Inelastic Near-Surface Interactions

Proceedings of
The Werner Brandt Workshop
on Penetration Phenomena

April 12–13, 1984

Pollard Auditorium/CER Building
Oak Ridge Associated Universities

Sponsored by
Oak Ridge National Laboratory
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Oak Ridge National Laboratory

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*Even though this paper was not presented at the workshop, it is related to the contribution of S. J. Pennycook and should be included.*
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Stanley Stern has performed magnificently the difficult and arduous task of editing the proceedings of all past workshops. Transcribing the presentations and ensuing discussions from magnetic tape to written word is an art form that requires innate talent, loving care, and knowledge of much physics. Since our summary report this time has, of necessity, been prepared without Stanley's expert ministrations, we do not attempt to record any discussions but only present material garnered from each participant. Thus we have material ranging from manuscripts that could be accepted as is by a refereed journal to skeletal remnants of vugraph transparencies. We believe, however, that the final result contains much to interest the radiation physicist and that the present report will, at the very least, guide the reader to relevant recent literature.
INTRODUCTION

R. H. Ritchie

A series of Informal Workshops on Penetration Phenomena of Charged Particles in Matter was begun at New York University in 1977 by Professor Werner Brandt. The 1982 workshop in this series was sponsored by the NSF and held in Honolulu. The locale was Oak Ridge, Tennessee the following year, and the meeting took place just after Werner's untimely death. A list of the topics covered in this series follows.

Informal Workshop Series

1977 - Wake Phenomena - NYU
1978 - Current Stopping Power Problems - NYU
1979 - Low-Energy Particles - NYU
1980 - Matter Under Extreme Conditions - NYU
1981 - Exotic Projectiles - NYU
1982 - Dynamic Screening and Effective Charge - Honolulu
1983 - Properties of Ion-Induced Tracks in Matter - ORNL

It was my privilege to work closely with Werner in the area of radiation physics for many years and to help him in organizing these workshops from the beginning. My colleagues and I at Oak Ridge National Laboratory propose to continue this series under the title "Werner Brandt Workshops" in the hope that the spirit of informality and excitement that was a hallmark of these meetings can be continued in the future.

These workshops have established a new mode of information exchange among workers in radiation physics, in part through the issuance of informal summary reports.

Werner devoted most of his lifetime in research to topics related to various aspects of penetration phenomena. He felt that this area contained a wealth of interesting, diverse, challenging, and important problems. We, his friends, think this is still true.

"For out of olde feldes, as men seith,
Cometh al this newe grain fro yeer to yeer;
And out of olde bokes, in good feith,
Cometh al this newe science that men lere."

Geoffrey Chaucer
Werner Brandt: In Memoriam

Werner was a delightful, gifted, many-faceted man. He was proud of his family and of his academic background. He spoke in glowing terms of the talents and achievements of all of his children.

He took deep enjoyment in physics, art, music, and friends. He was charming, witty, cultivated, and urbane. He liked living in New York and was very forceful in defending the city against uninformed critics.

Although I first met him in 1956, it was in 1968 that our collaboration really began. I had the great privilege of working on physics over a span of more than a dozen years with Werner in many interesting places around the world--New York, Munich, Moscow, Paris, Tokyo, Cambridge, Odense, Gothenberg, Aspen, and at almost all of the National Labs, including Oak Ridge. Werner's style in collaboration was very exciting and interesting. A typical session of 3-4 days in a given location would begin in an office with as much blackboard area as could be found. A list of topics would be put on the blackboard in short order with priority given to two or three items. A copy of Dwight's Table of Integrals was mandatory and, if possible, a copy of Abramowitz and Stegun.

This would be followed by much discussion, speculation, and qualitative reasoning. Werner operated on a profound intuitive level out of a broad understanding of fundamental physical principles. Typically, he would graph roughly the important functional relations in a given problem area before even beginning to calculate an effect quantitatively. Then the blackboard work would begin in earnest, often continuing into the morning hours. When ordinary mortals began to wilt, he would exhort us to plunge on with renewed energy. His unflagging vigor and optimism at the most difficult times were deeply inspiring.

After sufficient progress on a problem, the outlines of a paper would begin to appear. At that point Werner would become very elated. He loved producing a manuscript characterized by style, elegance, and precision. Once I was told by an Israeli physicist that a particular paper of Werner's was the best written of any that this individual had ever read. I looked up the paper and read it carefully. It was a model of economy, vigor, and clarity. I couldn't think of any way of changing a single sentence without doing damage. Werner was not only a stylist in producing papers, but excelled in crafting vividly descriptive phrases--among these are "vicinage effect," "peri-nuclear physics," "convoy electrons," "Pauli excitations," and many others that are now standard nomenclature.

At the end of a working session of 3-4 days, Werner would insist on a summing-up and a listing of desiderata for us to think about and work on toward our next meeting.
Werner had an uncanny ability to assemble large masses of data from many different sources, to understand the underlying principles, and to reduce the data by appropriate scaling procedures to what was often a single "universal" curve. I quote from one of his papers—"Similitude in Atomic Processes." "...It is the study of small deviations from the general behavior which brings into proper perspective the domains where a deeper analysis of the phenomena is necessary and where new scientific discoveries are possible." I think that these ideas motivated him through all his career and underlay his most important contributions to physics.

He pioneered in the study of collective effects in atoms, Bose-Einstein condensation of excitons, the systematic study of inner-shell excitations of atoms by swift charged particles, the $Z_1^3$ effect, the charged particle wake, energy losses by clusters of ions, and collective phenomena in radiation physics. We have already heard of his contributions in positron physics.

Werner was an outstanding leader in radiation physics, the area with which I am most familiar. He began a prestigious series of January Workshops here at New York University in 1977, establishing a new mode of information exchange among the most active workers in radiation physics.

Especially sympathetic to the plight of the physicist working diligently in isolation, Werner was responsible for guiding more than one such physicist into the mainstream of research.

Werner had an excellent sense of humor and was one of the most interesting people I have ever met. I learned many things from him—from how to choose binoculars to how to make a transatlantic flight and arrive at the destination as fresh as a daisy.

He served as a member of the Visiting Committee for our division at Oak Ridge National Laboratory for three years and was a loyal and articulate supporter of basic research and the researcher to Laboratory management.

Werner did many things for me and was a true and courageous friend. I will miss him always.

R. H. Ritchie

[Presented in a memorial service for Werner Brandt at New York University, February 28, 1983.]
Enhanced Electron Scattering in Small Spheres

David Penn

National Bureau of Standards
Washington, D.C.
INELASTIC ELECTRON SCAT
(AND PHOTON ABSORPTION) FROM
SMALL SPHERES (30-300 Å)

P. APPELL R. RENDELL

FOCUS ON MECHANISM

CLASSICAL EXPLANATION

RESPONSE DESCRIBED BY

\[ \epsilon(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i/\tau)} \]

LOSS \( \propto \frac{1}{\tau} = \frac{N_F}{l} + \frac{1}{T_{ee}} + \frac{N_F}{R} \)

BOUNDARY SCATTERING SUPPORTED
BY EXP + INDEPENDENT ELECTRON THEORY (30 Å)

RESPONSE OF SURFACE ELECTRONS
NOT BOUNDARY SCATTERING
Figure 7. $\varepsilon_2(\lambda = 405\text{ nm})$ of Ag particles, against $1/R$. Experimental: • from absorption halfwidth, ○ from Kramers–Kronig analysis. Curves FPE1 and FPE2: free path effect with $L_{\text{sphere}} = R$ and $L_{\text{sphere}} = \frac{3}{4}R$. Curve KK: Kawabata–Kubo effect.
SIMILARITIES BETWEEN IES AND PA

BOTH DUE TO EXCITATIONS, PLASMONS + E-H PAIRS

BOTH CHARACTERIZED BY SMALL MOMENTUM TRANSFERS

FORMALISMS VERY SIMILAR

PA SIMPLER, BETTER UNDERSTOOD

NON-LOCAL EFFECTS

SURFACE ELECTRON DISTRIBUTION
\[ A = \int d^3 r \, R \cdot \nabla \cdot (E(r) \times H(r)) / I_0 \]

Complex material \( g = G \)
Free electron material
Impurities \( \nu_L = \frac{N_F}{\xi} \)
Walls
\[ P_{EXC} \sim \nabla \cdot (\mathbf{E} \times \mathbf{H}), \text{ find } \mathbf{E}, \mathbf{H} \]

SPHERE

CLASSICAL THEORY (MIE)

ABSORPTION OR LOSS DUE TO

SURFACE PLASMONS

NON-LOCAL

E-H EXCITATIONS AT SURFACE

AS WELL AS SURFACE PLASMONS

SCIB "KLYWER"

GENERAL "FEIBELMAN"
\[ \bar{D} = \varepsilon \bar{E} \quad \text{means} \]

\[ \bar{D}(\vec{r}) = \int d^3r' \delta(\vec{r}, \vec{r}') \bar{E}(\vec{r}') \]

\[ \vec{r} \]

\[ \vec{r}' \]

FREE ELECTRON GAS

\[ \varepsilon \rightarrow \varepsilon(\vec{r} - \vec{r}', \omega) \]

\[ \varepsilon = \varepsilon_k + \varepsilon_t \]

\[ \bar{D}(\vec{q}) = \varepsilon(q, \omega) \bar{E}(\vec{q}) \]

CLASSICAL

\[ \varepsilon(\vec{r}, \vec{r}', \omega) = \delta(\vec{r} - \vec{r}') \varepsilon(\vec{r}, \omega) \]

\[ D(\vec{r}) = \varepsilon(\vec{r}, \omega) E(\vec{r}) \quad \varepsilon_v = 1 \quad \varepsilon_m(\omega) \]

VACUUM

METAL
CLASSICAL: PLANE

\[ \vec{A}_i \vec{A}_r \vec{A}_t \] PLANE WAVES
WAVE VECTORS
AMPLITUDES
(MAX EQS + BC)
General Soln (Kliener)

A NOT PLANE WAVE

\[ \nabla \times \nabla \times \mathbf{A} - (\frac{\mu}{\epsilon}) \epsilon \cdot \mathbf{A} = 0 \]

(GAUGE OF \( \phi = 1 \))

\[ \epsilon \cdot \mathbf{A}(\mathbf{r}) = \int \delta(\mathbf{r} - \mathbf{r}') \epsilon(\mathbf{r}') \cdot \mathbf{A}(\mathbf{r}') \, d^3 \mathbf{r}' \]

1) \( \mathcal{E} \) INCLUDES SURFACE
2) \( \mathcal{E} = \mathcal{E}_x \wedge \mathcal{E}_t \)
3) FREE ELECTRON
4) SCIB
$$\nabla \times \nabla \times \vec{A} - \left( \frac{\omega}{c} \right)^2 \vec{e} \cdot \vec{A} = 0$$

$$\text{SCEB} \} \in (\bar{\pi}, \bar{\pi}') = \varepsilon^{(0)}(\bar{\pi} - \bar{\pi}') + \varepsilon^{(0)}(\bar{p} - \bar{p}', \bar{s} + \bar{s})$$

$$\nabla \times \nabla \times \vec{A} - \left( \frac{\omega}{c} \right)^2 \vec{e}^{(0)} \cdot \vec{A} = \alpha \times \delta(\bar{s})$$
CROSS SECTIONS FROM Al(100) NORMAL EMISSION, P-POLARIZATION

FERMI LEVEL
- • CRYSTAL 1
- • CRYSTAL 2
- - THEORY

SURFACE STATE AT Γ
- • CRYSTAL 1
- • CRYSTAL 2

HEALTH (Arbitrary Units)

AREA (Arbitrary Units)

PHOTON ENERGY (eV)
SPHERE

\[ \nabla \times \nabla \times \vec{A} - \left( \frac{\omega}{c} \right)^2 \vec{E} \cdot \vec{A} = 0 \]

CLASSICAL E LOCAL

\[ \nabla \times \nabla \times \vec{A} - k^2 \vec{A} = 0 \quad ; \quad k^2 = \left( \frac{\omega}{c} \right)^2 \varepsilon \omega \]

\[ \nabla \cdot \vec{A} = 0 \]

USE SPHERICAL COORDS

MIE

\[ \vec{A}(\vec{r}) = \sum_{l=\{\ell m\}} a_l \vec{M}(k, \vec{n}) + b_l \vec{N}(k, \vec{n}) \]

(1) \nabla \cdot \vec{A} = 0

(2) \nabla \times \nabla \times \vec{A} - k^2 \vec{A} = 0

(3) \vec{A} is in SPHERICAL COORDS.
**PLANE WAVE ON SPHERE**

\[
\vec{A}_\omega = k e^{i k \vec{r}} = \sum_{L \geq 1} \left[ \vec{M}_{L \omega} - i \vec{N}_{L \omega} \right]
\]

\[
\vec{A}_e = \sum \left[ a_k^e \vec{M}_k - i b_k^e \vec{N}_k \right]
\]

\[
\vec{A}_m = \sum \left[ a_k^m \vec{M}_k - i b_k^m \vec{N}_k \right]
\]

**BC** \( E_{\|} , H_{\|} \) \( \text{cont} \)

\( l=1 \) **only for** \( k \omega R < 1 \)
\[ \hat{\mathbf{A}}_0 = \hat{x} A_0 e^{i k_0} \rightarrow \hat{x} A_0 \quad \text{const } \hat{z} \text{ field} \]

\[ k_0 R = 1 \]

\[ \phi_m = \frac{A_0}{2 + \varepsilon} \quad \varepsilon = 1 - \frac{\omega_p^2}{\omega^2} \]

\[ 2 + \varepsilon = 0 \quad \text{if } \omega = \omega_p / \sqrt{3} \]

**Peak Width Due to** \( \varepsilon \)
ADD e-h Pairs (E non-local)

\[ \nabla \times \nabla \times \vec{A} - \left( \frac{\omega}{c} \right)^2 \vec{E} \cdot \vec{A} = 0 \]

MODEL PROBLEM

\[ \nabla \times \nabla \times \vec{A} - \left( \frac{\omega}{c} \right)^2 \vec{E}_0 \cdot \vec{A} = \vec{J}_{\text{surf}} \]

(1) REPRODUCES QM CALCALS (K+F)
FOR PLANE SURFACE

(2) IF \( \varepsilon_1 \to \varepsilon_{\text{plasmon pole}} \),
REPRODUCES RUPPIN

(3) IF \( \varepsilon_1 \to 1 \), REPRODUCES MIE

NUMERICAL RESULTS FOR CASE

\[ \frac{\omega}{c} R \ll 1 \]
\[ \varepsilon_0 = \varepsilon_{\text{Lind-Mermin}} \]
\[ \nabla \times \nabla \times \vec{A} - \left( \frac{\omega}{c} \right)^2 \varepsilon_0 \cdot \vec{A} = J_{\text{SURF}} \]

\[ J_{\text{SURF}}(\vec{r}) = \delta(\rho - R) \left[ \hat{\phi} \psi_{(\phi, \psi)} + \hat{\theta} \phi_{(\theta, \phi)} \right] \]

\[ = \delta(\rho - R) \sum_l \left[ \alpha^l \vec{\rho} \times \nabla \gamma^l + \beta^l \vec{\rho} \times (\vec{\rho} \times \gamma^l) \gamma^l \right] \]

\[ \vec{P}(\vec{r}) = \int d\vec{r} \sum_l \left[ a^l(k) \vec{M}^l(k, \vec{r}) + b^l k \vec{N}^l(k, \vec{r}) \right. \]
\[ \left. + c^l \vec{L}^l(k, \vec{r}) \right] \]

**Solve for \( \vec{A} \) in terms of \( \alpha^l, \beta^l \) then use b.c.**
$R=25\,\AA$
$\lambda_s=4$

\[ \alpha \]

- $\omega_p \tau = 10^3$
- $\omega_p \tau = 10$
- $\alpha_{\text{Mie}}, \omega_p \tau = 10\pi \cdot 10^3$
- $\alpha_{\Pi}, \omega_p \tau = 10$

$\omega/\omega_p$
\[ A_{cl} = \frac{4\omega R}{c} \delta m \alpha \]

\[ \alpha = \frac{\varepsilon - 1}{\varepsilon + 2} \quad \varepsilon = 1 - \frac{\omega_p^2}{\omega(\omega + i/\gamma)} \]

\[ A_{cl} \approx A_{sc18} \quad \text{if} \quad \frac{1}{\gamma} \approx \frac{N_F}{R} \]

"SCREENING REDUCES BOUNDARY SCATTERING"

"HOWEVER \[ \frac{1}{\gamma} \approx \frac{N_F}{R} \] IS IN SEMI-
QUANTITATIVE AGREEMENT WITH EXP"

"RECALL IMPORTANCE OF SURFACE ELECTRON DENSITY FOR PLANE"
**Theory for Sphere That Describes Surface Excitations - Non-Local - Real Surface Profile**

OK for IES or PA

\[ \frac{\lambda}{R} \rightarrow \text{const.} \text{ field} \]

\[ \nabla^2 \phi = 0 \]

\[ r_2 - r_1 \approx 10 - 20 \]°

\[ r_1, R, r_2 \]

\[ \begin{align*}
    r < r_1 & \quad \frac{\phi^<}{r} = -E^< r \cos \theta \\
    E_r^< &= E^< \cos \theta \\
    E_\phi^< &= E^< \frac{\partial}{\partial \phi} \cos \theta \\
    D &= \varepsilon E^< \\
    \hline
    r > r_2 & \quad \frac{\phi^>}{r^2} = -E^> (r - \alpha (r^3)) \cos \theta \\
    E_r^> &= E^> \left( 1 + 2 \alpha \frac{r^2}{r^3} \right) \cos \theta \\
    E_\phi^> &= E^> \left( 1 - \alpha \frac{r^2}{r^3} \right) \frac{\partial}{\partial \phi} \cos \theta \\
    D &= \varepsilon E^>
\end{align*} \]

**Classical Desc. if \( r_1 = r_2 = R \)**

\[ \rightarrow E_r \text{ discontinuous} \]

**Unknowns** \[ E^<, \alpha \neq E^<_{cl}, \alpha_{cl} \]
\( \forall r < R \)
\[ \phi^\prime = -E^\prime \cos \theta \]

\( \forall r > R \)
\[ \phi^\prime = -E^\prime (r - \alpha \frac{R^3}{r^2}) \cos \theta \]

\[ E = -\nabla \phi \]
\[ D = \varepsilon E \]

Assume
\[ E_\phi = E_\phi \]
\[ D_\phi = D_\phi \]
\[ D_R = D_R \]
\[ E_R \text{ unknown} \]

\[ \nabla \times E = 0 \Rightarrow \phi \frac{\partial}{\partial R} \left[ -\frac{2}{R} (RE_\phi) + \frac{2}{\phi} E_R \right] = 0 \]

Define
\[ d_R = \int_{r_1}^{R} \left[ E_R - E_R \right] \frac{dR}{\varepsilon (R^2)} \]

\[ E^\prime (1 + d_R (\varepsilon - 1)) = E^\prime (1 - \alpha^2) \]

\[ \nabla \cdot D = 0 \Rightarrow D_\phi \text{ constant} \]

\[ E^\prime (1 + 2\alpha^2) = \varepsilon E^\prime \]

In case of plane this method gives "Feibelman"
WE REQUIRE \( d_r \)

\[
d_r(w) = R \frac{\int d^3 r r (r - n) \delta p}{\int d^3 r r^2 \delta p} = \frac{\int d^3 z \delta p}{\int d^3 x \delta p} \uparrow \delta p (w)
\]

\[
d_\perp(w) = \frac{\varepsilon}{1 - \varepsilon} \int d^3 z \frac{d}{d^3 z} \int d^3 z' \varepsilon^{-1} (z, z'; w)
\]

EVALUATED BY FEIBELMAN FOR LIMITED RANGE OF \( w \)

EVALUATED BY SUM RULES

\[
d_\perp(w) = A (1 + a w^2 + b w^4) w \Theta (w_0 - w)
\]

\( A, a, b \) DEPEND ON ELECTRON DENSITY, DETERMINED BY LOCAL DENSITY CALS
Feibelman

PRESENT APPROXIMATION

$-\text{Im} k_F d_1$

$\omega$

$\Omega \rightarrow$
\[ A = 4 \frac{\omega}{c} R \Im \alpha \]

\[ \alpha = \frac{\tilde{\varepsilon} - 1}{\tilde{\varepsilon} + 2} \quad \tilde{\varepsilon} = \frac{\varepsilon}{1 + (\varepsilon - 1)/(d_p^2/R)} \]

\[ A_{cl} = 4 \frac{\omega}{c} R \Im \alpha_{cl} \]

\[ \alpha_{cl} = \frac{\varepsilon - 1}{\varepsilon + 2} \quad \varepsilon = 1 - \frac{\omega_p^2}{\omega(\omega + i\tau)} \]

**WHAT IS** \( \frac{1}{\tau} \) **IF** \( A_{cl} = A \)?

\[ \frac{1}{\tau} = \frac{N_F}{R} \hat{f}(\omega, R, R_A) \]

\[ = \frac{N_F}{R} \left( 1 - \left( \frac{\omega}{\omega_p} \right)^2 \right) \frac{\omega_p^2}{\omega} \frac{1}{N_F} \Im (-d_p) \]

**IS** \( \hat{f} \approx 1 \)? **IF SO WOULD APPEAR**

**WHY** \( \frac{1}{\tau} = \frac{N_F}{R} \) **IS IN SEMI-QUANT**

**AGREEMENT WITH EXP**
PA

\[ A_I(\vec{r}, t) = e^{i(\vec{k}_0 \cdot \vec{r} - \omega_0 t)} \]

\[ A_I(\vec{q}, \omega) = 8 (\vec{q} - \vec{k}_0) \delta(\omega - \omega_0) \]

IES

\[ \vec{r}_0 = (\vec{p}_0, \nu t) \]

\[ \rho(\vec{r}, t) = 8(\vec{r} - \vec{r}_0) = 8(\vec{p} - \vec{p}_0) \delta(\omega - \omega_0) \]

\[ \rho(\vec{q}, \omega) = \int d^3q_{\parallel} e^{i\vec{q}_{\parallel} \cdot (\vec{p} - \vec{p}_0)} \delta(\omega - \omega_{\parallel}) \]
FORMULATION OF F. + K.

\[ \vec{r}_o = (\vec{r}_0, N \tau) \]

\[ W(\vec{r}_o) = \int dV \bar{F}_z = e \int dt N \Delta E_z(\vec{r}_o, t) \]

INDUCED FIELD

\[ W = \int d^2 \vec{r}_o \ Re \{ W(\vec{r}_o) \} = \int d^2 \vec{r}_o \int d\omega k \omega P(\vec{r}_o, \omega) \]

\[ \therefore \text{ FIND INDUCED FIELD} \]
\[
\phi(r, t) = \phi(r, \omega) \\
\]

\[
\begin{array}{c}
\text{If } r < r_1, \\
\phi^*(r, \omega) = \phi_0^*(r, \omega) + \sum_{\ell m} \beta_{\ell m} \left( \frac{r}{R} \right)^{\ell} Y_{\ell m} \\
\text{If } r > r_2, \\
\phi^*(r, \omega) = \phi_0^*(r, \omega) + \sum_{\ell m} \gamma_{\ell m} \left( \frac{r}{R} \right)^{\ell+1} Y_{\ell m}
\end{array}
\]

Assume \( E_0, D_1, D_2 \) given by above (with \( r_1 = r_2 = R, D = EE \))

\( E_r \) unknown
USE $\nabla \times E = 0$, $\nabla \cdot D = 0$

TO OBTAIN $B_{lm}$, $\gamma_{lm}$

IN TERMS OF

$\left(\begin{array}{c}
\frac{r_2^2}{r_1^2} \\
\frac{\delta P^{(lm)}}{r_1^2}
\end{array}\right) = R \int d\mathbf{r} \mathbf{r} (\mathbf{R} - \mathbf{r}) \delta P (\mathbf{r}, \omega)$

$\int d\mathbf{r} r^2 \delta P (\mathbf{r}, \omega)$

INDUCED SURFACE CHARGE DENSITY

$\delta P (\mathbf{r}, \omega) = \sum_{lm} \delta P^{(lm)} (\mathbf{r}, \omega) \gamma_{lm} (\hat{n})$
\[ P(q_{\parallel}, \omega) = \frac{4}{\pi \hbar v^2} \left[ \frac{1}{3} \frac{R^3}{Q^2} \text{Im} \left( \frac{1}{\varepsilon(Q, \omega)} - 3 \sum_l (2l + 1) (j_l(QR))^2 \frac{d^{(l)}_n}{R} \left( \frac{1}{\varepsilon(Q, \omega)} - 1 \right) \right. \right. \]
\[ \left. \left. - \frac{R}{Q^2} \sum_l l(l + 1)(2l + 1) \right) \right] \times \text{Im} \left[ \left[ \left( 1 - \frac{d^{(l)}_n}{\partial r} j_l(QR) \right)^2 \left( \frac{1}{\varepsilon(Q, \omega)} \right) \left( 1 - \varepsilon \right) \right] \right] . \]

\[ Q^2 = \frac{q^2_{\parallel}}{b_{ll}^2} + \left( \frac{\omega}{\omega_c} \right)^2 \]

\[ \Delta = \epsilon l + (l+1) \left( 1 + \epsilon (\epsilon - 1) \frac{d^{(l)}_n}{R} \right) \]

\[ l = 1, \quad d^{(l)}_n = 0 \quad \rightarrow \quad \Delta = \epsilon + 2 \]

AGREES WITH CLASSICAL LIMIT
OF F. + K. IF \( d^{(l)}_n = 0 \)

SMALL \( Q \) DOMINATE \( ; \) SMALL \( l \)
\[ d_{\mathcal{N}}(\omega) = d_{\mathcal{N}} \left( \frac{q}{k_{FR}}, \omega \right) \approx d_{\mathcal{N}}(\omega) \]

\[ P(\mathbf{q}, \omega) = P_{S}(\mathbf{q}, \omega) + P_{B}(\mathbf{q}, \omega) \]

\[ QR \ll 1 \]

\[ P_{S} \sim \frac{1}{Q^{2}} \quad P_{B} \sim 1 \]

\[ P_{S}(\mathbf{q}, \omega) = \frac{2e^{2}}{\pi \hbar v_{F}} \frac{R^{3}}{Q^{2}} \text{Im} \alpha(\omega) \]

\[ \alpha(\omega) = \frac{\bar{\epsilon} - 1}{\bar{\epsilon} + 2} \quad \bar{\epsilon} = \frac{\epsilon}{1 + (\epsilon - 1) \frac{d_{\mathcal{N}}}{R}} \]
$\omega_p \tau = 10^3$

- Non-Local
- Classical

$P_s/N$ vs $\omega/\omega_p$ for different $\rho$ values.
(1) Classical Calc of IES + PA allows for surface plasmon excitation.

(2) Non-local Calc include surface e-h pairs.

(3) If \( \frac{1}{\lambda} = \frac{N_F}{l} \), then surface e-h pairs give large enhancement in IES + PA

(i) \( \text{SCIB} \sim 10 \)

(ii) "Real" \( \sim 10^2 \)

(4) Classical Theory + \( \frac{1}{2} = \frac{N_F + N_F}{R} \)

"Agrees" with EXP, indep. elect. th.

Interacting electrons give

(i) \( \frac{1}{3} \sim 0.1 \frac{N_F}{R} \text{ SCIB} \) (Boundary scat mechanism wrong)

(ii) \( \frac{1}{3} = \frac{N_F}{R} b(w, R) ; \quad b > 1 \)
The Energy Loss by Swift Electrons to Metallic Layers

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I want to demonstrate that the problem of stopping power for charged particles, which are near to conducting surfaces, is in an exact way related to the problem of metal optics. I then give a simple exact relation between stopping power and reflection coefficient. The calculation of the reflection coefficient is discussed in the hydrodynamic approximation, which is capable of including in a straightforward manner the excitation of charge density perturbations near the surface, damping of plasma waves, excitation of band transitions via a measured dielectric function $\epsilon(\omega)$, retardation of the field propagation. The electromagnetic fields excited by an incoming wave near the surface derived from the hydrodynamic approximation are compared to those derived by the most sophisticated electron gas calculation. The fields agree in all characteristic properties even when the surface model is kept extremely simple. Therefore, there is no doubt that also the stopping power will be given correctly by this approach.

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\[ F_x = -e_0 E_x \]

\[ \Delta W = F_x \Delta x \quad \text{change of energy of particle} \]

\[ \frac{dW}{dx} = -e_0 E_x < 0 \quad \text{energy loss per unit path length} \]

= stopping force

\[ \Rightarrow \text{calculate induced electric field} \]
First: field of the moving electron: \( q S(x,y,z,t) \)

\[
\vec{E}(x,y,z,t) = \iiint E_0(\vec{k},\omega) e^{-i(\vec{k}\cdot\vec{r} - \omega t)} \, d^3k \, d\omega
\]

\[
\vec{E}_0(\vec{k},\omega) = -i \frac{4\pi q}{(2\pi)^3} \frac{\omega\vec{\nu} - c^2 \vec{k}}{\omega^2 - \vec{k}^2 c^2} e^{-i k_z z_0} \delta(\omega - k_x v_x)
\]

\[
\int d^3k \quad \vec{E}_0(\vec{k},\vec{y},\omega,\vec{z}) = -\frac{q}{\pi} \frac{1}{2\lambda^2 c^2} \left[ \frac{\nu_0 \vec{v} - (k_x \vec{k}_y + \lambda \vec{\nu}_0 \vec{z}_0)}{c^2} \right] e^{\frac{\nu_0 \nu_0}{c^2} - k_x^2 - k_y^2}
\]

\[
\lambda = \sqrt{\frac{\omega_0^2}{c^2} - k_x^2 - k_y^2}
\]

The moving charge is a source of a electromagnetic waves (linearly polarized)

\( \nu \to 0 : \omega \to 0 \)

\[
\vec{E}(x,y,z) = \iiint -\frac{q}{2\pi} \frac{i\vec{k}}{k^2} e^{-i \frac{k(x-v_0 t)}{c^2}} \, d^3k
\]

\[
= -\nabla \iiint \frac{q}{2\pi k^2} e^{-i \frac{k(x-v_0 t)}{c^2}} \, d^3k = -\nabla \frac{q}{|x-v_0 \vec{z}|}
\]
Induced field = sum of all reflected waves

calculation of reflected amplitudes:
Decomposition into s- and p-polarisation:

\[ \mathbf{E} = \frac{E_y k_x - E_x k_y}{k_x^2 + k_y^2} (k_y, -k_x, 0) + \frac{E_x k_x + E_y k_y}{k_x^2 + k_y^2} (k_x, k_y, 0) + (0, 0, E_z) \]

reflected amplitudes:

\[ A_s(k_x, k_y, \omega) = \mathcal{T}_s(k_x, k_y, \omega) E_s \]
\[ A_p(k_x, k_y, \omega) = \mathcal{T}_p(k_x, k_y, \omega) E_z \]

\[ A_x(k_x, k_y) = A_s + A_p = -\frac{k_x}{k_x^2 + k_y^2} \left( \mathcal{T}_p + \mathcal{T}_s \right) E_z + \mathcal{T}_s E_x \]

Induced field:
\[ A_x(x, y, z) = \iiint dk_x dk_y d\omega A_x(k_x, k_y, \omega) e^{i(k_x x + k_y y + k_z z - \omega t)} \]
Metal optics

Hydrodynamic approximation includes:
- excitation of charge density fluctuations
- damping of plasmon waves
- excitations of band transitions via measured $E(\omega)$
- retardation of field propagation

$$E_\| (\omega, K) = E_b (\omega) - \frac{\omega^2}{\omega^2 - \beta^2 k^2}$$

Boundary conditions: $E_{x_2}, E_n$ continuous, $j_n = 0$
Calculation of the reflection coefficient $r_p$

Solution above surface:
$$\vec{E}(\vec{r}, z) = e^{i(k_x x - \omega t)} (\vec{E}_e \ e^{i\lambda z} + \vec{E}_T \ e^{-i\lambda z})$$
$$\lambda = \left( \frac{\omega^2}{c^2} - k_x^2 \right)^{\frac{1}{2}}$$
$$\vec{E}_e = E_e \left( -\frac{1}{\lambda}, 0, 1 \right)$$
$$\vec{E}_T = E_T \left( \frac{1}{\lambda}, 0, 1 \right)$$

Solution below surface:
$$\vec{E}(\vec{r}, z) = e^{i(k_x x - \omega t)} (\vec{E}_\perp \ e^{i\alpha z} + \vec{E}_\parallel \ e^{-i\beta z})$$
$$\alpha = \left( \frac{\omega^2}{c^2} - k_x^2 \right)^{\frac{1}{2}}$$
$$\beta = \left( \frac{\omega^2}{c^2} - k_x^2 \right)^{\frac{1}{2}}$$
$$\vec{E}_\perp = E_\perp \left( -\frac{1}{\alpha}, 0, 1 \right)$$
$$\vec{E}_\parallel = E_\parallel \left( k_x / \sigma, 0, 1 \right)$$

Boundary conditions:
$$E_x \ \text{continuous}: \quad \lambda E_T + \alpha E_\perp - k_x / \sigma E_\parallel = \lambda E_e$$
$$\epsilon E_x \ \text{continuous}: \quad -E_T + \epsilon_e E_\perp + \epsilon_b E_\parallel = E_e$$
$$J_n \ \text{continuous,} \ j_n = 0: \quad \sigma_\perp E_\perp + \sigma_\parallel E_\parallel = 0$$
$$E_\parallel = E_b - \frac{e_{12}^2}{\omega (\omega + i\gamma) - \beta^2(k_x^2 + y^2)}$$
$$\sigma = \frac{i\omega}{4\pi} (\epsilon - 1)$$
$$E_T = \sqrt{\rho} (k_x, \omega) E_e = \frac{\epsilon_b \lambda - \alpha \epsilon_e + k_x / \sigma \frac{e_b - e_e}{e_b}}{\epsilon_b \lambda + \alpha \epsilon_e - k_x / \sigma \frac{e_b - e_e}{e_b}} E_e$$
\[ \tau_s = \frac{\lambda - 2\varepsilon}{\lambda + 2\varepsilon} \]

\[ \tau_p = \frac{\lambda \varepsilon - 2\varepsilon + \frac{k_x^2 + k_y^2}{\varepsilon} - \frac{\varepsilon - c_b}{\varepsilon_b}}{\lambda \varepsilon + 2\varepsilon - \frac{k_x^2 + k_y^2}{\varepsilon} - \frac{\varepsilon - c_b}{\varepsilon_b}} \]

\[ \varepsilon = \varepsilon_b \cdot \frac{\omega^2}{c \cdot (\omega^2 + c^2)} \]

\[ \lambda = \left( \frac{\omega^2}{c} - k_x^2 - k_y^2 \right)^{\frac{1}{2}} \]

\[ \varepsilon = \left( \frac{\omega^2}{c} - k_x^2 - k_y^2 \right)^{\frac{1}{2}} \]

\[ \varepsilon = \left( \frac{\omega^2 - c^2}{\varepsilon_b} - k_x^2 - k_y^2 \right)^{\frac{1}{2}} \]
The capability of the hydrodynamic approximation can be seen from a comparison of the fields near the surface with those derived from microscopic calculations.

From H. Kumpa, F. Forstmann, Surface Sci. 159, 536 (83)

Fig. 2 Variation of the electric field $E$, normal to the surface. The fields are normalized by the modulus of the incoming wave. (— — — ) From Kiewer [28], (— — ) from the hydrodynamic approximation, (— — — ) from standard optics $\gamma/\omega_p = 10^{-5}$; top, $\omega = 0.8\omega_p$; bottom, $\omega = 1.2\omega_p$.

Fig. 3. Normalized electric field near the surface. $\xi(z) = \frac{|E_z(z) - E_z(\infty)|}{|E_z(\infty)|}$:
(— — — ) $\Re\xi(z)$; (— — ) $\Im\xi(z)$; left side, according to Feibelman [2]; right side: hydrodynamic approximation with surface step and homogeneous damping.

\begin{align*}
\eta(z) &\quad \omega/\omega_p = 1.18 \\
\eta(z) &\quad \omega/\omega_p = 0.95 \\
\eta(z) &\quad \omega/\omega_p = 0.63
\end{align*}
We are studying the questions:

How does \( \frac{dW}{dx} \), \( \frac{d^2W}{dx \, dw} \) or \( \frac{d^3W}{dx \, dK \, dw} \) depend on

- Distance from surface
- Velocity of particle
- Dielectric function of metal (silver)
- Thickness of metal layer

Compare different approximations for \( T_s \) and \( T_p \)

Influence of retardation expected for electrons above 100 eV

Also calculate \( A_z = \text{Image force} \)

Velocity dependence
Notes on the Inelastic Scattering of Fast Electrons in the Aloof Geometry

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These experiments were done in a commercially available Scanning Transmission Electron Microscope (STEM) equipped with a Field Emission gun. Typical operating parameters are given in the figure. Briefly, a small, usually diffraction limited, spot is formed at the specimen plane by the use of two lenses. An aperture in the back focal plane of the final lens defines the range of angles which makes up the coherent spherical wavepacket. A smaller range of angles are collected in the far field beyond the specimen and are energy analyses, usually by a magnetic sector spectrometer as shown. Therefore, the scattering geometry does not consist of simple plane wave transitions, but is better described as a spherical wave going to a plane wave.
When using a large probe with a small angular spread, surface losses are enhanced relative to bulk losses. This is illustrated by the work of Petersen (1977) who found strong $l=1$ resonant surface plasmon, but not much else. There are probably two major reasons for this: 1) As illustrated, quite a lot of the particle surfaces are excited by grazing incidence electrons. Since the surface plasmon probability has a singularity for this condition, quite a lot of scattering is expected. 2) The surface scattering falls off as the cube of the scattering angle, while the bulk scattering decays as the square of the scattering angle. Therefore, a small scattering angles the surface losses are enhanced. In the STEM, we may strike the particle with a probe which is nominally much smaller than the particle diameter. Therefore we may eliminate the grazing incidence geometry. Next, as was pointed out above, this scattering involves transitions from an electron state which has a broad angular composition, thus favoring the broader distribution characterizing the bulk losses.

**Previous Results in Small Al Particles**

![Graph](image1)

H. Petersen (1977)

Nominal 20 Å Cride-F
Aluminum

**Present Results**

![Graph](image2)

Single 50 Å Al core
with 40% layer of Al$_2$O$_3$

6. Of Interest to Al, Cu, and Fe Base Cores
In fact, strong bulk properties are found in structures whose size is less than 5nm. The illustration shows an example of an aluminum sphere with 4nm of oxide on its surface, sitting on a thin carbon film. The energy loss plots from various positions within the sphere clearly reflect the compositional variations that are present.

**Experiment:**

Al spheres: evaporation in 4 Torr Ar
picked up by LN2 cooled Carbon Substrah
If the STEM energy loss spectrometer is set to one particular energy loss value, while the probe is scanned across the specimen, we may generate an "energy filtered image" which represents a probability map for the inelastic scattering which is selected. In this illustration, we show this map for a cluster of small Al spheres with the elastically scattered electrons, with those that have lost energy to the surface plasmon (6.7eV), the bulk plasmon (15eV), the aluminum oxide bulk plasmon (23eV) and to an unusual mode at 3.4eV which will be shown later to consist of a coupling of surface plasmons on adjacent spheres. Notice that the resolution of this technique is limited by the probe size only. The interaction of the particle system with the passing electrons is long ranged, however, strong variations in the strength of this interaction occur for very small changes in the probe position. Hence, quite sharp features occur (for instance the sharp lines at the particle interfaces in the 6.7eV image).
To start comparing these kinds of images with the calculations, we should start with a simpler geometry. In this illustration, we have the experiment for the aloof interaction of a fast electron traveling parallel to an infinite metal plane. The "plane" in this case is a rolled edge of an Al film (with a radius of about 100 microns). The Al film, of course, has an oxide coating. The scattering results as a function of distance from the oxide-vacuum surface are shown. The bulk scattering is absent. The surface loss is prominent and can be seen more than 30nm from the surface. There is a broad loss near 15eV which isn't obvious in other scattering geometries, although it should be noted that the bulk plasmon would be expected to obscure this intensity if the bulk Al is linked.
We model this experiment with a simple electrostatic boundary value problem. We solve Poisson's Equation with a source term determined by the fast electron. This is done after fourier analyzing in the directions parallel to the plane surface to simplify the problem. The electrostatic potential applied by the passing electron decays exponentially with a characteristic length controlled by the wavelength of the surface plasmon which is generated.

Model A Three Layer System

1. External Applied Potential
   \[ \phi_{\text{ext}} = -\frac{q\kappa e}{\kappa} \frac{e^{-\lambda(y-b)}}{\lambda^2} s(y-b) \]

2. System Response Potential
   \[ \phi_2 = \frac{\beta}{\varepsilon_2} e^{\lambda y} + A e^{\lambda y} \]
   \[ \phi_1 = \frac{\beta}{\varepsilon_1} e^{\lambda y} + B e^{-\lambda y} + C e^{\lambda y} \]
   \[ \phi_0 = \frac{\beta}{\varepsilon_0} e^{\lambda y} + D e^{-\lambda y} \]
   \[ \beta = -\frac{q\kappa e}{\kappa} e^{-\lambda b} \]
We compute the energy loss probability and find that the probability decays exponentially away from the oxide surface, and that it has resonances which depend on the detailed behavior of the dielectric responses of the Al the oxide and the vacuum. Also, the resonance value is shifted by variations in the oxide thickness, and indirectly through the exponential decay, by the impact parameter. Finally, we integrate over scattering angle to get the total scattering from the spherical wavepacket to the final plane wave. A simple integration over all possible surface plasmon scattering wavevectors (\( \lambda \)) gives \( K(\lambda) \). The integration for this work was done numerically. We finally obtain the scattering probability as a function of impact parameter an energy loss.

Compute the energy loss probability

\[
P(\omega, b) = -\frac{e}{\hbar \omega} \int \frac{3}{2} \left( \phi_0(\omega, x, b) - \phi_{\text{ext}}(\omega, x, b) \right)_{x=vt} \, dx
\]


\[
I_{\text{K}} \left( \frac{1}{\epsilon + i} \right)
\]

Obtain:

\[
P(\omega, b, \lambda) = -\frac{4\pi e^2}{\hbar \nu} \frac{\lambda(b-a)}{\lambda} \frac{\epsilon = \epsilon_0}{\lambda} \frac{\text{Im} \left( \frac{\lambda_2 + \epsilon_0 e^{2\lambda a}}{\lambda_2 \lambda_0 + e^{2\lambda a}} \right)}{\lambda_2 \lambda_0 + e^{2\lambda a}}
\]

\[
\lambda_{ij} = \frac{\epsilon_i - \epsilon_j}{\epsilon_i + \epsilon_j}
\]

\[
\epsilon_2 \rightarrow \text{Al} \quad \epsilon_1 \rightarrow \text{Al}_2\text{O}_3 \quad \epsilon_0 \rightarrow \text{Vacuum}
\]

finally:

Average over scattering

\[
P(\omega, b) = \int \int \int \frac{2\pi}{q_{\text{max}}} q^* \int_0^{2\pi} d\theta \, P(\omega, \lambda, b) \, Reff
\]

\[
\lambda^2 = g^2 \cos^2 \theta + \frac{\omega^2}{v^2}
\]

\( q \): Transverse wavevector

\( \theta \): angle between \( q \) & metal surface

\( v \): parallel wavevector
We use the Lindhard dielectric function for Al and tabulated values for the oxide as shown below. Notice the broad region between 12 to 23 eV where the oxide dielectric constant is close to zero. This produces unusual intensity in the experimental result.

**Dielectric Constants**

**Al:**
Lindhard form $\epsilon(q,\omega)$
to include damping $\omega_{0}$

**$\text{Al}_2\text{O}_3$:** Measured for amorphous $\text{Al}_2\text{O}_3$

![Graph showing the real part of the dielectric function for $\text{Al}_2\text{O}_3$]
The result for the energy loss intensity at an impact parameter of \(1\text{nm}\) is shown below. The energies for the completely relaxed (infinite oxide) and unrelaxed (clean metal) surface plasmon energies are indicated by vertical lines. The phase and magnitude of the surface plasmon electric potentials are shown also. These help us to visualize the spatial symmetry of the indicated resonances. The model successfully describes the qualitative behavior of the incompletely relaxed surface plasmon. In addition, it predicts the increased scattering intensity above \(12\text{eV}\). This is seen to be a result of the small value of the oxide dielectric constant in this range. If the oxide dielectric constant is set equal to a constant 3.65, this intensity disappears, leaving the one strong surface resonance.
We now plot the scattering probability as a function of impact parameter for two different energies -- the surface plasmon and the diffuse intensity at 18eV. The intensity at the oxide surface is normalized to the observed value. The variation with impact parameter is in quite good agreement. The scattering would thus seem to follow the simple classical electrostatic ideas very well.
A comparison of the results from the parallel and perpendicular incidence experiments show a shift in energy of the observed surface resonance. Referring to the probability of scattering shown above, notice that larger impact parameters tend to favor longer wavelength (smaller lambda) surface plasmon generation. These longer wavelength plasmons penetrate the oxide to a greater extent and are thus less completely relaxed. Therefore, the observed shift to higher energies in the grazing incidence experiment can be explained.

[Graph showing energy loss vs. energy with annotations for b=1nm and Bulk]
When the small spheres are not well separated, there sometimes appears a strong resonance at roughly half the energy of the normal surface plasmon. This illustration shows a comparison of the energy loss results for two essentially identical spheres -- one quite alone and one situated near another, larger sphere. The new resonance is striking.

**Energy Loss Result for Two 36 nm Spheres.**

- Next to Large Sphere
- Isolated on Carbon Film.

---

**Energy Resolution 1eV**

Collection half angle of Specimen $\sim 3$ mR (250 μm)

100 KeV

Probe $\sim 1$ mm

Obj. Aperture 50μm
Taking the lead from the calculation above, we model the electrostatic problem in bi-spherical co-ordinates appropriate to a two sphere system. These are summarized below. The important point to remember here is that these are only partially separable, so that no formal eigen solutions are available. However, when the two spheres are close together as in this case, Poisson's equation is separable to a good approximation.

\[ \Psi = \sqrt{\cos \theta - \cos \phi} \sum \left( A_n e^{-(n+\frac{1}{2})\mu} + B_n e^{(n+\frac{1}{2})\mu} \right) P_n(\cos \phi) \]

\[ \frac{\partial^2 \Psi}{\partial \phi^2} + \frac{1}{\sin \phi} \frac{\partial \Psi}{\partial \phi} + \frac{n^2 - 1}{\sin^2 \phi} \Psi = 0 \]

We apply boundary conditions: 
\( \Psi \) finite, \( \Psi \) continuous, \( \frac{\partial \Psi}{\partial \mu} \) continuous

And we obtain a coupled set of equations for \( B_n \) in the oxide.

\[ n \frac{d^2 B}{d \mu^2} + \left( E_n + \frac{n}{n+1} \right) B_n + E_n B_{n+1} = 0 \]

\[ G_n = \frac{(2n+1)\mu - 2 \mu^2 e^{2n\mu}}{(2n+1)\mu - 2 \mu^2 e^{2(n+1)\mu}} \]

\[ \gamma^2 = \frac{E_0 - E_n}{E_n + E_i} \]

\( E_0 \) of the oxid. 
The determinant of this matrix of coefficients define the frequencies \( \omega \) that produce non-trivial solutions for the \( B_n \).
We then can solve for the secular equation that gives us the resonant mode frequencies as a function of the relative sizes and separation of the two spheres. This form is close to that found for a thin metal film, when the surface plasmons on the top and bottom surfaces couple. The spherically symmetric problem for an oxide coated Al sphere embedded in an infinite Al matrix gives results almost identical to the bi-spherical problem. This is because the dominant process here is the interaction of the surface plasmon fields across the interface at the point of touching of the two spheres.

The secular equation appropriate to each mode, \( m \), can be found by diagonalizing the determinant, setting it equal to zero.

\[
2 \varepsilon_{m}^{2} = E
\]

For Al spheres embedded in Al\(_{2}\)O\(_{3}\) this result becomes

\[
E_{m}(\omega_{\pm}) = -\varepsilon_{0}(\omega_{\pm}) \left[ \tanh \left\{ \frac{1}{2}(\kappa+\frac{1}{2})(\mu_{1}-\mu_{2}) \right\} \right]
\]

With \( \varepsilon_{0}(\omega) \) being the frequency dependent dielectric constant for Al\(_{2}\)O\(_{3}\).

The solutions for \( \Psi \) take the form shown in the next slide.

Notice:

This form is identical to form for thin film of Al where surface plasmons on the top & bottom surfaces couple. Here

\((\mu_{1}-\mu_{2})\) takes the place of thickness \( t \).

(Good for \( \mu_{1}, \mu_{2} \) small \( \Rightarrow \) spheres close together)
Below is the result for the calculated energies compared with the measured values for several sets of particles of varying sizes. Notice that the dominant excitation seems to be the one with the highest symmetry. The $n=0$ mode has high dipole symmetry. On the other hand, the normal surface losses appear to favor the high angular momentum modes. This appears to be an impact parameter effect and will be discussed below.

Electrostatic Calculation for Energy of Spherical and Bispherical Surface Modes in all Spheres

![Graph showing energy vs. angular momentum for spherical and bispherical symmetries](image)
The approximate solutions for the electric potential are given below. The antisymmetric solution (also the low energy solution) is interesting because of its large dipolar character. We can compute the response electric field for these solutions, and find that in the region between the two spheres the field is perpendicular to the fast electron trajectory. Thus no work can be done and no energy loss occurs. This idea can be tested.

The solutions for $\psi^\pm$ become

\[
\psi^+ = F \sum (1 \pm 1) e^{-\sqrt{(n+\frac{1}{2})(\mu+\frac{1}{2})} \frac{1}{2}} \frac{\sin \left( (n+\frac{1}{2})(\mu+\frac{1}{2}) \right)}{\sin \left( (n+\frac{1}{2})(\mu+\frac{1}{2}) \right)} P_n(\cos \theta)
\]

\[
\psi^- = F \sum e^{\sqrt{(n+\frac{1}{2})(\mu-\frac{1}{2})} \frac{1}{2}} \frac{\sin \left( (n+\frac{1}{2})(\mu-\frac{1}{2}) \right)}{\sin \left( (n+\frac{1}{2})(\mu-\frac{1}{2}) \right)} P_n(\cos \theta)
\]

\[
F = \sqrt{\cos \mu - \cos \theta}
\]

* Note $\psi^- = 0$ for $\mu = \frac{\mu_1 + \mu_2}{2}$

* Note also the strong dipole character (1) for $n=0$ mode.

* $-\nabla \psi$ defines electric fields which are perpendicular to $\nabla \mu$ when $\mu = \frac{\mu_1 + \mu_2}{2}$ (Blue dots).

* The fast electron should not be able to couple to the system under this condition.
Below we show the energy loss scattering probability for three incident probe positions. At the position B, the response field would be perpendicular to the fast electron trajectory and so the bi-spherical mode is not present. At the position C, the bispherical mode is present. At positions A and B, coupling to the normal spherically symmetric mode is seen. This must arise from the excitation of very short wavelength surface plasmons on the surfaces of both spheres. These would not couple strongly because of their short decay lengths outside the surfaces of the spheres. Therefore the observed energy would correspond to that of the ordinary surface plasmon.
The large angular momentum modes are favored on isolated spheres probably for the following reason. Since the probability is likely a strong function of the impact parameter (e.g. exponential for the flat surface), small impact parameters are favored. For small impact parameters, the sphere is polarized by a very highly non-uniform electric field produced by the probe. Therefore we expect to see a surface plasmon comprised of many short wavelength modes. When the electron passes far away, the applied field is relatively uniform, leading to a long wavelength small angular momentum mode. This was also suggested above as an explanation for the observed shift in the plasmon energy. (It is interesting that there this idea resulted in an energy shift upwards due to field penetration of the oxide, rather than downwards due to excitation of a lower angular momentum mode.)

Why are $l \rightarrow \infty$ spherical modes favored at the same time that $n=0$ bi-spherical modes are observed?

Consider:

Since scattering is some strong function of $b$ (e.g. $e^{-2b}$ above) we expect strongest scattering for small $b$.

So $l$ is large

Notice that this is just the opposite of that for photons or large $e^-$ probes.
For the bispherical case, the small impact parameter position is close to the center of asymmetry of the system. Therefore, as was pointed out above, the fast electron cannot lose energy to the system. Therefore, only large impact parameter events are allowed and the low momentum modes are excited. Notice that the orientation of the axis connecting the two spheres is most important. If the same system is tilted so that the response field between the two spheres has a component parallel to the electron trajectory, the excitation probability is finite.

\[ h = \alpha + \mu \]
\[ A/\varepsilon \]

<table>
<thead>
<tr>
<th>Bispherical Case</th>
</tr>
</thead>
<tbody>
<tr>
<td><img src="image" alt="Bispherical Diagram" /></td>
</tr>
</tbody>
</table>

But by the arguments above, scattering is not allowed in the position where \( b \) is small. Hence:

\[ b \text{ is always large} \]
\[ n \text{ is constrained to be small} \]

Hence: Not only are impact parameter and volume geometry important, but impact parameter orientation is, as well.
All this makes a very nice story, but it isn't the whole truth. Up to now, simple classical electrostatic calculations with bulk dielectric constants have been adequate to explain the experimental results. The illustration below shows the bulk plasmon scattering probability as a function of sphere size for three different probe sizes which are nominally much smaller than the smallest sphere. These are expected to show a linear increase as the column of matter which is illuminated by the probe gets longer. However, pronounced oscillations are observed whose wavelength varies in some complicated manner with the probe size or shape. The most likely reason for this behavior was pointed out by Ritchie in his original paper on surface plasmons. Since the number of conduction electrons in the material must remain constant, the introduction of surface charge density waves at the surfaces requires that the spectral strength of the bulk plasmon be reduced. In the presence of a single externally applied wavelength (such as the parallel applied momentum, governed by the energy loss), the surface plasmon scattering probability is periodic with the system size, and so oscillations are also seen in the bulk plasmon probability.

**Bulk Plasmon Response**

*Overall linearity → e ≈ a*

Expected result occurs due to plasmon spatial quantization parallel to incident probe. Controlled by $\frac{\omega_i}{\omega}$. 
In this case, however, the parallel applied momentum change does not vary as the probe size is changed. Apparently, the momentum change perpendicular to the probe trajectory controls the oscillation spacing. This suggests that the wavepacket nature of the incoming electron needs to be considered when the inelastic scattering intensity is calculated. It seems possible therefore that the phase of the incoming electron -- relative to the outgoing electron and possibly to the phase of the plasmon potentials-- may be important to the scattering intensity. A formulation of this possibility has been formulated by Rose and his student Kohl and is shown below.

Following Rose, Optik V5 139 (1976)

The fast electron potentially becomes

\[ \psi_f = \{k, \} e^{i\phi} \{k, \} e^{i\phi} \]

Then

\[ \omega = \sum_f \left| <f| V_{gh}|i> \right|^2 + |k_f| V_{gh}|i> |^2 \]

\[ + e^{i\phi} <f| V_{gh}|i> \left< f| V_{gh}|i> \right> \]

\[ + e^{-i\phi} <f| V_{gh}|i> \left< f| V_{gh}|i> \right> \delta(\omega - \omega_f) \]

The cross terms are identified as "mixed dynamical form factor"

\[ = S(\phi, t, \omega) \]

It describes the dynamic correlation in time of fluctuations in the system with different spatial periodicities.

\[ S(\phi, t, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \rho_{gh}(t) \rho_{gh}(0) e^{i\omega t} dt \]
They predict that off diagonal matrix elements are fundamental to the calculation of the energy filtered images. For the dipole mode of the single sphere, they show two resonant modes, one parallel to the probe trajectory and one perpendicular to the trajectory. The spatial dependences of the response are summarized below. The prediction for a single sphere is that the scattering intensity has a peak at the metal surface, decays uniformly outside the surface, and becomes constant as the fast electron passes close to the center. If the strong dipolar symmetry of the bispherical mode is considered, then this calculation predicts that the intensity should go to zero as the impact parameter is made small. This was pointed out above.

For dipole Surface Plasmons

\[ \frac{d\mathcal{I}}{dE} = \frac{\eta E^2}{E_0^2} \frac{R}{q_m} \omega A_{\perp} \delta(\omega - \omega_A) \left[ |A_{\parallel}(\omega)|^2 + |A_{\perp}(\omega)|^2 \right] \]

without oxide layer

---

Spherical case

Bispherical case
These predictions are tested below for the spherical and bi-spherical cases. The results are not striking, but seem to confirm the general features of the calculation.

Preliminary Comparison with Experiment

![Diagram showing comparison between spherical and bi-spherical modes]

6a. \(A_n \propto A_k\)  
6b. \(\left| A_n \right|^2 \propto \left| A_k \right|^2\)  

Normalized Probe Position

Radius
To return to the oscillations in the bulk plasmon scattering, we need to formulate the problem as shown below. Briefly, we have a mixed dynamical form factor as described by Kohl and Rose. This form factor has structure in it which is controlled by the particle system. Next we have a complicated phase dependent microscope transfer function which is oscillatory. If these two functions have similar spatial frequencies they should beat and give a pronounced oscillatory behavior to the probability response. It remains to be seen in detail if this explanation can account for the observed oscillation in the bulk plasmon intensity.

\[
\frac{dI}{de} = \int_0^{\Theta} S(\hat{g}, \xi, \omega) e^{-i(y(\xi) - i(0, \xi))} d\theta, d\xi
\]

Microscope Transfer Function

\[
y(\xi) = k_0 \left( \frac{\xi C_0 \xi}{2} - \frac{\Delta f}{2} \right)
\]

\[
k_0 = \frac{1}{\sqrt{C_0}}
\]

Thus \( \frac{dI}{de} \) will oscillate as a function of \( \Theta \) with a periodicity determined by \( C_0 \).

This result demonstrates that

a) interference is possible

b) it might be useful

c) Explains bulk plasmon data
These studies have only begun in an area which promises many surprises and interesting physics. Clearly the unusual nature of the STEM probe is highlighting features which haven't been prominent before. This is annoying sometimes, when clear micro-analytical information is desired. But on the other hand it is encouraging, because the most appropriate probe for the ultra small systems should be the one that has a similar size and momentum content. The unexpectedly strong and sometimes unusual response to this type of probe, is probably a good indication that this idea is correct.
Wavepacket Electron Scattering

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WAVEPACKET ELECTRON SCATTERING

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INTRODUCTION

The aloof scattering of swift (20-1MeV) electrons has been of interest for many years to electron microscopists. Rather than being concerned with high energy (core) loss processes (where most of the energy transfer takes place), the interest has been in the most probably scattering processes, i.e. phonon and plasmon losses (electron microscopes count electrons). Some details of these processes and electron microscopes can be found in references 1-4.

One recently developed area has been the use of relatively small (~5Å) electron wavepackets in a scanning transmission electron microscope or STEM (see references 5-8). Current commercial instruments produce a fairly well characterised (in both amplitude and phase) probe whose form can be simply varied and which can be steered on the angstrom scale. The main use of this instrument has been for microanalysis, but some more academic applications have been to investigate the spatial and energy loss dependance of aloof scattering (refs 9-15, the article by Phil Batsan in these proceedings and Figure 1). Fairly good agreement has been obtained between the experimental results and classical calculations (A. Gras-Marti, these proceedings).

However, if one looks carefully at the Quantum Mechanics, it is a little surprising that the agreement should be so good. Classical approaches, which consider electrons as projectiles, take no account of diffraction. This is an enormous scattering effect at any electron energy (due to relativistic effects), as illustrated in Figure 2. Secondly, because of their very small masses, there are hardly any circumstances when a point charge approximation is valid for electrons, and the Quantum Mechanics requires use of a dispersing wavepacket.
In this note, the formal analysis of the electron wavepacket produced by a STEM instrument is sketched, and some of the diffraction effects briefly described. A substantially more detailed (and better referenced) analysis has been submitted elsewhere (16). It is hoped that this will encourage some needed analysis of the complicated inelastic processes occurring in this experimental geometry.

2. THE STEM WAVEPACKET

The STEM instrument produces a demagnified image of an electron source (cold Field Emission tip in a dedicated STEM) on the specimen as shown in Figure 3. Several magnetic condenser lenses together with a magnetic objective lens are normally used, although mathematically the process can usually be considered by dealing only with the objective lens. Almost any post specimen signal (e.g. X-rays, electrons, acoustic waves, induced current) can be monitored, either with a stationary probe or by a raster scan across the object to produce an image.

Assuming correct alignment of the microscope, the incident wavepacket can be written (with standard electron microscopical notation):

\[ \psi(r) = \exp(-2\pi iz'/\lambda) \int S(u') P(u') \exp(-2\pi i p' \cdot u') d^2u' \]

where:

- \( r \) - three dimensional position vector
- \( z' \) - coordinate along the beam direction (optic axis)
- \( p' \) - vector in the object plane normal to \( z' \)
- \( \lambda \) - electron wavelength
- \( u' \) - wavevector component normal to \( z' \)
- \( S(u') \) - Fourier spectrum of the electron source
- \( P(u') \) - Pupil function of the lens

and a small angle approximation is used. For a defocus \( \varepsilon \) and a spherical
aberration $C_s$, the pupil function can be written
\[
P(u') = \exp(-i\pi(\varepsilon u'^2 + \frac{1}{2}C_s u'^4)) \quad |u'| < u_{ap}
\]
\[
= 0 \quad |u'| > u_{ap}
\]
where $u_{ap}$ is the radius of the objective aperture. In general, $\psi(r)$ at the entrance surface is a decaying, oscillatory function with respect to $r$.
Reasonable approximations for it are a complex Gaussian or (with a little more justification) a Bessel function with a complex argument.

3. BLOCH WAVEPACKET EXPANSION

In this section we sketch the standard Bloch wave expansion for swift electron diffraction(1). For convenience we will employ the non-relativistic wave equations, which strictly should contain relativistically corrected rest-masses and wavelengths.

Considering Schroedinger's equation for the swift electron in the form
\[
(V^2 + (8\pi^2m_e/\hbar^2)[E+V(r)])\psi(r) = 0
\]
where $V(r)$ is the crystal potential and the remainder of the notation is standard, we look for a series solution based upon Bloch waves,
\[
B_j(k_j,r) = \sum_g C_j^{(g)}(k_j)\exp(-2\pi i(k_{g}+g).r)
\]
Substituting 4 into equation 3 with a Fourier expansion of the potential
\[
V(r) = \sum_g \exp(2\pi i g.r)
\]
we obtain
\[
\{1-(k_j+g)^2 \hbar^2/(2m_eE)}C_j^{(g)}(k_j) + \sum_g V C_j^{(g)} = 0
\]
the standard matrix equation for high energy diffraction. This equation only has solutions for specific values of $k_j$, these values being different for each 'j' solution for the coefficients $C_j^{(g)}(k_j)$. The standard representation for the $k_j$ values is a dispersion surface (e.g. 1-3, 16), similar in principle to that used in solid state theory.
The general solution can now be constructed based around the Bloch waves in the form

$$\psi(r) = \sum_j A_j(k_j) B_j(k_j, r) \, d^2k_j$$

where \( A_j(k_j) \) is determined by the boundary conditions on the entrance surface \((z=0)\), and by virtue of the Bloch wave orthonormality is

$$A_j(k_j) = \int B_j^*(k_j, p) \, \psi(p, z=0) \, d^2p$$

With an incident wavepacket, \( A_j(k_j) \) will be some smoothly varying function over a range of \( k_j \) values. The combined term \( A_j(k_j)B_j(k_j, r) \) therefore represents a smoothly varying function of \( k_j \), and as such constitutes a "Bloch wavepacket". The difference between a conventional wavepacket and a Bloch wavepacket is that the former is based upon free electron plane waves, whereas the latter uses the Bloch waves (Wannier functions) for electrons in a periodic potential. We note that Bloch and Wannier wavepackets are standard tools in solid state physics. The wavefunction for the electron is a sum of these Bloch wavepackets, but for most purposes it is convenient to separate these and consider each as a different Boson (i.e. non-interacting) electron wavepacket inside the solid.

To continue the analysis, we now make the substitution

$$k_j(u) = \frac{z}{\lambda} - s_j(u) z + u$$

and writing

$$A_j(k_j) = A_j(u), \quad C_j^*(k_j) = C_j^*(u)$$

(with the energy dependence implied), equation / can be written in the form:

$$\psi(r) = \sum_j A_j(u) B_j(u, p) \exp(2\pi i [1/\lambda - s_j(u)] z) \, d^2u$$

Now taking out the phase term \( z/\lambda \) by putting

$$\psi'(r) = \psi(r) \exp(-2\pi iz/\lambda),$$

which is equivalent to considering an electron wave propagating through the crystal using a rest frame on the zone axis \((z)\), and neglecting any 'z'
diffraction by using an averaged potential $\overline{V}(\rho)$ along the zone axis repeat, $\psi'(r)$ satisfies

$$\left( V_{\rho}^2 + \frac{8n^2mV(\rho)}{h^2} \right) \psi' = \frac{4\pi i}{\lambda} \frac{\partial \psi'}{\partial z} + 4\pi^2 s_j^2(\rho) \psi'$$

Since $1/\lambda \gg s_j(\rho)$, the last term can be neglected in which case equation 13 mimics the time dependent Schrödinger equation, with $z$ 'time-like' and $s_j(\rho)$ 'energy-like' (17). We are now dealing with waves propagating in a quasi two-dimensional solid, and approach which has been extensively used for convergent beam diffraction (e.g. 3, 17) and channelling radiation (e.g. 18).

We next employ a number of concepts standard in wavepacket optics and solid state theory. Expanding as a Taylor series

$$s_j(\rho) = s_j(\rho)|_{\rho = \rho_0} + (\rho - \rho_0).\nabla s_j(\rho)|_{\rho = \rho_0} + ...$$

we may identify a "Group velocity" (similar to the Poynting vector for X-rays) with respect to $z$ of $G_j(\rho) = \nabla s_j(\rho)$, together with higher order terms which mimic the electron optical effects of defocus, astigmatism, coma and other aberrations. This is for a pseudo rest frame along the $z$ axis. In the standard frame with the specimen stationary we employ a group velocity expansion using $k_j$, the standard propagation vector being normal to the dispersion surface. In terms of solid state theory, the second derivative term in the expansion of $s_j(\rho)$ is proportional to the effective mass tensor for the $j$th Bloch wavepacket when considered as a particle. The electron density (equivalent to energy in light optics) for each Bloch wavepacket propagates independent of the other wavepackets along the direction of the Group Velocity, dispersing in the process from the higher order terms of $s_j(\rho)$.

(The standard caveat for wavepacket propagation should be remembered, namely that the higher order terms may severely effect this process.)
4. WAVE PACKET EFFECTS

For reasons of space, we will not become involved here in the detailed interpretation of the mathematical model. For this we refer to a more detailed paper (16). Instead two parts only will be discussed here, namely the "classical" (particle-like) interpretation of the model and some of the coherent interference effects that are possible in the inelastic scattering.

4.1 Classical interpretation

A classical (Eikonal) interpretation considers particles propagating along the Group Velocity directions. Hence rather than one particle moving through the solid, the classical treatment (including diffraction) has a multiplet of particles all propagating in slightly different directions. Depending very strongly upon the precise diffraction conditions, the multiplet may either propagate in parallel directions (channelling) or spread out. Depending therefore upon the crystal orientation, the volume inside which the electron excites aloof events can change.

4.2 Interference inelastic effects

One consequence of this type of analysis is that interference effects arise due to the localization of the inelastic scattering. Localization, either over a broad region for plasmons or at an atom for a core excitation, implicitly involves a wavepacket for the inelastically scattered wavefunction; the uncertainty in momentum of a wavepacket permits spatial definition by the Uncertainty Principle.

We now consider an experiment with a range of (monoenergetic) momenta incident and one particular final momentum and energy sampled. If there are a range of momentum transfers associated with a particular inelastic scattering event, one cannot distinguish between the different momentum transfer events if the final momentum (and energy) are the same. The different events are
thus summed coherently in the outgoing wave.

Hence with an incident wavepacket and a dispersion of momentum transfers (for a particular energy-loss), the final signal is in general sensitive to both the amplitude and phase of the scattering matrix. (We note that this result is inherent in the analyses of Van Hove (19), Rose (20) and Kohl (21). It also follows from the "standard" result that inelastic scattering is sensitive to the form of the incident wavepacket if the wavepacket size is smaller than or comparable to the range of the inelastic potential.)

This implies that the amplitude and phase of the scattering matrix may be measurable by STEM. Experiments to check this are currently in progress.

Summary

We have very briefly detailed herein some of the more complicated effects that can occur with a quantum mechanical wavepacket analysis for a STEM. At present the analysis is in a relatively immature form - some of the broad details are apparent, but many points still require more formal theoretical analysis and experimental testing. For instance, it is not clear whether a more rigorous time dependant analysis will reveal other features. Hopefully future work will clarify some of these points.

Acknowledgments

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REFERENCES

**FIGURE CAPTIONS**

**Figure 1**

STEM energy loss filtered images of MgC smoke particles, with the energy loss value (in eV) marked on the Figures. The main bulk plasmon loss is at 22 eV, that of the surface plasmons at 18 eV. Note that the loss function is essentially continuous from 6 eV upwards. Thus the strong surface signal at 13 eV reflects the surface enhancement of interband transitions (see references 11 and 12).

**Figure 2**

Convergent beam pattern from about 100 nm of Si at 100kV on a (111) zone, courtesy of R. Carpenter. The central disc (magnified in the insert) corresponds to the incident beam (a range of momenta), everything else is due to diffraction. This is identical to the diffraction pattern that would be obtained in a STEM.

**Figure 3**

Schematic diagram of a STEM.
Fig. 3
Comments on the Image Contrast from Inelastically Scattered Electrons

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Comments on the IMAGE CONTRAST FROM INELASTICALLY SCATTERED ELECTRONS

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There is considerable interest in forming images from inelastically scattered electrons, both for studying the scattering process itself, and to obtain information on surface structure and composition. Inelastic surface excitations are prominent features of the energy loss spectrum for reflection off bulk surfaces at grazing angles,1 or in aloof geometry from surfaces of small particles.2−4 Such images can be obtained at high resolution in a dedicated scanning transmission electron microscope (STEM) with a field emission gun simply by adjusting the electron spectrometer; images at various energy losses and width of energy window can be compared at the same objective lens setting, if the specimen is placed outside the lens field. In an energy selecting TEM or in a STEM using a high-excitation objective lens, the post-specimen lens field will act differently on electrons of different energy making quantitative evaluation difficult.

A further complication arises since the inelastically scattered electrons can also undergo elastic scattering, which can dominate the image contrast. Experimentally, dividing the inelastic image by the elastic image (obtained at zero energy loss) is a simple way to remove the elastic contrast. But this is an approximation, since the elastic contrast is only qualitatively preserved at small energy losses. An electron elastically scattered to θ will be scattered an additional θE by inelastic scattering, which will effectively act as a small phase randomisation, reducing the contrast produced by the original elastic scattering. As the energy loss,
and $\theta_E$, increase image contrast becomes less and less preserved by the inelastically scattered electrons.\(^5\) It may be possible to simulate the elastic portion of an inelastic image by increasing the collection angle of the zero loss electrons by the average inelastic scattering angle (several $\theta_E$) before taking the ratio.

Normally one wishes to extract chemical information from the inelastically scattered electrons. Preservation of elastic contrast is only concerned with the trajectory of the fast electron. The trajectory may be confined to surface regions, but the resolution of the chemical information and therefore the surface sensitivity depends on the volume of specimen contributing to the inelastic interaction, which may be much larger. High resolution chemical information is only carried by electrons inelastically scattered through large angles (a manifestation of the uncertainty principle). The chemical resolution $d$ is given\(^6\) by combining the STEM probe size with the mean square impact parameter $b_{\text{RMS}}^2$ for inelastic scattering from zero to $\theta_{\text{max}}$

$$d = (r_p^2 + b_{\text{RMS}}^2)^{1/2}$$

where $r_p$ = probe radius

$$b_{\text{RMS}} = \frac{\hbar v \theta_{\text{max}}}{\Delta E} \left[ (\theta_{\text{max}}^2 + \theta_E^2) \ln(\theta_{\text{max}}^2/\theta_E^2 + 1) \right]^{-1/2}$$

$v$ = electron velocity

This function is shown in Fig. 1 and shows two asymptotes, one at $b_{\text{RMS}} = \frac{\hbar v}{\Delta E} \left[ \ln(2/\theta_E) \right]^{-1/2}$ corresponding to including all angles up to the cutoff angle $\theta_C$, and the well-known expression\(^7\) $b_{\text{RMS}} = \frac{\hbar v}{\Delta E}$ when only small angle scattering is included. For energy losses in the energy range of surface plasmons, chemical resolution can only be of the order of 3–10 nm.
resulting in poor surface sensitivity in either aloof or reflection geometry. Poor chemical resolution means that preservation of elastic scattering contrast, is expected (see, for example, the paper by Milne and Howie,\(^1\) Fig. 3, where surface steps are visible through carbon contamination.) A possible way to improve the chemical resolution and therefore the surface sensitivity of the inelastic signal would be to exclude low angle scattering events from the collected signal.\(^8\) For example, an annular collection aperture could be used. Improving the chemical resolution would also reduce the contribution of elastic contrast to the inelastic signal. However, it is experimentally difficult since most of the inelastic intensity, particularly at low energy losses of surface plasmons, tends to be concentrated at small scattering angles. Low signal intensity would result and problems would arise with multiple scattering (small angle inelastic plus phonon scattering) contributions.

A more fruitful way might be to explore core loss edge structures, which contain information on the conduction band states. They are much more strongly localized than plasmon losses (see Fig. 1) and would, therefore, carry high resolution information,\(^9\) and show strong surface sensitivity. There would be no need to collect electrons "off-axis", so that efficient signal collection would also be achieved.

References
1. R. H. Milne and A. Howie, these proceedings.
References (Contd.)


Fig. 1. RMS impact parameter for inelastic scattering of 80 keV electrons with $\theta_{\text{max}}$ equal to (a) 0.1 mrad, (b) 1 mrad, and (c) 10 mrad.
EELS and Images of GaAs(110) Surfaces

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EELS and Images of GaAs(110) Surfaces

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The recent developments in Reflection Electron Microscopy (REM) (e.g. Osakabe et al. 1980; Yagi et al. 1979) have renewed interest in the interaction of high energy electron beams with surfaces at glancing angle. Here a Scanning Transmission Electron Microscope (STEM) has been used to image surfaces of cleaved GaAs(110) crystals and to examine the energy losses incurred by the electrons.

The classical theory of dielectric excitation by a moving charge has been used to give the 'probability' of energy loss of an electron beam running parallel to a surface (Howie 1983) and these predictions are in quite good agreement with results obtained using MgO smoke cubes (Howie and Milne 1984). These equations can be modified to allow for a small angle of incidence (θ) which gives the total probability of surface excitation as

\[ Q(\theta) = \frac{e^2}{4\pi\varepsilon_0hv}\int_0^{\infty} \frac{1}{\omega} \text{Im}\left(\frac{-2}{\varepsilon + 1}\right) d\omega \]  

showing that the form of the energy loss spectra is independent of θ. In practice, there are considerable changes in the spectra as the angle of incidence is increased, using a parallel electron beam (fig.1). This is due to the depth of penetration and path length of the electrons in the sample which are not included in equation (1). Very low angles of incidence show multiple surface plasmon peaks and as θ increases energy losses from bulk plasmons start to dominate (fig. 1). These spectra give an indication of how the electron paths vary inside the sample with angle of incidence. The areas under the Ga and As peaks at 316 eV and 1323 eV respectively were also observed at different incoming directions of the electron beam, but while quite large changes were observed, the relationship between the various angles of incidence and collection were rather complicated.
Images of the surface showed small steps (fig. 2) and these were present even on a sample which had a layer of carbon contamination, as shown by the speckled contrast. Many of the electrons used to form these images have undergone energy loss (fig. 1) and the effect of this was determined by energy filtering images and these show preservation of the contrast mechanisms (fig. 3). It has been shown (Howie and Milne, 1984) that the surface plasmons generated under normal REM conditions decay fairly slowly into the bulk - 10 nm and so are not sensitive to such effects as surface reconstructions or thin layers. The long wavelength of the surface plasmons excited and the strong concentration of the inelastic scattering at small scattering angles help to explain the similarity between various energy filtered images and consequentially the unfiltered surface images can be interpreted in terms of elastic scattering effects.

REFERENCES

Osakabe, N. et al. (1980) Surface Sci. 97 393
Yagi, K. et al. (1979) Surface Sci. 86 174
FIGURE CAPTIONS

Figure 1a  Reflection EELS spectra taken at increasing angles of incidence (A) shows multiple surface plasmons at $\theta = 2\theta_B$ ($\theta_B =$ Bragg angle)

(B) single surface plasmon at $\theta = 4\theta_B$

(C) bulk plasmons becoming evident at $\theta = 8\theta_B$

Figure 1b  Ratio of 1st plasmon peak/zero peak flattens off as electrons go far enough into the specimen to excite bulk plasmons.

Figure 2a  Crystallographic steps in $\langle 001 \rangle$ and $\langle 110 \rangle$ directions.

Figure 2b  An overlayer of carbon can be detected by the change in the contrast from the surface but the steps on the GaAs are still clearly visible.

Figure 3  Energy filtered images using the zero loss peak (upper) and surface plasmon peak (lower).
Multiple Surface plasmon peaks

 Bulk plasmons start to dominate

Angle of Incident Beam (Bragg Angles)

Fig 1b
Electron Excitation of Dielectric Wedges

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ELECTRON EXCITATION OF DIELECTRIC WEDGES.

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ABSTRACT

Recent experimental data on Electron Energy Loss Spectroscopy of the interaction between small crystallites and beams travelling at a fixed beam-solid surface distance are analyzed in terms of the surface and bulk excitation modes of parabolically shaped wedges. The probability of excitation of the surface modes is calculated for an electron travelling parallel to the wedge surface, either outside or inside the dielectric wedge. The main features of available experimental data for MgO crystallites can be explained by the theory.

EXCITATION OF WEDGE MODES BY FAST ELECTRONS.

Experimental data.
Model calculations.
Relativistic effects.

THE EXPERIMENT

EEL data
30 keV e⁻ beam →

Explore surface excitations,
Fixed impact parameter.
Aloof interaction
THE MODEL

Given $E(w)$, relate surface and bulk excitations to $e^-$ energy losses.

Flat surface.

Parabolically shaped wedge.

- parabolic cylinder coordinates.
- non-retarded analysis.

\[ \nabla^2 \phi = -4\pi \rho \]
\[ \rho(\vec{r},t) = e \delta(\vec{r} - \vec{x}(t)) \]

Specific energy loss
\[ -\frac{dW}{dz} = \frac{e}{\sqrt{t}} \frac{\partial \phi^i}{\partial t} \bigg|_{\vec{r} = \vec{x}(t)} \]
\[ \phi^i = \text{induced or reaction potential} \]

Excitation probability
\[ -\frac{dW}{dz} = \int dw \cdot tw \cdot \frac{e\rho}{dz}\,dw \]
Fig. 1.- Parabolic cylinder coordinates. The parabola
\[ y = \xi(1 - \frac{x^2}{n_0^2}) \] defines the dielectric wedge.

The wedge occupies the region \( n < n_0 \), and the beam
can travel either in vacuum \( (n > n_0) \) or through the wedge \( (n < n_0) \),
through the point \((\xi_i, n_i, z_i)\).
Fig. 2.- Wedge profiles for different values of the parameter $n_0^2$. Note that in the experiments$^{(1,2)}$ the beam passes at a distance $\pm 2\text{nm}$ from the wedge.
Fig. 3.- Excitation probability of surface modes, eq. (13a), for aloof electrons travelling in front of the edge of the wedge, at a distance ~2nm. The parameters $n_0^2$ are: (a) 0.5 nm, (b) 20nm. The latter case resembles a flat semi-infinite wedge. The probability in curve b) has been divided by a factor 5. The electron beam energy is 80keV. (For $\hbar w \gg 21$eV both curves overlap and the curve a has not been plotted for clarity).
Fig. 4.- Excitation probability of surface and bulk modes, eqs. (13a) and (16a), for electrons travelling: (E) along the edge, at a distance of 2nm from the edge; (F) along a lateral surface, at a distance of 2nm from it, and (B) through the bulk of the wedge, along the symmetry plane and at a distance of ~20nm from the edge. The spectrum (B) has been divided by a factor 4.

The wedge parameter is $a_0^3 = 0.5$nm and the electron beam energy is 80 keV.
Fig. 5.- Excitation probability, for given modes $W$, eqs. (13a) and (16a), for electron beam paths at varying distance $d$ from the lateral wedge surface. The paths are far away from the edge, ($n_i^2 \approx 20 \text{nm}$) and range from inside ($d<0$) to outside ($d>0$) the wedge.

Wedge parameter $n_o^2 \approx 20 \text{nm}$. 
RELATIVISTIC BEAMS - Retardation effects.

Planar interface

Maxwell eqn. (Hertz vector)

Retarding force $\rightarrow$ energy loss

$$- \frac{dW}{dx} = -e E_x (st, 0, z, t)$$

$$= \int_{0}^{\infty} dw \, tw \, \frac{\partial \gamma P}{\partial x \, dw}$$

Radiative surface excitation:

Cherenkov criterion $\epsilon' \beta^2 > 1$

(for MgO, $\hbar w \leq 10$ eV).
Fig. 6. Geometry of the interaction of a charged particle Ze, moving with velocity \( v \), at a distance \( z_o \) from the interface between media 1 and 2, characterized by the complex dielectric functions \( \varepsilon_1(w) \) and \( \varepsilon_2(w) \).

Fig. 7. Complex dielectric function of MgO, from ref. (10).

\[ \varepsilon' = \text{Re}(\varepsilon) \quad \text{and} \quad \varepsilon'' = \text{Im}(\varepsilon) \]
**Fig. 8.** Ratio between the probability excitation function for a non-relativistic beam, eq.(20), and a relativistic beam, eq.(19), travelling externally to a given solid surface. For a 100 keV electron beam. The parameter on the curve is the distance from the beam to the surface. The dielectric material is MgO.
Fig. 9. Probability excitation function for a relativistic beam travelling externally to a solid and at a distance of 20 Å, from eq. (20). The parameter on the curves is the energy of the beam. In the experiment reported in ref. (8) the beam energy is 80 keV. The dielectric material is MgO.
Electron Energy Loss Spectroscopy Applied to Surface Studies

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TECHNIQUE:

- ELECTRON SCATTERING FROM SOLID SURFACES TO STUDY GEOMETRIC AND CHEMICAL PROPERTIES OF SURFACES (ALSO ELECTRONIC PROPERTIES) VIA

- ANALYSIS OF CHARACTERISTIC ENERGY LOSS FEATURES IN SCATTERED ELECTRON ENERGY SPECTRUM ASSOCIATED WITH SURFACE VIBRATIONS (OR ELECTRONIC EXCITATIONS)

- COMMON ACRONYMS: ELS, EELS, HRELS, LEELS
ELECTRON ENERGY LOSS
SCATTERING PROCESSES
FOR SURFACES

1) Dipole Scattering — Coulomb scattering from
long range electric field fluctuations

Selection Rule: Sensitive only to dipole
moments perpendicular to surface

\[
\text{-} \quad \text{+} \quad \text{-} \quad \text{+}
\]

Small angle scattering: \( \Delta \theta = \frac{\hbar w}{2E_0} \)

\( \Rightarrow \) 2 step scattering

2) Impact Scattering — short range scattering
which gives large deflection angles

\( \Rightarrow \) diffuse scattering

3) Negative Ion Resonance — electron capture
in excited molecular state and subsequent
electron emission
ELECTRON ENERGY LOSS SPECTROSCOPY (EELS)

\[ e^{-}(E_p) \rightarrow e^{-}(E_p-\omega) \]

- Sensitive to dipoles perpendicular to surface
- Analogous to IR

\[ 0 \quad 100 \quad 200 \]

SCATTERED ELECTRON ENERGY LOSS, \( \omega \) (meV)

- Elastic peak
- Vibrational energy loss peaks

- Sensitive to all orientations
- No IR analogue, more like Raman

INFORMATION OBTAINED FROM EELS

- Identity of and discrimination between atomic and molecular species
- Site of atomic or molecular species
- Orientation of molecular bonds
Applications of EELS

1) Surface Geometry - Adsorbate Site Location

2) Surface Chemistry: identification of adsorbate species

- Compare observed vibrational energies of molecular adsorbates to free molecule energies observed via IR (with caution)

- Hydrocarbon vibrational modes - observe C-H stretching and bending modes which are relatively isolated from bonding effects - good fingerprints

3) Surface crystal properties

- surface phonon dispersion
SCHEMATIC OF EEL SPECTROMETER SYSTEM AT ORNL
EELS Examples

System:
Al(100)

Cu(110) + O

Cu(100) + NO
Cu(111) + NO
Cu(110) + NO

W(001) + H

Type of Information:
Electronic Structure
Adsorption Site Determination
Surface Chemistry
Substrate Structure
Energy Loss Profiles
Al(100), g = (0,0)
\[ \phi = 0^\circ \]
\[ \theta = 15^\circ \]
\[ T = 85^\circ K \]
\[ E_p = E_{Bragg} = 139.0 \text{ eV} \]

Intensity \( I/I_0 \times 10^{-7} \)

Energy Loss, \( W \) (eV)
Surface Plasmon Dispersion

Energy Loss Profiles
Al (100)
θ = 0
θ = 15°
T = 300°K
E_p = 137.5 eV

Zones Levels
23°
21°
20°
19°
18°
16°
15°
14°
13°
12°
11°
10°
9°
8°
6°
5°

Intensity (arbitrary units)

Energy Loss, \(W\) (eV)

W = 10.3 eV

Specular
Cu(110) + O

CLEAN

Anneal (b): 500 K, 10 min

20 L O₂ at T = 80 K

Anneal: 400 K, 10 min

Anneal (b): 500 K, 10 min

• room T exposure \(\Rightarrow (2\times 1)\)

• \(c(6\times 2)\) reported previously - but only for high exposures (\(>10^3\) L) at \(T>300\) K
Cu(110) + O

Exposure Dependence

- **(a)** Cu (110) + O
  - O₂ exposures at T = 25 °C followed by 10 min anneal at T = 250 °C
  - E₀ = 10 eV
  - θ = 80°

- **(b)** O₂: 0.5 L
  - 49 meV (395 cm⁻¹)

- **(c)** O₂: 2.0 L
  - 49 meV

- **(d)** O₂: 6.0 L
  - 49 meV
  - O₂: 20 L
  - 49 meV

No coverage dependence ⇒ shielded site
49 meV ⇒ multiple bonding
CONCLUSIONS:
Atomic oxygen is located beneath the surface in the troughs of the (110) surface. The only difference between the observed LEED structures is the density of sites filled.

Oxygen coverage from XPS:
\[ \frac{2}{3} \] monolayer  \[ \frac{1}{2} \] monolayer
INTERPRETATION OF NO(ad) VIBRATIONAL SPECTRA

\[ \nu = \text{stretching mode} \]
\[ \delta = \text{bending mode} \]
COMPARISON OF DATA TO OTHER SYSTEMS

- **Observed Loss Energies for Cu(110) + NO**

\[
\begin{array}{lll}
\text{^14NO} & 195 - 194 \text{ meV} & 106 - 104 \text{ meV} \\
\text{^15NO} & 191 - 190 \text{ meV} & 104 - 102 \text{ meV} \\
\Delta E = 4 \text{ meV} & & \Delta E = 2 \text{ meV}
\end{array}
\]

- **Nitrosylplatinum Complexes (Miki et al.)**

\[
\begin{array}{lll}
\text{NO stretch} & \text{bend} & \text{NO-Pt stretch} \\
\text{^14NO} & 212.25 \text{ meV} & 64 \text{ meV} & 36 \text{ meV} \\
\text{^15NO} & 208.5 & 67.7 & 35.8 \\
\Delta E = 3.75 \text{ meV} & \Delta E = 1.3 \text{ meV} & \Delta E = 0.2 \text{ meV}
\end{array}
\]

- **Gas Phase Molecular Vibrational Energies (Shimanouchi)**

\[
\begin{array}{lll}
\text{NO stretch} & \text{bend} & \text{N-N} \\
\text{^14N}_2\text{O} & 159.3 \text{ meV} & 73 \text{ meV} & 275.7 \text{ meV} \\
\text{^15N}_2\text{O} & 156.8 & 70.9 & 267.2 \\
\Delta E = 2.5 \text{ meV} & \Delta E = 2.1 \text{ meV} & \Delta E = 8.5 \text{ meV}
\end{array}
\]

\[
\begin{array}{lll}
\text{^14N}_2\text{O} & 163.4 \text{ meV} & 93 \text{ meV} & 200.6 \text{ meV} \\
\text{^15N}_2\text{O} & 161.9 & 91.7 & 195.9 \\
\Delta E = 1.5 \text{ meV} & \Delta E = 1.3 \text{ meV} & \Delta E = 4.7 \text{ meV}
\end{array}
\]

Specific loss peak assignments are normally made by comparing energies and isotopic energy shifts to previously studied systems.
Energy Loss (cm$^{-1}$)

$\text{Cu}(110) + {}^{14}\text{NO}$
$E_p = 10\,\text{eV}$
$\theta_1 = \theta_2 = 80^\circ$
$T = 80\,\text{K}$

$\text{Cu}(110) + {}^{15}\text{NO}$
$E_p = 10\,\text{eV}$
$\theta_1 = \theta_2 = 80^\circ$
$T = 80\,\text{K}$

**NO peak behavior** for $T \geq 115\,\text{K}$, no decomposition seen
CO shift remains when NO vanishes
$\theta$ observed after heating
LEED $\Rightarrow N$ also present following dissociation
Cu(110) + NO

NO Gas Phase
$r_e = 1.15 \text{Å}$

- $I_{\text{Bend/I Elastic}}$
- $I_{\text{Stretch/I Elastic}}$

Caution: Intensities may be influenced by unseen species.
$\text{Cu(100) + NO}$

$\text{Cu(111) + NO}$

$\text{NO(a)}$: singly bound, atop position
$\text{NO(b)}$: multiply bound, bridge position
Temperature Dependent Desorption

Cu (III) + 15NO

When NO is present only in the bridge position, dissociation and recombination as N₂O occurs upon heating.

Conclusions:
1. NO(15) is pre-desorptive species
2. NO(1) is pre-dissociative species
XPS indicates the presence of $N_2O$ at high exposures in contrast to the EELS results. The explanation may be a second $N_2O$ state at high coverages which is parallel to the surface.
Information on substrate geometry in adsorbate systems can sometimes be obtained with EELS.

W(001) RECONSTRUCTION MODELS

\((\sqrt{2} \times \sqrt{2})R\ 45^\circ\) - CLEAN

"ZIG-ZAG" P2 mg

\((\sqrt{2} \times \sqrt{2})R\ 45^\circ\) - H

"DIMER" C2 mm

Commensurate Phase

\(\bigcirc\) RECONSTRUCTED W SURFACE LAYER

\(\bigcirc\) W SECOND LAYER

\(\bullet\) HYDROGEN ATOMS
**Adsorbate Site Symmetry**

Atomic Adsorbate - 3 vibrational modes are possible:
- $v_1$: adsorbate-substrate stretch
- $v_2$: bending (wagging)
- $v_3$: bending (asym. stretch)

For W(001), substrate has 4-fold symmetry.

If adsorbate site has 4-fold symmetry, $v_2 = v_3$  
⇒ maximum of 2 modes observed and bonding is to single atom or hollow site.

If site has 2-fold symmetry, $v_2 \neq v_3$  
⇒ maximum of 3 modes observed and bonding is at bridge site.
$W(001) + H$

$T = 300K$

Hydrogen Exposure:

1 L

$\theta = -10^\circ$

.05 L

$\theta = -10^\circ$

SPECULAR $\theta = 0$

ENERGY LOSS (keV)
Confinement of Low-Energy Electrons Near Metallic Surfaces for Plasmon Generation


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CONFINEMENT OF LOW-ENERGY ELECTRONS
NEAR METALLIC SURFACES FOR PLASMON GENERATION*

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ABSTRACT

A novel technique is used in studying the energy loss spectrum of aloof (nontouching) electrons with surfaces. Metallic foils perforated by microchannels restrict the maximum impact parameter to the channel radius (20-200 nm). The passage of electrons (20-2000 eV) near the cylinder walls results in prominent energy-loss structure. In silver, a resonance loss peak at 3.5-3.6 eV is attributed to the formation of surface plasmons on the silver channels. The problem is modeled by a classical calculation of a charge moving parallel to a plane surface whose response is given by tabulated optical properties. The differential probability is calculated assuming the lower limit to the impact parameter is determined by surface roughness. The calculations are compared with the experimental data.

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Fig. 1. The maximum probability for coupling with surface plasmons is obtained at grazing incidence. The range of the s.p. potential outside the bulk is exponentially damped in distances of the order of 100 Å. The phase velocity of the induced s.p. tends to couple to the electron's velocity. Thus to see the effects we need electrons to pass very close to the surface at the grazing incidence.

Fig. 2. Historically, there have been numerous experiments involving reflection off surfaces or penetration through thin foils. Unfortunately these probe the plasmon fields from infinity to the surface, stimulating volume plasmons and other energy loss channels, and, being dominated by the collision with the surface, provide little direct information about the exponential nature of the s.p. fields.
ALOOF FORWARD ELECTRON LOSS SPECTROSCOPY

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- Probe the near-surface field coupling of surface plasmons and near-grazing (but non-touching) electrons at low energies. (20-2000eV)

- Requirements:
  - Angle: $\theta_s(\theta) = \theta_s(0)/\cos \theta$
  - Distance: $\delta_s \sim e^{-Kx}$, $\lambda = \frac{2\pi}{K} \approx 100\text{Å}$
  - Velocity: $v_e \approx \frac{\omega}{K_x}$

Fig. 1

- Historical:
  - Reflection & Transmission
    - probe plasmon fields from infinity to surface
    - volume plasmons & other energy loss channels stimulated
    - little direct information on the near-surface region (dominated by intimate contact with surface)
  - ALOOF (non-touching)
    - High energy: STEM on MgO and Al particles
      - ~100 KeV electron beam focused near a surface
      - impact parameter control: ~1nm
      - momentum resolution ~ 0.5 Å⁻¹
      - energy resolution ~ 1eV
      - volume plasmon production through penetration
  - Low energy: Parabolic trajectory near a plane
    [Lecarte, Ballu, Nowack, PRL 38, 36 (1976)]

Fig. 2
There have been a limited number of aloof studies. The nicest experiments utilize the tightly focused beam of electrons available in a STEM. MgO and Al particles have been studied by this means. These use a high energy beam so that precise control of the impact parameter can be had. Typical transverse momentum control is ~0.5 Å. The energy resolution is limited by primary beam energy and width to ~1 eV. One can choose to stimulate volume plasmons by moving the beam to pass through the particle.

The experiments at low energies have been very limited. The first attempt utilized a uniform electric field to force a parabolic trajectory of the electrons just above a Mo surface. An obvious difficulty is maintaining an adequately close approach to the surface without touching it. Nevertheless, a peak in energy loss was seen at ~1 eV which was attributed to a s.p. on the Mo surface.

**Fig. 3.** We are using a much more straightforward approach to confine impact parameter which was developed within our group. We have been able to prepare metallic foils containing submicron-sized channels of radius ~200 Å and up. The electrons which strike the foil are blocked from detection, while the electrons which pass through the channels have a maximum impact parameter given by the cylinder radius, which we can measure. If the plasmon wavelengths generated are of the order of the cylinder dimensions, one might expect to see some effects of geometrical dispersion. The momentum resolution of such an experiment is greatly dependent upon the electron optics and varies with energy but typically is ~0.1 Å⁻¹ (sufficiently large to encompass all aloof scattered electrons at this energy range). A great advantage in using low energies is the energy resolution of ~0.1 eV. We can change the excitation probability by modifying the channel radius and length.

**Fig. 4.** The dispersion curve of a free electron gas for a vacuum cylinder (upper) (with radius corresponding to 200 Å and 470 Å for silver) and for the inverse geometry of a dielectric cylinder in a vacuum (lower) is plotted. The vertical dashed lines correspond to phase velocity coupling at the indicated electron energies. For the energy range of the present experiment, even for the smaller cylinder radius, the dispersion is very flat and becomes even more so for silver which has the volume plasmon energy very close to the surface plasmon energy.
- Microchannels through a metal foil

- rigorous confinement of impact parameter
  \( b \leq \text{radius of channel, } 200 \text{ to } 2000 \text{Å} \)
- geometrical dispersion \( \tilde{K}_{sp} = \tilde{K}_x + \tilde{K}_y \)
- momentum resolution \( \sim 0.1 \text{ Å}^{-1} \) (typ.)
- energy resolution \( \lesssim 0.1 \text{ eV} \)
- excitation probability governed by channel radius
  and channel length \( (P \sim 10^{-7} \text{ to } 10^{-5}) \)

**Fig. 3**

---

**Fig. 4**

Surface Plasmon Dispersion-Cylindrical Geometry
Fig. 5. We use two methods to fabricate these channeled foils. Begin with a commercially available micropore filter to be used as an evaporation base. (The filters are made by etching the track damage caused by high-energy particles which pass through a polycarbonate membrane.) Evaporate silver @45° while rotating the filter. This coats the inside walls of the channels to a depth equal to the channel diameter. Dissolve the filter. Reevaporate on both sides to narrow the channels and provide a clean surface.

Fig. 6. Typical SEM picture of a replicated foil with random positions and axes orientations up to ~30°.

Fig. 7. Taking a look at a rolled-over foil, we see a channel protruding so that the filter material has almost completely been removed, leaving the exposed channel.

Fig. 8. Hole size distribution of a filter replication.

Fig. 9. A second method to produce channeled foils is to use a computer-controlled SEM to expose a thin membrane of an electron-sensitive resist (PMMA). The exposed areas dissolve during subsequent development. The membrane is coated on both sides to present a metallic surface for interaction. This shows a pattern of hexagonal close-packed channels ~1000 Å diameter, ~3000 Å apart.

Fig. 10. A closer look at these channels. The advantage here is that the channel axes are all aligned, and a higher density can be achieved.

Fig. 11. Hole size distribution.

Fig. 12. The experimental apparatus for observing energy losses by electrons passing through the microchannels is represented schematically here. An electron gun injects a beam into an electrostatic analyzer at about 10-30 eV, producing a beam with a width of 50-100 meV. A zoom lens accelerates the beam to the desired interaction energy where the electrons encounter the sample. The electrons which make it through the channels are then decelerated by another zoom lens for injection into an energy analyzer. This is a fixed angle at 0°, but the lens should be able to capture all the aloof scattered electrons.
ORNL-DWG 81-15129

Fig. 5

Fig. 6
Channel Size Distribution

Fig. 11

Fig. 12
Fig. 13. A typical energy loss spectrum. Most of the electrons which pass through the channels do not suffer any loss. An asymmetric peak is centered in the neighborhood of the silver surface plasmon (3.63 eV).

Fig. 14. Comparing this to optical data, we see: elastic peak position, energy loss spectrum for the silver foil, optical surface loss function—the surface analogue of the bulk optical loss function Im(-1/ε), and likewise for a silver film which has several hundred angstroms of gold overcoated. The optical data is in qualitative agreement with the electron data for both position and asymmetry. However, there is an obvious broadening which we will attempt to explain later.

Fig. 15. A family of experimental energy losses from 30 to 1200 eV. The low energy-loss tails are real and predicted from the theory. The resonance peak dominates over the continuum losses until the very low energies.

Fig. 16. To model the situation theoretically, we have used:
(1) Quantum mechanical calculation, (2) classical calculation based upon cylindrical geometry, and (3) finally a classical calculation based upon planar geometry which has proven adequate for the experimental parameters used to date. The first equation gives the work done on a charged particle per unit distance traveled moving a distance y above a surface with dielectric function ε(ω). To compare with experiment, we need the differential probability with energy so that the integral may be removed. Finally, we integrate over the cylinder cross section, keeping the upper limit constrained due to small-scale roughness on the cylinder walls. (The radius of the cylinder is "a" and the effective roughness height which would absorb or scatter electrons out of the beam is "d").

Fig. 17. Total surface plasmon probability dependence. "d" was adjusted to best fit the data and is consistent with measured roughness values for evaporated silver films. The integrated probability increases with energy and decreasing channel diameter (forcing more electrons near a surface).

Fig. 18. The foils produced by electron beam lithography have their axes aligned so that we can probe angular effects by tilting the foil in the beam. As the channels are tilted, the free aperture through which
- Energy spectrum after passage through silver microchannels

Fig. 13

Fig. 14
Fig. 17

Electron Energy (eV)

Probability

$\alpha = 300 \text{ Å}$

$\alpha = 1600 \text{ Å}$

$d = 25 \text{ Å}$

$L = 2a$

Fig. 18

Integrated Transition Rate

$\frac{L}{D} = 2.25$

Ratio:
Perimeter to Area

$\theta$ (degrees)
the electrons pass is formed by the intersection of two ellipses. Since the excitation probability is a strong function with distance from the wall, we might expect as a rough approximation that all transitions occur very close to the wall. As we study the ratio of the area of a perimeter strip to the free aperture area, we see reasonable agreement with experimental data.

Fig. 19. Finally, this shows a comparison of the shapes between the theoretical and experimental resonances. The dotted curve is theoretical and must be broadened by the experimental instrumental width to get the solid curve to compare with experimental points. We see an additional broadness in the experimental curve which we are presently attributing to roughness effects. For example, the fundamental mode of a surface plasmon standing wave on a silver sphere occurs at 3.5 eV. The combination of planar resonance and localized resonances (on roughness features) may be required to explain the shape.

Fig. 19

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Aloof Scattering of Low-Energy Electrons Near a Submicron Sphere

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We consider an electron moving at constant velocity in the $z$ direction with speed $v \ll c$ at distance $x_0 > a$ from the center of a sphere of radius $a$. The sphere is assumed to be sufficiently small that electrodynamic retardation effects are negligible, and the response of the sphere at angular frequency $\omega$ is characterized by the local dielectric function $\varepsilon(\omega)$. The data on $\varepsilon(\omega)$ is obtained from measurements of the bulk optical properties, e.g., reflectivity and absorption coefficient measurements.

If the $z$ component of the electric field ($E_z$) of the surface charge density induced on the sphere is calculated and evaluated at the electron, one may obtain the energy loss per distance traveled by multiplying $E_z$ by the charge on the electron. This is $dW/dz$, which is supplied by the external agent which maintains the constant velocity. The differential probability for losing energy between $\omega$ and $\omega + d\omega$ is found from

$$W = \int_{-\infty}^{\infty} \frac{dW}{dz} \, dz = \int_{-\infty}^{\infty} d\omega \, \omega \frac{dP}{d\omega}$$

In the case at hand, in Hartree atomic units ($\hbar = m = 1$),

$$\frac{dP}{d\omega} = \frac{16a}{2\pi v^2} \sum_{\ell=0}^{\infty} \sum_{m=0}^{\infty} \frac{(2 - \delta_{\ell m})}{(\ell+m)! (\ell-m)!} \left( \frac{\omega a v}{v} \right)^{2\ell} \frac{2^m}{m!} \left( \frac{\omega a}{v} \right)^{2m} \Im \left[ \frac{\varepsilon(\omega) - 1}{\varepsilon(\omega) - \varepsilon_{\ell}} \right]$$

where

$$\varepsilon_{\ell} = - \frac{\ell + 1}{\ell}$$
and \( K_m(\omega a/v) \) is the modified Bessel function of the order \( m \), and \( \text{Im} \ f \) is the imaginary part of the complex function \( f \).

As \( x_o \) is made closer to \( a \), more and more terms in the series must be kept; but for \( x_o \gg a \) the series is approximately the dipole terms \( (\ell = 1) \). These are essentially the same as given in the well-known text by J. D. Jackson.\(^1\)

In aloof scattering through microchannels in a silver foil, it is likely that surface roughness features dominate the energy losses.\(^2\) A simple model of roughness on the nanometer scale is provided by the energy loss probability calculated for a series of spheres. This neglects substrate effects such as the imaging of a given sphere in its supporting material; but other effects, such as the direct electron-planar surface interaction can be added to the above analysis. A comparison of this model with experiment is currently under way, and the initial work is very encouraging.

By including relativistic effects, the model developed above may be applied to the case in which the incident electron beam is that of a scanning-transmission electron microscope, a case of much current interest. Quantum effects in profiling the beam are of importance in this instance, but the simplest model is often the best starting point, and the relativistic correction to the above results provides this.

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The Theory of Inelastic Atom-Surface Scattering

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The Theory of Inelastic Atom-Surface Scattering

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I. Introduction

I would like to discuss a few ideas on recent work in the inelastic scattering of low energy atoms by solid surfaces. But first, for those who may not be familiar with this field of research, let me review briefly the sorts of experiments that this is related to. Figure 1 shows schematically the situation, a nearly monenergetic ($\Delta E/E \sim 1\%$) and well collimated ($\Delta \theta \sim \frac{\theta}{2}$) beam of light atoms or possibly molecules is incident on the surface at an angle $\theta_i$, and particles reflected at angles $\theta_f$ are detected. For most work on inelastic scattering the detector must have a time of flight analyzer. A partial list of projectiles and representatives of classes of surfaces typically used in such experiments is shown on Fig. 1. The interesting point is that the projectile energies are in the 25 $\sim$ 100 meV range (thermal energy particles, i.e. 70 K $\sim$ 300 K) and consequently their de Broglie wavelength is of the order of the surface lattice spacing and the energies are in the same range as those of the phonons. Thus diffraction can be observed from which one can obtain the structure and corrugation of the surface. Inelastic scattering measurements can give information on the surface lattice vibrations. Comparison of data with theory can give the atom-surface interaction and the repulsive part of this potential gives directly the surface electron density profile, since currently it is believed that the two are proportional.
The general characteristics of surface scattering are roughly outlined in Fig. 2. The incoming atom first sees the attractive $1/z^3$ Van der Waals potential, then close to the surface the force is strongly repulsive due to Pauli exclusion of the atomic electron cloud overlapping with the tail of the surface electron density. Between these two regions is an attractive well which contains the bound or physisorption states. A list of readily observed effects is shown in Fig. 2 along with a few early references. The initial research was done by Stern, Estermann and Frisch in a series of experiments in the late 20's and early 30's, these experiments being the second confirmation of the hypothesis of de Broglie's matter waves. Little work was done until the late 60's with the advent of energetically well resolved supersonic nozzle beams. The experiments of Fisher stimulated a revival of the theory and the field has been building rapidly ever since.

Fig. 3 shows the definitive experiment on the benchmark system, He scattered at a LiF surface. The incident beam energy is 21 meV, corresponding to a wave vector of 6.4 Å\(^{-1}\). The diffraction peaks are well defined and very strong compared to the specular beam. This immediately makes it clear that this must be described by a strong scattering theory; perturbation theory is inadequate. In between the diffraction peaks is a very small background (typically down in intensity by a factor of order $10^{-3}$) and this contains inelastically scattered particles, the subject of this discussion.

Fig. 4 is an example of resonant scattering with the bound (or adsorption) states in the physisorption potential well. (This process is also called selective adsorption.) This resonant scattering occurs when the incident beam can diffract into the bound state. This bound state
channel is an intermediate state, degenerate with the other emerging elastic beams, and this oftentimes causes a substantial rearrangement of the diffracted intensities. These can be seen as maxima, minima, or combinations of the two as one follows the intensity of a diffracted beam as a function of one of the incident beam parameters (energy, azimuthal angle, or polar angle). Fig. 4 shows a plot of the specular beam for the 21 meV He/Cu(113) system [21 meV He incident on a Cu(113) surface] and the resonances of the three bound states with the (10) evanescent diffracted beam (or alternatively stated, with the (10) surface reciprocal lattice vector) are visible as minima. Fig. 5 shows a stronger scattering system, 21 meV He/Cu(115). This demonstrates that resonances can be a dominant influence on the diffraction curve, and furthermore both maxima, minima, and mixed structures can appear. The signatures of these resonances have been studied by Wolfe and Weare (Phys. Rev. Lett. 41 (1978) 1663) and by García, Celli and Goodman (Phys. Rev. B19 (1979) 634. Resonances are stressed here because they can also be phonon assisted, situations in which the incident beam can resonate with the bound state via an inelastic exchange. This is a very important process in inelastic scattering.

II. Kinematics of Inelastic Scattering

A good deal of experimental work prior to 1980 (i.e. Williams and Mason; D. Miller; B. Feuerbacher; Boato et al.) had shown that individual inelastic processes could be observed, although they were limited primarily to looking at the acoustical region of the Rayleigh phonon mode. However, they did seem to indicate two very important points:

1.) single phonon events dominate if the projectiles are not too energetic (< 100 meV), and
2.) Rayleigh modes seem to contribute to the inelastic scattering more strongly than the bulk modes.

The kinematics of a single phonon process are shown in Fig. 6. The conserved quantities are energy (accounting for the exchanged phonon) and momentum parallel to the surface. Because of the broken symmetry perpendicular momentum is not conserved. Upon combining these two conservation laws the phonon frequency $\omega$ can be expressed as a parabolic function of $\Delta K$, the total exchanged parallel wave vector. This can be plotted as a function of $\Delta K$ on the same graph as the surface projected phonon dispersion relation. This is shown at the bottom of Fig. 6. The crossings of the parabola function (dashed lines) with the dispersion curves are possible phonon transfers for the given incident and detection angles. Since the bulk and Rayleigh modes are often well separated, a time of flight analysis can distinguish between them.

The dashed parabola to the right in Fig. 6 is a special case. At one point the scan parabola and Rayleigh dispersion curve are tangent. In this case there is an infinite density of phonon states contributing to the scattering cross section and this causes a weak singularity at that angle. Such processes are called "kinematical focusing" (Benedek, 1975) and they can be used to enhance certain modes, or even to map out parts of the dispersion curve without a time of flight analysis.

An example of the work done by the group of Toennies et al. at Göttingen is shown in Fig. 7, for He at $\sim 20$ meV incident on LiF(001) in the $<100>$ direction. The scan parabola for the incident angle of 64.2° (the detector is at a fixed angle of 90° to the incident beam) shows the possibility of observing two creation events and one annihilation event with the Rayleigh modes as well as an exchange with bulk modes with small
△K. All of these events appear as corresponding peaks on the time-of-flight intensity plot. (There are also two additional observed peaks which are well understood experimental artifacts.)

A series of experiments like this can be used to map out the entire surface projected dispersion relation as shown for the case of LiF in Fig. 8. The dotted line is an older calculation due to the group of de Wette; the full curves are by Benedek et al. calculated with some additional surface relaxation as indicated by the experiment. However, there still remains a small discrepancy in the Rayleigh modes at the band edge which is not understood to date.

This is just an example of what has been done. The Göttingen group has done extensive work on many of the alkali halides (even seeing surface optical modes on NaF), on semiconductors such as Ga As, and on the metals Au, Ni, and Cu. It is fair to say that this group has raised the technique to the point where we can call it a true surface phonon spectroscopy. Fortunately, the next speaker at this workshop is one of the people most responsible for making it work.

III. Dynamics of Inelastic Scattering

One of the fortunate aspects of the theory is that at the present point it is not extremely complicated in concept, although detailed calculations can be somewhat lengthy. Figure 9 gives some relevant equations. Typically one starts from the transition rate \( w_{fi} \) expressed in terms of the transition matrix \( T_{fi} \). The experimentally measured quantity is the differential reflection coefficient (Manson & Celli, 1970)

\[
\frac{dR}{dE_f d\Omega_f}
\]
which is the differential cross section per unit surface area. This is obtained by multiplying $w_{fi}$ by the appropriate density of states and then averaging over initial crystal states and summing over final crystal states. The average over crystal (or phonon) modes is the non-trivial part but recently this has been extended to all orders in perturbation theory for a harmonic lattice (Armand and Manson, 1984).

Often a calculation in the distorted wave Born approximation will suffice and the salient features of such a result are shown in the middle of Figure 9. The important features are the factor of $1/\rho$ where $\rho$ is the surface mass density, the square of a spatial matrix element $|<f|v|i>|^2$ which contains the particle surface interaction; the factor $|e_z|^2/\omega$ is the phonon spectral density (or its surface normal projection); $n(\omega)$ is the Bose-Einstein factor; and the $\delta$-functions contain the kinematical conservation laws. The interaction potential $V(r,u)$ is split into a "large" or distortion term $U$ and a remainder $v(r,u)$, and often the remainder is expanded to first order in the surface displacement $u$.

A model which is frequently used is the "corrugated hard wall" (CHW) in which the repulsive potential is replaced by an infinite step (hard wall) which is corregated and also vibrates with an appropriate spectral density. A recent calculation using this model is shown in Fig. 10 and compared to selected data for LiF. It is clear that most of the dominant features, including Rayleigh modes (marked with a dot) and bulk acoustic modes (LA) are reproduced.

Resonant inelastic scattering is quite important and the essential theory is shown on the last two lines of Fig. 9. The resonance can be projected and the T-matrix $T_{fi}$ written as the sum of a direct term $h_{fi}$ and a resonant part with a self energy in the denominator. Since the self
energy $h_{bb}$ is complex the denominator never vanishes and there are no divergences. The direct term $h_{f1}$ is just a matrix similar to $T_{f1}$ but does not contain the resonant state in the intermediate sum. If there should be more than one resonance or near resonance occurring simultaneously, the formalism can be readily generalized. Single phonon processes can be built into this formalism and a recent calculation, again using the corrugated hard wall model, is shown in Fig. 11. The top series, a, is the time-of-flight data taken at closely spaced angles. The second row, b, is the distorted wave Born approximation, and the third row, c, is the calculation including resonances. Clearly, under some circumstances the resonance conditions can greatly enhance certain modes. This points out a significant advantage of the atom scattering technique, by using resonance and/or kinematical focusing the experiment can be "tuned" to enhance the response to certain modes which might otherwise be weak contributors to the scattering intensity. The observation of surface optical modes in NiF was done in precisely this manner (Skofronick et al. 1984).

Other points worth mentioning, Figs. 10 and 11 indicate that a single phonon approximation can be quite adequate in spite of the fact that there is substantial loss of unitarity due to multiple phonon and other possible processes. In addition the Rayleigh mode scatters strongly in comparison to the bulk modes and hence is readily visible.

IV. Inelastic Effects on Resonance Lineshapes

While on the subject of resonances, it is of interest to discuss the effect of inelastic exchange on elastic resonance lineshapes. It is clear that inelastic exchange plays a large role in resonances because the atom is near the surface for long periods of time. In fact it is this effect
which makes resonance scattering enhance the inelastic signal. It is also reasonable to suspect that there is a large amount of multiphonon exchange under resonant conditions. These multiphonon exchanges have been successfully modeled by two methods: 1) the use of a small complex part in the potential near the surface (optical potentials), and 2) the addition of thermal attenuation on the scattering amplitudes through Debye-Waller factors.

In order to see how these methods work, a few equations are written down on Fig. 12. The first line gives the intensity $I_G$ of an elastic diffracted peak. The next two lines are a repeat of Fig. 9 for the coupled T-matrix equations when an isolated resonance is projected. (The extension to multiple resonances is straightforward.) Thus, with obvious definitions, the diffraction intensity is of the form $I_G = I^0_G \frac{|1-i\beta/(x-i)|^2}{|1-i\beta/(x-i)|^2}$. This is a general form for a resonance and can have at most one maximum and one minimum as shown in the small graph in Fig. 12. The width of the structure is given by the imaginary self energy, $\text{Im}(h_{bb})$, and the signature (maximum, minimum, or mixture of the two) is dictated by the two parameters $\text{Re}(b)$ and $\text{Im}(b)$. For example, if $|\text{Re}(b)| \gg |\text{Im}(b)|$ it can be readily shown that there is only one extremum and this is a minimum for $-2 \leq \text{Re}(b) \leq 0$ and a maximum otherwise. The simulation of inelastic scattering, either by an optical potential or by Debye-Waller factors has the tendency to make $\text{Re}(b)$ smaller in magnitude. Thus in the frequent situation in which $\text{Re}(b) < -2$ in a purely elastic calculation (corresponding to a maximum in the resonance), the addition of a small amount of simulated inelastic scattering can increase $\text{Re}(b)$ into the region $-2 \leq \text{Re}(b) \leq 0$ and the resonance changes abruptly from a maximum to a minimum. Simultaneously, $|\text{Im}(h_{bb})|$ is increased and the resonance becomes broader.
Fig. 13 shows a typical calculation illustrating this. The system is 21 meV He/Cu(113) and the resonance involves the (10) evanescent diffraction beam and the ground state. The curve marked a is the purely elastic calculation using a potential which reproduces the elastic curves off-resonance nearly exactly. At resonance the elastic calculation is clearly in disagreement with the experimental points of curve b which shows a broad minimum. However, the multiplication of the elastic scattering amplitudes by a Debye-Waller factor corresponding to the thermal attenuation of the experiment at the measured surface temperature of 70 K changes the resonance to the broader sharp minimum in curve d. Curve c is with the addition of an average over the angular spread of the incident beam. This method has been used successfully to explain a number of experiments, and was first used for the case of 21 meV He scattered by a graphite surface (Hutchison, Phys. Rev. B22 (1980) 5671).

V. Thermal Attenuation

Thermal attenuation has been mentioned several times above and we will now take a closer look at this as our last topic for consideration. Figure 14 gives a few of the equations currently used in interpreting this effect. In keeping with other types of scattering such as bulk neutron or x-ray diffraction, the thermal attenuation seems to roughly follow a Debye-Waller formula. This is shown in the first line of Fig. 14 where each intensity is multiplied by $\exp(-2W)$ with $W = \langle(\Delta k \cdot \mathbf{u})^2\rangle$. Since the perpendicular momentum exchange is relatively large, $\Delta k_z = |k_{iz}| + |k_{fz}|$, the exchange of parallel momentum can usually be neglected. For sufficiently large temperatures $T$ we have $\langle u^2 \rangle \ll T$ and hence a logarithmic plot of intensity versus temperature should give a straight line.
The most extensive studies of this effect are by Lapujoulade et al. and Fig. 15 shows a fairly typical example, 63 meV He/Cu(110) with four diffraction peaks showing relatively constant slopes on a logarithmic plot. Clearly, the Debye Waller analysis is an approximation with the correct tendencies. There are two classes of theories from which such a Debye-Waller picture can be extracted, the corrugated hard wall model (Benedek & Garcia (1981); Armand & Manson (1981)), and semiclassical approximations (Levi (1979); Levi & Suhl (1979); Meyer (1981); and Schinke & Gerber (1983)). However, there are two main difficulties with such theories, they often do not get the slopes of the logarithmic intensities correctly and these two models often do not give good results for the elastic intensities (this latter objection is particularly true for the hard wall models). Furthermore, a strict Debye-Waller analysis is really not a good explanation for the experimental data, because the logarithmic intensities as a function of temperature tend to be decreasing functions, but with a pronounced curvature as is more clear from Figs. 16 and 17.

As a more fundamental approach we have examined the thermal attenuation by calculating exactly the inelastic effects through the perturbation series (Armand and Manson (1984)). In this approach we start by noting that the intensity $I_G$ of a diffracted peak can be written in terms of the thermal average of the T-matrix as shown in Fig. 14. The thermal average $\langle T_G \rangle$ can be evaluated to all orders in perturbation theory through the use of integral representations of the Green function denominators. (We mention in passing that this method can also be extended to treat inelastic scattering to all orders.) Extensive data exists for the He/Cu(100) system for which the diffraction peaks other than the specular are negligible. The potential model is shown in the last equation on Fig.
14 and is a Morse potential with a repulsive part which vibrates with the amplitude $u$. This potential is chosen because, when used with the appropriate static corrugations, it gives excellent agreement with elastic scattering data for all of the stepped copper surfaces which have been studied to date. Note that it exhibits the proper exponential decay in the repulsive part, since it is now understood that this should be proportional to the surface electron density. The elastic calculations are very sensitive to the slope parameter ($\kappa = 1.05 \text{ \AA}^{-1}$) showing that atom scattering apparently measures the slope of the electron cloud very accurately. The vibrational displacement $u$ is taken from the phonon spectral density calculated by Armand for the Cu(100) surface.

The results of the calculations are shown on Figures 16 and 17. On both of these figures the calculations (solid lines) use a phonon spectral density which is averaged over a cell of four surface atoms. This accounts for the fact that the incoming He atom is large and interacts simultaneously with several surface atoms (i.e. the Armand effect). In addition the spectral density was softened by a multiplicative factor of $\frac{1}{2}$ which in part accounts for correlations with more than 4 atoms, for other correlation effects (Garcia & Maradudin, 1984) and for the fact that the He atom is really interacting with the tail of the surface electron distribution which vibrates with a smaller amplitude than the ions themselves. The important point is that the curvatures in the calculated intensities match the experimental points very well. This is a clear demonstration that thermal attenuation, particularly for soft surfaces, is not adequately described by a simple Debye-Waller factor. This can be understood by noting that the important higher phonon exchange processes come from higher order terms in the perturbation series, unlike the situation in neutron or x-ray scattering.
Figure 18 shows the importance of treating the potential well exactly. There the solid curves are for a number of different well depths D. Clearly an exponential repulsive potential (D = 0) can underestimate the inelastic scattering by a large amount. The two dashed curves show the approach from an exponential potential to the hard wall limit by increasing \( \kappa \). (The curves shown are for \( \kappa = 2.5 \text{ \AA}^{-1} \) and \( \kappa = 5 \text{ \AA}^{-1} \), the latter being essentially the hard wall limit.) An interesting calculation is the curve shown by the dash-dot trace. This is a calculation in which the true well depth of 6.35 meV has been accounted for by adding it to the perpendicular part of the particle energy (this is known as the Beeby approximation). It is somewhat surprising to find that this is such a poor approximation to the exact result.

Finally, we note that the calculations in Figs. 16-18 were carried out with the exchange of a single virtual phonon. This is further theoretical confirmation of the experimentally observed fact that single phonon events are dominant as long as the particle energy and surface temperature are not too large.

VI. Conclusions

We have reviewed above a few of the aspects of inelastic scattering that are of current interest. Clearly in some of these areas the experiment is pushing the theory quite hard. The technique of atom-surface scattering has some rather remarkable advantages for investigating surface phonons and the most important are:

1) single phonon processes are dominant over a large range of conditions,

2) Rayleigh waves give surprisingly strong signals, and
3) Kinematical focusing and inelastic resonances can be used to select and enhance the intensities due to certain vibrational modes.

Already we can say that this is a tool for surface phonon spectroscopy, and we have shown here that investigations of resonances and thermal attenuation can give further information on the particle-surface interaction. Atom-surface scattering has already proved its extreme usefulness in structural analysis. In the future the extreme sensitivity to surface impurities will surely be exploited as a useful diagnostic tool for looking at adsorbed particles, adsorbed layers, and even disorder. Inelastic scattering will surely be useful for studying surface phase transitions and reconstructions. In short, atom surface scattering is rapidly becoming an accepted and essential addition to the list of techniques used in surface analysis.
Atomic and Molecular Surface Scattering

He, Ne, Ar, H, H₂, NO, LiF, NaF, NaCl; Cu, Ag, Au, Pt, W; GaAs, Si, C (graphite, diamond)

Low energies

\[ E_i \sim 25 - 100 \text{ meV} \quad (70° - 300°K) \]

\[ d \sim a \text{ and } E_i \sim tw \]

Information to be gained

1) Structural — i.e. diffraction
2) Phonons — surface lattice vibrations
3) Atom-Surface Potential
4) Surface electron density profiles

Fig. 1
General Characteristics of Surface Scattering

Readily observed effects
1. diffraction
2. rainbow pattern
3. bound state resonance
4. threshold resonance
5. inelastic effects

Early references
Gerhard, Endermann + Stern, Fisch + Stern (1929-33)
Fisher, Bhara, Kolthau + Scott RGD (1968) p. 1227
Cabrera, Celli + Manson, PRB 32, 346 (1969) [CCM]
Fig. 2. Scattered intensity of He diffracted from (001) LiF; $\theta_i = 0^\circ$; (a) (110) and (b) (100) azimuth. The shaded region is the angular range covered by the bolometer box. The specular peak is extrapolated from measurements at small incident angles.

Fig. 3. Scattered intensity of He diffracted from (001) LiF; $\theta_i = 30^\circ$; (a) (110) and (b) (100) azimuth.
FIGURE 12: Spéculaire en fonction de l'angle d'incidence sur Cu(113) à $E_i = 21$ meV.
Inelastic Atom-Surface Scattering

$$E_i \rightarrow E_f = E_i \pm \hbar \omega$$

**Kinematics**

$$E_i = E_i \pm \hbar \omega$$  \text{energy}  \hspace{1cm}$$E_f = E_i \pm \xi + \xi$$  \text{parallel momentum}  \hspace{1cm}$$k = (K, k_z)$$

**Scan Curve**

$$\hbar \omega = E_i \left[ \frac{\sin^2 \theta_f}{\sin^2 \theta_i} \left( \frac{\Delta K}{k_i} + 1 \right)^2 - 1 \right]$$

$$\omega = \omega(K)$$

Fig. 6
Fig. 4: The interpretation of measured time-of-flight spectra is illustrated. The original measured spectrum shown at the top is transformed to an energy scale in the picture below. The locations of the maxima are interpreted with the aid of a scan curve (----) for 64.2° in an extended zone diagram. From the intersections of the energy loss peak locations with the scan curve the corresponding momentum transfers are determined.
Fig. 1: In part a) the scattering angles and coordinates used to describe phonons are defined. Part b) shows a calculated surface phonon dispersion curve \(^{14}\) for phonons with amplitudes along the \(z\)-direction and neutron (\(\bigcirc\)) and Helium atoms (\(\bigcirc\)) measurements for LiF along the \(\langle100\rangle\) direction. Part c) shows the corresponding density of states \(^{14}\). Note that \(1 \times 10^{13}\) rad/sec = 6.582 meV.
Dynamics of inelastic scattering

\[ W_i = \frac{\text{const}}{x^2} |\Psi_i|^2 \delta(\xi - \xi_i) \]

\[ T_i = \Psi_i + \sum \frac{\Psi_k}{\xi - \xi_k} e^{i\xi_k} T_{-i} \]

\[ \frac{dK}{d\xi d\Omega} = \sum \left< \frac{m(2m\xi_i)^n}{(2\pi)^{3/2}} \right> \left< \Psi_i \right> \]

Distorted wave Born approximation

\[ \frac{dK}{d\xi d\Omega} = \frac{m^2}{2\pi^3 \hbar^2} \sum \left| \frac{\langle \Psi_i | \langle \Psi | \rangle \rangle}{\omega(\theta)} \right|^2 \left| e_\theta(\beta) \right|^2 \]

\[ \times \left\{ \frac{\eta(\theta)}{\eta(\theta) + 1} \right\} \delta(\xi - \xi_k - \xi_i) \delta(\xi - \xi_i - \omega(\theta)) \]

where

\[ V(x, u) = U + V \approx U + v(x) + v(y) u_z + \ldots \]

\[ u = \sum \frac{\xi}{\sqrt{\hbar^2 \omega(\theta)}} (a_{q_+} + a_{q_-}) e^{i q \cdot \xi} \]

Resonant scattering — projection

\[ T_i = \Psi_i + \frac{h\xi}{\xi_i} \frac{h\xi_i}{\xi_i - \xi_i - h\xi} (\text{isolated resonances}) \]

\[ h\xi_i = \Psi_i + \sum \frac{\Psi_k}{\xi - \xi_k} e^{i\xi_k} h\xi_i \]

Fig. 9
FIG. 7. Same as Fig. 3 for \( \theta_i = 40.5^\circ \) and \( k_i = 6.10 \text{ Å}^{-1} \). D labels the diffused diffraction peak, due to the diffraction of atoms with incident velocities different from the nominal velocity.

IV. ANALYSIS OF TOF SPECTRA

In this section we compare the calculated one-phonon reflection coefficient with some selected TOF spectra. Out of the systematic comparison made for LiF(001) over a large number of different incidence angles and documented in Appendix F of Doak's thesis, we have chosen the more interesting and informative spectra. They are divided in two groups. The first group (Figs. 3–8) contains spectra which are not affected by resonances with bound states of the atom-surface potential and are in good agreement with theory. In the second group (Figs. 9–13) we collect some spectra showing clear effects of resonances as predicted from the kinematics and known bound-state energies. In each figure the kinematical parameters \( \theta_i \) and \( k_i \) and the surface temperature are indicated. The vertical lines correspond to \( \Delta K = G = (\text{integer}) \times 2\pi/a \), where \( a = 2.01 \text{ Å} \) is the nearest-neighbor ion distance. Since the experimental integrated intensity is strongly modulated by initial-state resonances—an effect not included in the theory—the spectra are plotted in arbitrary units.

A. Nonresonant inelastic spectra

Figures 3–6 show four spectra for equally spaced values of \( \theta_i (= 18^\circ, 36^\circ, 54^\circ, \text{and} 72^\circ) \). Inelastic scattering occurs essentially in two regions, corresponding to phonon-creation and -annihilation processes, which, with each new angle, shift to the left by approximately an integer \( G \) value. The relative intensities of the different structures change considerably with angle. At the extreme angle \( \theta_i = 18^\circ \) phonon-annihilation processes are not resolved from the background, while at the other extreme, \( \theta_i = 72^\circ \), only annihilation processes are observed.

At first glance, the calculated spectra appear to be in remarkable agreement with experiment and suggest that a complete interpretation of the TOF spectra should be possible. The sharper experimental peaks correspond quite

Fig. 10
FIG. 3. TOF spectra for He scattering from LiF(001) along (100) for $k_f = 6.06 \text{Å}^{-1}$ and $\theta_f = 63.2^\circ$, 64.2°, and 65.2°. The experimental data (a) are compared to the calculated nonresonant spectra (b) and to the results of the full calculation (c). The resonant factor alone is shown in (d) for two values of the surface temperature $T_s$. The plot for $T_s = 300 \text{K}$ (full line) is multiplied by 3. The effect of $T_s$ is included in the manner of Hornsby (Refs. 11 and 13), with the assumption of a root-mean-square surface displacement $0.0307 \text{Å}$. The maxima (marked by arrows in (a)) correspond to the inelastic resonances for $\hat{N} = (1,1)$ and $n = 1,2,3$ (as shown in (d)). Almost degenerate with the $n = 2$ peak, and affecting its shape, is an out-of-plane inelastic resonance for $\hat{N} = (2,0)$ and $n = 0$, which by itself gives a minimum.

$E_i + D$, where $D$ is the well depth; $\vec{G} + \vec{Q}$ stands for parallel momentum $\vec{K}_f + \vec{G} + \vec{Q}$ and energy $E_i + \hbar \omega + D$. The effect of adding $D$ to the energy is to change the perpendicular momentum from $k_z$ to $p = (k_z + \frac{4}{3} \hbar k^2 + 2mD/\hbar^2)^{1/2}$. The calculation of the $S$-matrix elements for a hard corrugated wall is described below.

The elastic and inelastic amplitudes $B(\vec{G})$ and $B(\vec{G} + \vec{Q})$ for scattering from $V_e + V_i$ are obtained by
Inelastic effects on Resonance lineshapes

\[ I_0 = | \frac{\sigma}{6i} - \frac{2\pi i m}{k^2 k_1 k_2} T_{gi} |^2 \]

\[ T_{gi} = h_{oi} + \frac{h_{cb} h_{bi}}{\epsilon_i - \epsilon_b - h_{bb}} \quad \text{(isolated resonance)} \]

\[ h_{pq} = v_{pq} + \sum_{r} v_{pr} \frac{1}{\epsilon_i - \epsilon_r + i \Gamma_r} h_{qr} \]

\[ I_0 = I_0^* |1 - \frac{ib}{\epsilon - i\Gamma}|^2 \]

\[ \epsilon = \frac{[\epsilon_i - \epsilon_b - \text{Re}(h_{bb})]}{\text{Im}(h_{bb})} \]

\[ b = \frac{N_0 h_{cb} h_{bi}}{[(\Delta_0 - i N_0 \epsilon_i) \text{Im}(h_{bb})]} \]

Optical potentials

Chao & Thompson (1976)
Wolf & Warren (1978)

Thermal attenuation (Delva-Wells, Hutchison & Celii (1980), Evans & Celii (1992), Montovani et al. (1983))
Mantovani et al. (1983)

He/Cu(113) (10) resonance
$E_i = 21\text{meV}$

Fig. 13
Thermal Attenuation

\[
I_c = I_c(T=0) e^{-\langle (\Delta k \cdot u) \rangle^2} = I(0) e^{-2\mathcal{W}} = I_0 e^{-\alpha T}
\]

Ref:
- Hand, W. G.
- Benedito & Garcia
- Armand & Manson
- Semiclassical
  - Levi; Levi & Subl
  - Mayer & Schinke & Gerber

Soft Potential + thermal attenuation

\[
I_c = |\delta_i^2| e^{-\frac{2\pi i m}{k_i k_e k_o} \langle \langle T_{gi} \rangle \rangle}^2
\]

\[
\langle \langle T_{gi} \rangle \rangle = \langle \langle v_{gi} \rangle \rangle + \langle \langle \sum_{\delta} \frac{1}{\varepsilon - \varepsilon_i + i\xi} v_{gi} \rangle \rangle + \ldots
\]

\[
\sqrt{z(u^2)} = D \left\{ \frac{e^{-2\lambda(z-u)}}{v_0} - 2e^{-z^2} \right\}
\]

He/Cu (100)

\[
D = 6.35 \text{meV}
\]
\[
\lambda = 1.05 \text{Å}^{-1}
\]
\[
u_0 (Armand)
\]

Fig. 14
\[ I(c^+) = I(\text{He}) e^{-2W_0} \text{ (T is crystal temperature)} \]

\[ W_0 = \frac{1}{2} \langle \Delta E \cdot n \rangle^2 \]

\[ \langle \Delta E \rangle \propto T \text{ to } T^3 \]

\[ \Delta E = \hbar \omega + k_T \]

\[ \text{He/Cu(110)} \]

63 meV \( \theta \approx 31.5^\circ \)

Fig. 15

Lapujilash & Poreeen (1982)
He/Cu (100) 21 meV = $E_i$

Fig. 16
Fig. 17

\[ \text{He/Cu (100)} \]

\[ \bar{e}_i = 63 \text{meV} \]

\[ I^*_o \]

\[ T(K) \]

\[ 100 \quad 300 \quad 500 \]

\[ 71.6^\circ \]

\[ 66.7^\circ \]

\[ 19^\circ \]

\[ 38^\circ \]

\[ 51^\circ \]
Fig. 18

He/Cu(100)

\( \theta_i = 73.5^\circ \)

\( E_i = 21 \text{ meV} \)

\( D = 6.35 \text{ meV} \)
Surface Spectroscopy Using Inelastic Scattering of Helium Atoms

R. B. Doak

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Surface Spectroscopy Using Inelastic Scattering of He Atoms

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ABSTRACT

A low energy (~20 meV) neutral helium atomic beam has been scattered from crystal surfaces. Energy gain and loss due to annihilation or creation, respectively, of surface phonons may be measured by time-of-flight analysis of the scattered beam. A temperature range is found in which single phonon scattering dominates, allowing the frequency and wave vector of individual surface phonons to be determined and their dispersion relations plotted. Resonant interaction with bound states of the helium in the surface potential well is found to greatly affect the inelastic scattering cross-sections.
The use of neutral atomic beams to probe crystal surface properties dates back to the pioneering work of the Hamburg group around 1930.\textsuperscript{1-3} As with other surface probes, however, atom scattering was only of limited interest prior to the modern development of ultra-high vacuum techniques which allow crystal surfaces to be cleaned \textit{in situ} and maintained clean for reasonable lengths of time. Atoms are unique among the surface probes in that they are completely nonpenetrating and nondestructive. As such, the scattering of light atoms may be employed to measure those static and dynamic properties of the two-dimensional surface which are measured in the three-dimensional bulk crystal by neutron scattering.

A diffraction pattern of helium atoms scattered from LiF\textsuperscript{4} is shown in Fig. 1. As discussed above by Mason, the angles and amplitudes of the diffraction maxima provide information on both the periodicity and corrugation of the surface. It was also shown how changes in diffraction intensities and lineshapes as a function of surface temperature could provide considerable insight into the dynamics of the crystal surface. This dynamical information may, of course, also be obtained directly by energy analyzing the
Figure 1

Helium Diffraction

$k_i = 10.95 \text{ Å}^{-1}$

Helium on LiF (001) (110)

scattered beam to determine the frequencies of the surface vibrational modes. It is this technique of energy-analyzed inelastic helium scattering which will be discussed here. The energy range of interest is that of the surface phonons, a few tens of meV.

Fig. 1 already hints at one of the difficulties to be encountered in inelastic helium scattering. As seen in that figure, it appears that virtually all of the coherently scattered intensity emerges in the diffraction peaks. This is somewhat deceptive, as low energy inelastic interactions also contribute an unknown fraction of these peaks. Furthermore, the amount of inelastic scattering is a strong function of crystal properties, beam mass and temperature, and scattering geometry. Nonetheless, it does turn out that the intensity of coherent single phonon scattering is several orders of magnitude lower than that of elastic (diffractive) scattering for typical experimental conditions. This necessitates careful design of the experimental apparatus to allow very low signal levels to be detected.

The technique used to velocity-analyse the scattered beam is illustrated in Fig. 2. This "time-of-flight" (TOF) analysis is well known in inelastic neutron scattering. The beam is mechanically pulsed (by a slotted rotating chopper disk in the present case) and the flight time of the beam pulses from chopper to detector is measured, allowing
Figure 2

Source  Chopper  Target

Time of Flight (TOF)
the velocity of the beam atoms to be determined. Those atoms which undergo inelastic interactions at the crystal surface to annihilate phonons (anti-Stokes scattering) or create phonons (Stokes scattering) will arrive at shorter or longer flight times, respectively, than elastically scattered particles. An example of some of the earliest TOF measurements\(^5\) is shown in Fig. 3. The dashed line is the TOF profile of the incident beam and the solid line curve the amplified spectrum of inelastically scattered He atoms. There appear to be features due to both phonon creation and annihilation. This figure also illustrates the second major problem to be overcome in measuring surface phonons, that of TOF (or energy) resolution. Ideally, the resolution should be such that the different inelastic events in the TOF spectrum do not overlap.

The resolution is determined primarily by the velocity distribution of the incident beam. As a series of recent measurements\(^6,7\) has shown, the controlling parameter is the product of source pressure \(p_0\) and nozzle diameter \(d\). This dependence is illustrated in Fig. 4, where speed ratio \(S_{\|}\) of the supersonic jet is plotted as a function of \(p_0d\). To a first approximation, the FWHM of the velocity distribution of the beam is given by \(1.65/S_{\|}\). In fact, for helium beams nature steps in to give a hand as well. The dashed lines show calculations based on classical cross-sections
Figure 3

He:LiF(001) – TOF Expts.

Fisher and Bledsoe, J.V.S.T. 9, 814 (1972)
PRODUCTION OF HELIUM NOZZLE BEAMS

Figure 4

@77 K, typ. 5 μ, 200 atm

Brusdeylins, Meyer, Toennies, and Winkelmann
RGD Symposium, 1976
whereas there is a quantum mechanical enhancement (solid line curves) which considerably improves the resolution. Speed ratios of 100–200 are possible for a LN$_2$-cooled beam source (He beam energy $\sim$18 meV), giving a velocity resolution of about 1% and energy resolution (twice as large) of a few tenths of an meV.

A series of experiments with this high resolution beam source have been performed at the Max-Planck-Institute in Göttingen, Germany. The crystal surfaces studied include a variety of alkali halides$^{8-14}$ as well as, more recently, metal$^{15,16}$ and semiconductor$^{17}$ surfaces. The work with alkali-halide surfaces is illustrative of the atom scattering technique and the present discussion will be limited to that particular example. The apparatus used in the early measurements$^{18}$ is shown in Fig. 5. Note that the angle between incoming and scattered beam is fixed at 90° so that as the incident angle $\theta_1$ is varied, the angle of the scattered beam ($\theta_f = 90^\circ - \theta_1$) varies as well. One important feature of this apparatus is the inclusion of numerous differential pumping sections to reduce the helium pressure from typically 1.5 x 10$^5$ torr in the source to a partial pressure of 5 x 10$^{-14}$ torr in the detector chamber. This low background He pressure is important to detect the very low scattered He intensity.
Figure 5

Magnetic Mass Spectrometer

Detector Chamber

Detector

Differential Pumping Stages

Target Chamber

 Incident Beam Detector

Time of Flight (TOF)
A typical diffraction pattern\textsuperscript{10} is shown in Fig. 6. In the lower half of the figure, the abscissa has been expanded to show the angular half-width of the apparatus, typically \( \approx 0.1^\circ \) for the specular beam. The first order diffraction peaks are broader -- about half of this increase is due to grating dispersion of the non-negligible (although small) beam velocity distribution. The remainder is a geometric factor related to the target rotation geometry described above.

The measurements taken at one of the diffraction angles of Fig. 6 yields the TOF spectrum shown in Fig. 7. Virtually all of the intensity in the spectrum is at the elastic flight time. (The tiny peak to the right of the major peak is due to the beam rebounding from the rear of the detector.) When the amplification is increased to search for intensity between the diffraction peaks of Fig. 6, the result\textsuperscript{10} is as shown in Fig. 8, where the vertical scale has now been expanded by about three orders of magnitude. Clearly there is not only inelastic intensity to be found, but it is highly structured, showing obvious maxima at some angles and minima at others. Various markings are shown in this figure which will be discussed below. For the moment, it is of interest to pick out one of these tiny features and see what insight a TOF trace taken at the angle has to offer.
Elastic TOF:

LIF (001) <100>

$\theta \equiv 23.4^{\circ}$

$K_i = 5.99 \text{ A}^{-1}$

$T_i = 298 \text{ K}$

$\frac{\Delta t}{t} \leq 1\%$

$\Delta E_{\text{FWHM}} \approx 300 \mu \text{V}$

Run No. 302.00

$L_{\text{ToF}} = 1.015 \text{ M}$

$6.90 \mu \text{Sec/Chnl}$

$0.008 \times 10^6 \text{ MCA Cycles}$
$k_i = 6.17 \, \text{Å}^{-1}$

$T_{LiF} = 300 \, \text{K}$

He - LiF (001) $\langle 100 \rangle$
Choosing the maximum at $\theta_i = 31.5^\circ$ produces the TOF trace shown in Fig. 9. Since this angle is well away from the nearest diffraction peak, we expect to see no elastic scattering and there is indeed no TOF peak at the elastic flight time. There are, however, four inelastic peaks — three due to phonon annihilation, one due to phonon creation. Are these due to single phonon interactions or are multiple phonons involved? This question may be answered by investigating the temperature dependence of the spectrum as illustrated in Fig. 10. Over a range of temperatures increasing from near LN$_2$ temperature to 570 K (LiF bulk melting temperature 1115 K) the inelastic TOF peaks are seen to first increase in height then decrease and, at the higher temperature, broaden considerably. This is the hallmark of multiphonon scattering; the broad diffuse tails and decreasing single phonon intensity. The low intensity at low temperatures merely reflects the decreasing thermal population of the vibrational modes. Since the intensities peak near room temperature where single phonon interactions clearly still dominate, most measurements were carried out at that temperature. (This optimum temperature will, of course, depend on the crystal properties.)

Once it is known that single phonon interactions dominate, a spectroscopic analysis to extract the phonon frequency and wave vector is straightforward, requiring only
Inelastic TOF
Figure 10

LIF (ODJ) <10C> VARYING TARGET TEMP.

\( \Theta = 31.6^\circ \)

\( k_1 = 6.2 \text{ A}^{-1} \)

\( T_\gamma = 133 \text{ K} \)

\( T_\gamma = 210 \text{ K} \)

\( T_\gamma = 303 \text{ K} \)

\( T_\gamma = 393 \text{ K} \)

\( T_\gamma = 476 \text{ K} \)

\( T_\gamma = 570 \text{ K} \)

Scattered Intensity (Adv. Units)

Time of Flight (MSEC)
conservation equations (Fig. 11). To facilitate identification of the participating modes, it is of use to combine these relations to yield the so-called "scan curves," a relation between phonon frequency and total parallel momentum transfer $\Delta K$ (Fig. 11). This provides a family of parabolic curves (corresponding to different incident angles), which show the locus of frequency and wave vectors consistent with conservation of energy and momentum. One such scan curve is shown in Fig. 12 aligned under a TOF trace replotted against frequency. Superimposed on the scan curves are the sinusoidal-like dispersion curves for Rayleigh phonons. At those frequencies where these dispersion curves intersect the scan curves, we would expect to see maxima in the TOF spectra. As can be seen, all but two of the TOF peaks (two and five) can be associated with annihilation or creation of Rayleigh phonons.

By repeating TOF measurements under different conditions\textsuperscript{12} (e.g., varying incident angle, Fig. 13) it is then possible to sweep out different portions of $\omega$ vs. $\Delta K$ space. If values of $\omega$ and $\Delta K$ are calculated for each TOF maximum and plotted in an extended zone dispersion plot, the results are as shown in Fig. 14. The sinusoidal-like curves are theoretical predictions of Rayleigh dispersion curves for LiF(001). As can be seen, these agree well with most of the experimental curves. The most notable exceptions are two
Calculation of phonon frequency and wavevector

1) Energy Cons. \[ \frac{\hbar^2 k_f}{2m} = \frac{\hbar^2 k_i}{2m} + \omega \]

2) Momentum Cons. \[ \mathbf{k}_f = \mathbf{k}_i + \mathbf{Q} + \mathbf{G} \]

3) TOT relation \[ k_f = \frac{m}{\hbar} \frac{l_{td}}{\tau_{td}} \]

\[ \frac{\omega}{\omega_i} = \left( \frac{m l_{td}}{\hbar k_i \tau_{td}} \right)^2 - 1 \]

\[ \frac{Q+G}{k_i} = \left( \frac{m l_{td}}{\hbar k_i \tau_{td}} \right) \cos \theta_i - \sin \theta_i \]

(in-plane scattering, \( \theta_i + \theta_f = 90^\circ \))

"Scan Curve"

Combine equations of energy and momentum conservation to eliminate \( k_f \):

\[ \omega = \omega_i \left[ 1 + \frac{4k}{k_i} \right]^2 \tan^2 \theta_i - 1 \]
Figure 12

LiF (001) <100>

\[ \theta = 64.2^\circ \]
\[ q = 0.041 \]
\[ T_2 = 295 K \]

Scattered Intensity (10^3 counts)

Time of flight (m sec)

Scaled Intensity (arb units)

Phonon Frequency \((10^3 \text{ rad sec}^{-1})\)

Parallel Momentum Transfer \((q^2)\)

Scan Curve 04.0 17.0
Figure 14

[Diagram showing a dispersion curve with labels A, B, F, and C, and wave vector directions.]
strange curves -- one near each of the (11) and (11) diffraction curves. These turn out to be experimental artifacts; in fact, the incident beam velocity distribution is not monoenergetic but has extended very low intensity tails \(10^{-3}\) of peak. Diffracton of these tails produces "energy-shifted" peaks near the diffraction peaks which are of the same amplitude as and may be misinterpreted as phonon signatures (Fig. 15). Peak 2 in Fig. 12 is such a diffraction tail; in view of the deceptive nature of these structures they were dubbed "deceptons," until overruled by the AIP (Fig. 16) which also refused to accept such lovely, imaginative terms as "phonyons" or "spurions" (a term employed in inelastic neutron scattering)! Plotting "dispersion relations" for the deceptons clears the unexplained data of Fig. 14 as shown in Fig. 17. This removes the asymmetry from the extended zone plot, allowing the data to be "folded" into the irreducible section of the first Brillouin zone. Reduced plots are shown for LiF(001) and NaF(001) in Fig. 18. The line curves are again theoretical predictions, which are seen to fit well the data for NaF but less satisfactorily for LiF. In the latter case there is a noticeable discrepancy at the zone boundary. This is of interest since the Rayleigh waves penetrate only about one wavelength so that at large wave vector (short wavelength) the penetration is but a couple of atomic layers, and it is truly the elastic characteristics of the outermost surface
Figure 15

The diagram illustrates a graph with the function $f(k_i)$ on the y-axis and $k_i$ on the x-axis. The graph shows a peak at $k_i^*$, with lower values at $k_i$. The diagram also includes labels for different regions: $(\overline{1}1)$, $(00)$, and $(11)$. The terms "Decepton", $k_i^*$: false premise, and false assumption are indicated. The dashed lines represent the actual situation, while the solid line represents the false assumption.
August 2, 1983

Dr. J.P. Toennies
Max-Planck Institut
Stromungsforschung
Bottingerstrasse 4-8
3400 Gottingen, W. Germany
West Germany

RE: "Measurement of the Rayleigh surface-phonon dispersion curve for ..., code No. BD2363 scheduled for publication in Physical Review D.

Dear Dr. Toennies:

The article noted above is being published in the August 15 issue of Physical Review B, Vol. 28, issue No. 4, pp. 2104-2113.

This letter is to inform you that the editor will not accept the terms "deception," "phonyon," or "D spurion" in future articles.

The editor permitted "D spurion" to appear this time only because your article was in the later stages of production.

To repeat the "note to author" that was originally affixed to the manuscript, these terms are simply not acceptable; their usage is not justified. Please use more conventional, standard terminology in the future.

Yours sincerely,

Valerie L. Miller
Journal Supervisor
Physical Review B

cc: Dr. Peter Adams
LIF(001) [100] SURFACE PHONON DISPERSION CURVE
EXTENDED ZONE FLOT OF EXPT. MEASUREMENTS

PHONON FREQUENCY [10^13 RAD/SEC] -4.0 -3.0 -2.0 -1.0 0.0 1.0 2.0 3.0 4.0 5.0
WAVE VECTOR DELTA K [10^10 1/M] -4.0 -3.0 -2.0 -1.0 0.0 1.0 2.0 3.0 4.0

DECEPTIONS

θ_i 65.2 66.6 67.4 68.7 69.0 0.95 0.90 0.85
k_i / k_i^* 1.00 1.05 1.10 1.15

21.2 19.8 0.95 0.90 0.85

DECEPTIONS

25.8 24.8 23.4 22.6 21.2 1.00 1.05 1.10
Figure 18

LiF (001) <100>
Data from Doak, Toennies, et al.

- Chan, et al.
- Benedek, et al.

NaF (001) <100>
Data from Doak, Toennies, et al.

- Chan, et al.
- Benedek, et al.
which are being sampled. The experimental data here lie below the predicted values which is of particular interest since alkali-halide surfaces are believed to relax inward which would lead to a stiffer surface and higher frequencies relative to the theoretical curves for the unrelaxed surface.\textsuperscript{21} Thus, surface relaxation \textit{per se} is probably not the cause of the discrepancy. Other possible explanations are being investigated.

Returning to the angular distribution\textsuperscript{10} (Fig. 19), it can be said with certainty that all of the intensity away from the diffraction angles is due to single phonon inelastic scattering. Proceeding beyond the simple spectroscopic analysis (which requires only kinematic information) it is of interest to consider the information to be obtained from the amplitudes and shapes of the TOF peaks. The many minima and maxima in the angular distribution must arise through either enhancement of the phonon density of states or of the scattering cross-section. The former is expected to be slowly varying apart from tangency points of scan curves with the dispersion curves (at which the interaction with a given mode abruptly "turns on" or "turns off"). This effect, termed kinematic focussing by Benedek,\textsuperscript{22} is illustrated in Fig. 20. Considering the highest frequency annihilation events, it is seen that two peaks at $\Theta_i = 30.7^\circ$ should merge to form a single peak at $32.7^\circ$ which $\ldots$
Figure 19

He - LiF (001) <100>

$I/I_{(00)} \times 10^3$

$\Theta_1 [\text{degrees}]$

n = 32

(11)

(0,0)

(10)

(11)
entirely at larger $\theta_i$. A weak singularity is expected at the tangency point\textsuperscript{11} which could account for some of the features in Fig. 17, where the kinematic focusing is marked with regard to angle and approximate form by the shaded shapes at the bottom of the plot. However, if the TOF spectra are recorded at these above angles, as in Fig. 21, the two phonon peaks are seen to merge then disappear, as expected; but at the kinematic focusing angle the enhancement appears in all of the phonon peaks and not just that undergoing kinematic focusing. Thus, an additional factor must be entering here.

It is well known through elastic He scattering\textsuperscript{23} from surfaces that there is an attractive potential for He at the LiF surface and that four bound states exist in that potential well (Fig. 22). Resonant entrance into these states can dramatically change the elastic scattering cross-sections. As shown in Fig. 23, this bound state resonance or "selective adsorption" places the He atom into a two-dimensional Bloch state at the surface -- a quantum mechanical analogy to the classical "hopping" trajectory. The entrance into and exit from the bound state may occur either elastically (diffraction) or inelastically (phonon mediated). The energies of the four bound states are well known, allowing the incident angle for resonance to be calculated for any given scattering event. The incident angles
Figure 21

a) $\theta_i = 30.7^\circ$
$k_i = 5.97 \, \text{A}^{-1}$

b) $\theta_i = 32.7^\circ$
$k_i = 5.99 \, \text{A}^{-1}$

c) $\theta_i = 34.7^\circ$
$k_i = 5.99 \, \text{A}^{-1}$

Scattered Intensity [arb. units] vs. Time of Flight [msec]

- annihilation
- creation

[Graph showing data and annotations]
Bound states of He on LiF(001)

Lilienkamp and Toennies,
Figure 23

\[ V_{oo}(z) \text{ [meV]} \]

\[ z \text{ [Å]} \]

\[ \epsilon_0, \epsilon_1, \epsilon_2, \epsilon_3 \]

He:LiF(001)
corresponding to elastic transition into a bound state are shown in Fig. 19 for two different reciprocal lattice vectors. Considering first the (11) selective adsorption, it is seen that there is very good agreement between these angles and the position of maxima in the angular distribution. Since it is known from the TOF spectra that all of this intensity involves interaction with a phonon, it would appear that the helium atom has been elastically selectively adsorbed and then inelastically desorbed. The resonant interaction with the bound state enhances the amplitude at the corresponding angles. For selective adsorption via the (10) reciprocal lattice vector, the situation appears to be somewhat different. Here the resonance angles appear to correspond to minima in intensity. (Although not obvious the correspondence in angles is, in fact, exact as shown by recent, more careful measurements.)

A perhaps too heuristic argument for this observation may be based on the fact that the (11) reciprocal lattice vector lies in-plane whereas the (10) lies out of plane. Diffraction into a bound state via the (10) vector thus channels intensity into an out-of-plane direction from which a return to the in-plane direction requires interaction with a very large wave vector phonon -- an unlikely event.
Resonant events explain many, but not all, of the features seen in the angular distribution. In particular, the many maxima and minima near the specular peak do not seem to correlate with selective adsorption events. The maxima which indeed may be assigned to bound state resonances allow an additional piece of information to be extracted. The angular width of the feature should carry information on the lifetime of the bound state. There may be additional factors contributing to the peak width so, in fact, only a lower limit may be calculated. Deconvoluting the known angular resolution of the apparatus from the peak width and making use of the Uncertainty Principle yields the lower limits on lifetime shown in Fig. 24 (the additional kinetic energy of the atom in the bound state has been included in distance traveled L). The higher states appear to be longer-lived, presumably because the atom spends less time close to the surface, as seen in the probability distributions of Fig. 23.

To fully utilize the intensity information of the TOF spectra, it is necessary to develop a comprehensive scattering theory which includes a detailed calculation of the phonon density of states. Benedek has undertaken such a scattering calculation based on a distorted wave Born approximation (DWBA) which treats the phonon-induced changes in static surface corrugation as a perturbation. Comparison
Figure 24

Lifetimes of resonantly bound He atoms on LiF(001)

<table>
<thead>
<tr>
<th>n</th>
<th>$\Delta \theta$ (°)</th>
<th>$r$ ($10^{-12}$ sec)</th>
<th>$L$ (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>1.0</td>
<td>7</td>
<td>80</td>
</tr>
<tr>
<td>1</td>
<td>0.6</td>
<td>11</td>
<td>120</td>
</tr>
<tr>
<td>2</td>
<td>0.4</td>
<td>17</td>
<td>170</td>
</tr>
<tr>
<td>3</td>
<td>0.12</td>
<td>60</td>
<td>560</td>
</tr>
</tbody>
</table>
of an experimental TOF spectra with a theoretical calculation (Fig. 25) shows reasonably good agreement. The solid diamonds mark the Rayleigh modes; the regions where the scan curve cuts through the bulk bands produce broad maxima. The dashed line in the upper trace shows the envelope expected from the Bose-Einstein thermal population. This clearly produces the long, decreasing tails at high energy ($\Delta K$).

Fig. 26 shows another TOF spectrum which does not agree so well with theoretical predictions. One small maxima between the two leftmost Rayleigh peaks is not reproduced in the theoretical spectrum. This small peak is apparently again a signature of selective adsorption, although it is now the time reversed sequence of the previous selective adsorption process which enters. The atom is inelastically selectively adsorbed and elastically desorbed. The kinematics of the process may be visualized by superposing the requirements of the inelastic selective adsorption on the plot of scan curves and phonon dispersion curves. This is done in the lower part of Fig. 26 for the four different bound states. An intersection of bound state curve (solid line) with dispersion curve (dot-dashed line) and scan curve (dashed line) gives the $\omega$ and $\Delta K$ of such an inelastic selective adsorption/elastic desorption event. For $n = 0, 2, \text{and } 3$ the intersections of scan and bound state
**Figure 25**

LiF(001) <100>

- $\theta_1 = 72.2^\circ$
- $k_1 = 6.15$ Å
- $T_0 = 297$ K

**LiF(001) <100>**

- $\theta_1 = 36.0^\circ$
- $k_1 = 6.10$ Å
- $T_0 = 297$ K

G. BENEDEK, J. P. TOENNIES, AND R. B. DOAK

Figure 26


Physical Review B 28, 7277, 1983
curves lie outside the regions of phonon dispersion curves. For the \( n = 1 \) state, however, the two cross within the bulk band, and it is the enhancement of scattering amplitude of bulk band phonons which gives rise to the small peak in the upper trace. The various modes of interaction of the He with the surface are shown schematically in Fig. 27.

In conclusion, it appears the helium scattering is a very useful tool for obtaining information about surface vibrations. It has higher resolution and is more surface sensitive than electron scattering and is particularly well suited to studies of acoustic modes in the dispersive regime (although optical modes may also be measured). Inelastic helium scattering provides extensive information on the helium-surface interaction. Finally, the above comparisons show that theoretical modeling, even in the present rudimentary state, is already capable of making quantitative predictions to aid in the data interpretation.
Beam/Surface Interactions

(a) ELASTIC SCATTERING (DIFFRACTION)

(b) INELASTIC SCATTERING

(c) ELASTIC SELECTIVE ADSORPTION
    ELASTIC DESORPTION

(d) INELASTIC SELECTIVE ADSORPTION
    ELASTIC DESORPTION

(e) ELASTIC SELECTIVE ADSORPTION
    INELASTIC DESORPTION
REFERENCES


The $Z^3_1$ (Barkas) Effect

George Basbas

Physical Review Letters
Ridge, New York
**BARKAS EFFECT:**

\[
\frac{\text{Range}(\Sigma^-)}{\text{Range}(\Sigma^+)} = 1.03 \quad \text{at } v = 0.14c
\]

\[
\frac{\Delta E(\pi^+)}{\Delta E(\pi^-)} = 1.14 \quad \text{at } v = 0.05c
\]

(1963) Barkas, Dyer & Heckman

(1969) Heckman & Lindstrom

\[
\text{DATA } \Rightarrow \alpha z_i^2 + \beta z_i^3 \quad \text{for STOPPING POWER}
\]

\[
\uparrow \Rightarrow \beta > 0
\]

\((z_i \text{ carries sign of charge})\)
\[ \frac{\Delta E}{\Delta x} \text{ MEASUREMENT} \]

**Liquid Helium**

- **Resistance Thermometer**
- **Foil (}\Delta x\)**
- **Block**
- **Eo - \Delta E**
- **Eo**
- **Beam**

**Conventional Method**

1. **Measure Eo and Eo - \Delta E**.
2. **Need \Delta x for small target area**.

\[ \Delta E_{\text{difference}} \]
\[ \Delta x_{\text{nonuniform}} \]

**Calorimetric Method**

1. **Measure \Delta E directly** (±0.1%).
2. **Large beam diameter (≈1cm²) averages over thickness inhomogeneities**.

\[ \Delta E_{\text{direct}} \]
\[ \Delta x_{\text{average}} \]

**Uncertainties:**

- \( E_0 \)
- **Slt Scattering**
$w \ll 1$, the function $F(w)$ [Eq. (14)] depends only logarithmically on $w$ (cf. Fig. 3). That is, in the
limit of high-particle velocities our $Z_1^2$ correction becomes insensitive, as it should, to the choice of $\theta$.

We have made a calculation of the difference in stopping power due to the $Z_1^2$ effect between $Z_1 = 1$ and $Z_1 = 1$ particles in emulsion and find a $9-95\%$ difference at $1.2 \text{MeV/amu}$, in fair agreement with

\[ \frac{S_{\text{He}} - 4S_{\text{H}}}{S_{\text{He}}} \times 10^2 \]


\[ \frac{S_{\text{He}} - 4S_{\text{H}}}{S_{\text{He}}} \times 10^2 \]


\[ \frac{S_{\text{He}} - 4S_{\text{H}}}{S_{\text{He}}} \times 10^2 \]
\[ F = \frac{m}{r} a \]

\[ \ddot{r} + \omega^2 r = \frac{1}{m} \mathbf{f}(\mathbf{r}, \mathbf{v}) \]

\[ \dot{r}_0 + \omega^2 r_0 = \frac{1}{m} \mathbf{f}(\mathbf{r}, \mathbf{v}) = 2 \mathbf{f}(\mathbf{r}, \mathbf{v}) \]

\[ r_0 \propto z_1 \]

\[ \Delta E \propto z_1^2 \]

\[ \ddot{r}_1 + \omega^2 r_1 = \frac{1}{m} \mathbf{f}(\mathbf{r}, \mathbf{v}) \]

\[ \text{"polarization"} \]

\[ S = \frac{e^2}{\hbar} \int_{b}^{0} \Delta E(b) \, db \]

\[ = 2 \pi \int_{b}^{0} \Delta E_2(b) \, db + 2 \pi \int_{b_c}^{0} \Delta E_3(b) \, db \]

\[ \left. \frac{e^2}{\hbar} \frac{2m^2 v^2}{I} + \frac{z_1}{3} \right\} \]

\[ L_1 = \frac{e^2 \omega}{m v^2} \ln \left( \frac{w b_c}{v} \right) \sim \frac{3}{2} \pi \frac{e^2 \omega}{m v^2} \ln \left( \frac{v}{w b_c} \right) \]
\[-\frac{1}{n} \frac{dE}{dx} = S = \frac{4\pi z_1^2 e^4}{m v^2} z_2 L\]

\(z_2\) is TARGET ATOMIC NUMBER

\[L = L_0 + z_1 L_1 + z_1^2 L_2 + \ldots\]

Born Series Expansion

\[L_0 = \ln \frac{2mv^2}{I} - \frac{C(v)}{z_2}, \quad \text{NONRELATIVISTIC}\]

\(I = \text{MEAN EXCITATION ENERGY OF TARGET}\)

\(C(v) = \text{INNER SHELL CORRECTION}\)

Corrects for use of dipole approximation which is valid for \(v \gg v_{\text{shell}}\).

ASHLEY-RITCHIE-BRANDT:

\[L_1 = \frac{3}{2} \pi \frac{e^2 \omega}{mv^3} \ln \left( \frac{v}{\omega b_c} \right) \quad \text{for } v \gg \omega b_c\]

Bloch Term

\[L_2 \sim -1.2 \left( \frac{v_0}{v} \right)^2 \quad \text{for } v \gg v_0\]
FIG. 7. Experimental curves (fully drawn) for the Bethe logarithm ($L_0$), the $Z_i^1$ factor ($L_1$), and the $Z_i^2$ factor ($L_2$) for four target materials. Theoretical curves for $L_1$ (dashed) and $L_2$ (dot and dash) are Eqs. (7) and (6), respectively.

$L_0, L_1, L_2$ ——— DATA

$L_1$ ——— Ashley, Ritenze & Brandt $Z_i^3$ Term

with $b_c = $ QM Harmonic Osc. Radius

(Jackson & McCarthy)

$L_2$ ——— Bloch Term

**STATUS OF BARKAS EFFECT**

Barkas established that heavy particles with opposite charges lose energy at different rates and

1. If this is due to a $Z_i^3$ term in the stopping power, its coefficient is positive.

H.H. Andersen & collaborators established the departure from the $Z_i^2$ scaling law of the Bethe formula for positive ions with different atomic numbers.

And

2. If this is due to a $Z_i^3$ term its coefficient is positive.

Also: 3. From a three-parameter fit of a polynomial in $Z_i$ to the data, values for $L_0$, $L_1$, and $L_2$ are inferred.

**Theory:**

- 1. Distant collision contribution.
- 2. Impact-parameter cutoff for dist. coll.
- 3. Close collision contribution.

**Remarks:**

1. Because of the freedom of the impact-parameter cutoff, calculations with and without contributions from close collisions both match the data.

2. Stopping power data are incapable of distinguishing between the two theoretical approaches.

3. Measurements of single-electron inner-shell processes are needed to provide more detailed information.
### Calculations of the $Z^3$ Term for Energy Loss

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<th>&quot;Impact Parameter&quot; Dependence</th>
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<td>$b_c = \frac{\hbar}{2m\nu}$</td>
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<td>Hill &amp; Merzbacher PRA 9, 156 (1974)</td>
<td>Reproduces classical HO result, importance of close collisions cannot be taken for granted</td>
<td>QM Time-Dep. Perturbation Th.</td>
<td>QM Harm. Osc.</td>
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<tr>
<td>Morgan &amp; Sung PRA 20, 818 (1979)</td>
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<td>2nd PWBA Closure &amp; Dipole Approx.</td>
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<td>Sung &amp; Ritchie PRA 28, 674 (1983)</td>
<td>Unimportant</td>
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**Reviews & Surveys**

Angular and Thickness Dependence of the Specific Energy Loss of $(0.6 - 1.0) \nu_0$ Heavy Ions in Carbon


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ANGULAR AND THICKNESS DEPENDENCE OF THE SPECIFIC ENERGY LOSS
OF (0.6 - 1.0)v_o HEAVY IONS IN CARBON


Atomic Energy of Canada Limited Research Company

Chalk River Nuclear Laboratories

Chalk River, Ontario, Canada K0J 1J0

ABSTRACT

Specific energy losses have been measured for heavy ions at low velocities (0.6 ≤ v/v_o ≤ 1) in carbon with a time-of-flight (TOF) spectrometer. From the measured angle and thickness dependence, we find no evidence for pre-equilibrium stopping effects.

* On leave from GSI, Darmstadt, West Germany.
When a heavy ion beam penetrates through matter, the ions lose energy and change their original direction of motion through collisions with the target atoms. Generally the stopping processes are described by an electronic interaction arising from the electronic excitation of atomic or collective modes and by nuclear elastic collisions with the target atoms. The last mechanism is mainly responsible for the angular scattering, whereas its contribution to the stopping of heavy ions is only significant at low velocities. In the first approximation the two slowing down processes can be treated as independent events [1,2]. The validity of this basic assumption has been studied recently in new experiments [3,4,5] and theoretical investigations [6,7,8] treating simultaneously the energy loss and angular distributions of ions traversing the stopping medium.

In this paper we demonstrate that the geometrical arrangement of the detection system can lead to a totally different interpretation when studying the thickness dependence of heavy ion energy loss. A schematic of the experimental set-up is shown in fig. 1. A low intensity, collimated beam from the CRNL High Voltage Mass Separator (HVMS) is used to measure the angular and thickness dependence of heavy ion energy losses in thin foils. The energy losses were measured with a TOF spectrometer in transmission geometry with and without the stopping foil in front of the detector system. The two TOF detectors consist of thin carbon foils positioned perpendicular to the beam direction separated by a distance of \( \sim 83 \) cm. The secondary electrons released when the ions penetrate through the detector foils were electrostatically accelerated (1.6 kV) and magnetically deflected into microchannel plate assemblies [9]. These new detectors and the measurement
in the direct beam from the HVMS have achieved significant improvements in the time resolution compared with our earlier experimental set-up [10]. Furthermore, the new TOF spectrometer can be continuously rotated in the angular range, -10° to 37° relative to the incident primary beam direction (0°), with the axis of rotation lying in the target plane. The detector system could be positioned reproducibly with a precision better than 0.05° and had an acceptance angle of 0.17° in the horizontal plane, subtending a total solid angle of 0.03 msr. The absolute flight times were determined by: 1) varying the distance between start and stop detectors by a precisely measured amount (16 cm), and 2) by the electronic method described earlier [11]. The two methods yielded agreement within 0.1%. The detection efficiency for each detector was >96%. The thicknesses of the target foils were determined by measuring the energy loss of ~620 keV $^4$He ions in the same geometry and using the stopping power tables of Ziegler [12].

After we had observed the channeling disturbance in 0° measurements using different evaporated polycrystalline foils, similar to results described by Mertens et al. [13] and Schultz [14], we concluded that only carbon foils were suitable amorphous monoatomic stopping solids for our investigations. For example, in fig. 2 we show the corresponding TOF spectra obtained with 403 keV Mg ions in 19 μg/cm² evaporated Al foils. Part (b) of this figure demonstrates clearly the convolution of the channeled and randomly scattered beam in the 0° measurement, whereas at an emergent angle $\alpha=4.8^\circ$ only the random component determines the shape of the distribution. The critical angle for axial channeling in this case is ~3°.
Fig. 3 shows the measured thickness dependence of the specific energy loss for $^{20}$Ne in carbon. For these data $\alpha$ was $0^\circ$ and the mean velocity of the projectiles in the different foils was exactly $0.8 \, v/v_0$, which was achieved by tuning the magnetic field of the HVMS. The results demonstrate clearly the increase of the specific energy loss with increasing foil thickness, in agreement with our earlier data [5]. This characteristic feature was observed for all heavy ion energy loss data. The effect is more pronounced for heavier projectiles where the thickest target foil ($\sim 29 \, \mu g/cm^2$) was not thick enough to cause the saturation apparent for the Ne data. For example, in fig. 4, we present similar results for $^{209}Bi$ ions at $0.8 \, v_0$ in the same stopping foils.

In fig. 5 we show our measured energy losses for $0.8 \, v_0$ Ne ions in three different carbon foils measured at different emergent angles, $\alpha$. We observe that the relative change at fixed $\alpha$ is largest for the thinnest foil. The enlargements of the path length give a negligible contribution to the measured increase in stopping at the different angles. Using the data from fig. 5, we show in fig. 6 the corresponding specific energy loss for different emergent angles as a function of the target thickness. With increasing $\alpha$, the specific energy loss is decreasing with increasing target thickness, contrary to the $0^\circ$ measurement. We explain qualitatively our results by the increasing contribution of violent collisions (small impact parameters) with increasing foil thickness due to multiple scattering into the acceptance angle of the TOF spectrometer. Our results are qualitatively in agreement with Högberg [15], who measured the thickness dependence in C at lower velocities (4.5 to 46 keV) and with Skoog et al. [16]; we are not
in agreement with the results of Mertens and Krist [17], and Reid and Scanlon [18]. The latter groups found an increased stopping for their thinner foils. Mertens and Krist used carbon foils in the same thickness range as used here. They observed a decreasing specific energy loss with increasing thickness of the carbon foils and suggested a new model of pre-equilibrium stopping to explain their results.

Our combination of measured angle and thickness dependence is a possible explanation of the results of Mertens and Krist [15], who measured at a fixed emergent angle, \( \alpha \neq 0^\circ \), to avoid the influence of the channelled beam. Their detection angles were chosen in the range \( 3^\circ \leq \alpha \leq 6^\circ \), thereby simultaneously circumventing the high intensity problem in the initial beam direction.

From our systematic investigations we conclude there is no significant enhanced pre-equilibrium stopping as Mertens and Krist have suggested.

We are grateful for the technical assistance of G.A. Sims during the course of these measurements.
REFERENCES

REFERENCES (continued)


ANALYSING MAGNET

SLIT COLLIMATOR (X-Y)

SLIT COLLIMATOR (X-Y)

TEST CHAMBER & MONITOR DETECTOR

SLIT COLLIMATOR (X-Y)

TARGET

T.O.F. CHAMBER.

Fig. 1
403 keV $^{24}\text{Mg} \rightarrow \text{Al}$

a) NO TARGET, $\alpha = 0^\circ$

b) Al-TARGET, $\alpha = 0^\circ$

c) Al-TARGET, $\alpha = 4.8^\circ$

Fig. 2
$^{20}\text{Ne} \rightarrow \text{C}$

$v = 0.800 v_0, \alpha = 0^\circ$

$\frac{\Delta E}{\Delta X}$ (keV cm$^2$ µg$^{-1}$)

$\Delta X$ (µg/cm$^2$)

Fig. 3
\[ v = 0.80 \, v_0, \alpha = 0^\circ \]

Fig. 4
Fig. 5

\begin{equation}
\frac{\Delta E(\alpha) - \Delta E(0^\circ)}{E(0^\circ)} \times 1.25
\end{equation}

\begin{align*}
\text{20}^\text{Ne} & \rightarrow \text{C} \\
v & = 0.80 \, v_0
\end{align*}

\begin{align*}
\Delta \tau & = 5.764 \\
\times \tau & = 3.910 \\
\bullet \tau & = 1.964
\end{align*}
Relativistic Corrections to Stopping Powers

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RELATIVISTIC CORRECTIONS TO STOPPING POWERS*

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Abstract

Relativistic corrections to the nonrelativistic Bethe-Bloch formula for the stopping power of matter for charged particles are traditionally computed by considering close collisions separately from distant collisions. The close collision contribution is further divided into the Mott correction, appropriate for very small impact parameters, and the Bloch correction, computed for larger values. This division of the region of close collisions leads to a very cumbersome result if one generalizes the original Bloch procedure to relativistic energies. We avoid the resulting poorly specified scattering angle $\theta_0$ that divides the Mott and Bloch correction regimes by using the procedure suggested by Lindhard and applied by Golovchenko, Cox and Goland to determine the Bloch correction for relativistic velocities.


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The energy loss of swift charged particles in matter has been studied extensively for many years because of its importance for nuclear physics. The landmark classical theory of Bohr, the pioneering non-relativistic quantal contribution of Bethe, and the synthesis of Bloch have given firm theoretical foundation for many subsequent developments. Bethe extended his treatment to include relativistic corrections, using the Møller cross section for electron-electron collisions. Following this, Fermi's dielectric approach has led to understanding of relativistic corrections originating from large impact parameter collisions.

The theory of stopping up to that time had employed perturbation theory in which the stopping power for a particle with speed \(v\) and charge number \(Z\) \(\ll \frac{\hbar v}{e^2}\) is proportional to \(Z^2\). Contributions that involve other dependences on \(Z\) have since been studied theoretically and confirmed experimentally.

A) Ashley, Ritchie and Brandt (ARB) extended stopping power theory to include the polarization of target atoms by the projectile. The result was a correction term that, in lowest order, is proportional to \(Z^3\). The results of this development were confirmed in a quantal harmonic oscillator approximation that leads to identical formulas. The theory of ARB contains a scaled minimum impact parameter that they denoted by \(b\). It is of order unity and is essentially independent of the atomic number of the stopping material. Adjusting on relative stopping powers of \(^{13}\text{Al}\) and \(^{73}\text{Ta}\) for \(^1\text{H}\) and \(^4\text{He}\) yielded a first trial value \(b = 1.8\) Å. A later analysis using other data indicated that \(b = 1.4\) Å is a better value. Still more recently, following a suggestion by Brandt, Ishiwari and
co-workers\textsuperscript{(13)} have fitted some of their own data, allowing both $b$ and another quantity $\chi$ to be adjustable parameters.\textsuperscript{(14)} They find $b = 1.3\text{Å}$ and $\chi = 1.358$ to be good values. Other models set $b$ from the harmonic oscillator model,\textsuperscript{(15)} while Lindhard\textsuperscript{(16)} and Esbensen\textsuperscript{(17)} have chosen $b$ corresponding to the quantal minimum impact parameter, arguing that approximately equal contributions to the $Z_1^3$ correction term originate from small and from large impact parameters. More recently some of us\textsuperscript{(18)} have carried out a many-body analysis of the electron gas model and conclude in agreement with the original assumption of ARB that the principal contribution to the $Z_1^3$ term originates from the region of large impact parameters. Recent data on energy losses of heavy ions have been obtained by Feissel et al.\textsuperscript{(19)} and analyzed by them to obtain effective charges. They conclude that their data are better described in terms of the model of ARB\textsuperscript{(9)} with the choice of the minimum impact parameter made by Jackson and McCarthy\textsuperscript{(16)} than by the Lindhard theory.\textsuperscript{(15)}

B) At relativistic velocities, deviations from the Rutherford cross section give rise to contributions from small impact parameters\textsuperscript{(20,21)} that are the higher order in $Z$. These are referred to as Mott corrections.

C) The Bloch correction\textsuperscript{(4)} contributes terms of order $(Z_1)^{2n}$ (where $n = 2, 3, \ldots$) to the stopping power formula. This may be considered to arise from the fact that the minimum impact parameter must be chosen from quantal considerations at large $v$ ($b_{\text{min}}^Q \approx \frac{\sqrt{2} Z}{2mv}$) but from the Bohr classical argument, as $v$ becomes small ($b_{\text{min}}^C \approx |Z_1|e^2/m v^2$).

The purpose of this paper is to consider in detail the Bloch and Mott corrections to the standard formulas for stopping power. Eby and Morgan\textsuperscript{(20)} and Ahlen\textsuperscript{(21)} extended the original work of Bloch\textsuperscript{(4)} into the relativistic region ($v \approx c$). These theories use a center-of-mass
scattering angle $\theta_0$ originating in the Bloch approach. It is defined by the inequalities

$$1 \gg \theta_0 \gg \frac{\alpha}{\beta \gamma Z_1}$$

(1)

where $\alpha = Z_1 e^2 / \hbar c$ is $Z_1$ times the fine structure constant, $\beta = v/c$, and $\gamma = (1 - \beta^2)^{-1/2}$. It is introduced to divide the small impact parameter region into two parts: i) for $\theta < \theta_0$, the struck electron is described in the plane wave approximation; and ii) for $\theta > \theta_0$, the full (Mott) wave function is used. Since $\theta_0$ is not defined, this procedure leads to uncertainty.

An alternative to the Bloch procedure, which avoids the angle $\theta_0$ altogether, is to use the full Mott wave function over the entire range $\theta$. Golovchenko, Cox and Goland, following a suggestion by Lindhard, have shown that one may obtain the nonrelativistic Bloch correction in a few steps by working in center-of-mass coordinates. In this framework the struck electron is considered to be effectively free for angular momenta that are not too large. Thus the wave function of the electron in the field of the onrushing ion may be taken to be the exactly known Mott function. One has only to compute the energy transferred from the ion to the electron at a given CM scattering angle, multiply by the differential scattering cross section and the electron density to obtain the energy loss per unit path length. The result is then expressible in terms of a sum over phase shifts for various angular momentum quantum numbers and may be evaluated to yield, exactly, the Bloch correction, as Golovchenko, Cox and Goland have shown.

Here we extend this procedure into the relativistic region. We find a great simplification over the Bloch procedure as implemented by Ahlen.
in the relativistic region, since it is not necessary to introduce the quantity $\hat{a}_0 = \frac{m^2}{k^2}$ to divide impact parameter space into two regions in which different approximations to the collision dynamics are employed.

II. Theory

The energy loss per unit length, $dE/dx$, is taken to be proportional to the density of electrons in a medium in which the atomic density is $N$ and the atomic number of each atom is $Z_2$. Thus

$$\frac{dE}{dx} = N Z_2 \int dE(\theta) \frac{d\sigma(\theta)}{d\Omega} d\Omega,$$  (2)

where $d\sigma(\theta)/d\Omega$ is the differential scattering cross section for scattering through a CM angle $\theta$ of a free electron in the Coulomb field of the swift ion, and $dE(\theta)$ is the energy transferred to the struck electron in the laboratory system. We assume that the full charge of the swift ion is to be employed in this theory. From the work of Mott

$$\frac{d\sigma}{d\Omega} = \frac{i}{4k^2} \left| \sum \left\{ (2l + 1) \left( e^{2i\delta_{l+1}} - 1 \right) + \frac{2i\delta_{l+1}}{l \left( e^{2i\delta_{l+1}} - 1 \right) - 2l} \right\} \hat{P}_l(\theta) \right|^2 \left| \hat{P}_l(\theta) \right|^2 / 4k^2$$  (3)

The phase shifts $\delta_{l+1}$ and $\delta_{l-1}$ are given by

$$e^{2i\delta_{l+1}} = -\frac{i+1}{l+1} \frac{e^{-ir(\rho_{l+1}+1)}}{\Gamma(\rho_{l+1}-i\alpha/\rho)} \frac{\Gamma(\rho_{l+1}+i\alpha/\rho)}{\Gamma(\rho_{l+1}+1)} e^{-ir(\rho_{l+1}-1)}$$  (4a)
\[ e^{\frac{2i\xi}{\xi - 1}} = \frac{\xi - 1}{\xi + 1} \frac{\Gamma(\xi + 1 - i\alpha/\beta)}{\Gamma(\xi - 1 + i\alpha/\beta)} e^{-i\pi(\rho_\xi - 1)} \] (4b)

where \( \rho_\xi = (\xi^2 - \alpha^2)^{1/2} \), \( \alpha' = \alpha/\gamma \), and \( \Gamma \) is the gamma function.

The energy loss \( \Delta E(\theta) \) is the difference between the initial energy, \( W_0 = \gamma M c^2 \), and the final energy, \( W_f = \gamma (W_{CM} + \beta_{CM} c P_{CM} \cos \theta) \). Here \( \beta_{CM} = v_{CM}/c \), \( v_{CM} \) is the CM velocity and \( P_{CM} \) the CM momentum of the ion, which has mass \( M \). Since

\[ E_{CM} = M \beta/(M + M \sqrt{1 - \beta^2}) \] (5)

\[ P_{CM} = \gamma M_{CM} (\beta - \beta_{CM})/\sqrt{1 - \beta_{CM}^2} \] (6)

then

\[ \Delta E = m v^2 (1 - \cos \theta)/[(M + m \sqrt{(1 - \beta^2)})^2 - M^2 \beta^2] \]

\[ = m v^2 \gamma^2 (1 - \cos \theta) \]

\[ = \hbar^2 k^2 (1 - \cos \theta)/m \]

(7)

since \( m \ll M \). Note that \( k \) is the wave number of the electron in the CM system.
Equation (2) can be written in a more conventional form

$$\frac{dE}{dx} = \frac{4\pi NZ^2 Z_j^2 e^4}{mv^2} \left( \frac{E_0^2}{8\alpha^2} \right) \int_0^\infty \sin^2(1-\cos \theta) |F(\theta)|^2 d\theta = B_0 S$$

(8)

where $B_0 = 4\pi NZ^2 Z_j^2 / mv^2$

The relativistic corrections to $F(\theta)$ come from the phase shifts defined in Eqs. (4) which are to be compared with the NR phase shifts defined by

$$e^{i\delta_{\ell,0}} = \frac{\Gamma(\ell+1 - i\alpha/\beta)}{\Gamma(\ell+1 + i\alpha/\beta)} = e^{i\delta_{\ell-1,0}}$$

(9)

They agree in the limit $\alpha \sim \alpha/\gamma \sim \alpha$ and $\rho_{\ell+1} \sim 1 + 1$.

In presenting the relativistic correction to $dE/dx$ we write

$$S = S_{NR} + S_{RC} = S_{NR} + \frac{\beta^2}{8\alpha^2} \int_0^\pi \sin^2(1-\cos \theta) \left[ |F(\theta)|^2 - |F_0(\theta)|^2 \right]$$

(10)

where $F_0(\theta)$ is the NR limit of $F(\theta)$. The non-relativistic expression for $dE/dx$ is calculated as in Ref. (22), giving

$$S_{NR} = \sum_{\ell} \frac{1}{\ell+1} \frac{1}{\ell+1} + \psi(1) - \text{Re} \psi(1+i\alpha/\beta)$$

(11)

where $\psi$ is the digamma function and $\ell_{\text{max}} = 2mv^2/\chi_{\text{ad}}$ is chosen using the adiabaticity condition for large impact parameter collisions with a harmonically bound electron. The Bethe-Bloch NR form is recovered if the proper oscillator strength is included for each effective oscillator in the
target atom: then \( \nu_0 + 1 \), the mean excitation energy of the atom.

To compare with the approach of Ref. (21), we write the main result of that paper as

\[
S = \ln\left(\frac{2m^2 \gamma^2}{I}\right) + \psi(1) - \text{Re} \psi(1+i\alpha/\beta) - \beta^2 + G/2 + C_R
\]  

(12)

where \( G/2 \) and \( C_R \) are the contributions from \( \theta > \theta_0 \) and \( \theta \leq \theta_0 \), respectively.

Equation (12) becomes, after relativistic correction to the contribution from distant collisions is included

\[
S = \ln\left(\frac{2m^2 \gamma^2}{I}\right) - \beta^2/2 + \psi(1) - \text{Re} \psi(1+i\alpha/\beta) + S_{\text{RC}}
\]  

(13)

where the factor \( 2m^2 \gamma^2/I \) comes from the combination of \( C_R \) in Eq. (12) and the logarithmic term in the contribution from distant collisions, whose relativistic correction gives rise to a term equal to \(-\beta^2/2\). The relativistic Bloch correction, \( S_{\text{RBC}} \), according to the standard definition, excluded the lowest-order correction term \(-\beta^2/2\). Thus a comparison of Eqs. (13) and (10) shows

\[
S_{\text{RBC}} = S_{\text{RC}} + \beta^2/2 - G/2 = C_R
\]  

(14)

Since we do not treat \( \theta \) separately in the two regions to obtain \( G/2 \) and \( C_R \), our \( S_{\text{RC}} \) is the sum of the relativistic Mott correction \( (G/2) \) and \( S_{\text{RBC}} \) in addition to \(-\beta^2/2\). To conform with notation, we discuss \( S_{\text{RBC}} + G/2 = S_{\text{RC}} + \beta^2/2 \).
III. Results and Discussion

$S_{RC}$ in Eq. 10 can be calculated by using the standard orthogonality relations for Legendre functions and expressions for integrals of $\cos^\theta x$ products of Legendre functions. The results of this analytic integration can be expressed as

$$S_{RC} = \frac{\ell^2}{2a^2} \sum_{l=0}^\infty \frac{\ell(\ell+1) \left[ (\ell+1)^2 - (a/\beta)^2 \right] - (\ell+1) (2\ell+1) (2\ell+3) \left[ (\ell+1)^2 - (a'/\beta)^2 \right]}{(2\ell+3)} \frac{2i(\delta_{\ell-1} - \delta_{\ell+1})}{2\ell+1} - \frac{\ell(\ell+1)}{2\ell+1} \frac{2i(\delta_{\ell-1} - \delta_{\ell+1})}{2\ell+1} \text{Re}[e^{i\delta_{\ell-1} - i\delta_{\ell+1}}] \quad (15)$$

We have calculated this series using a computer program which utilizes Stieltjes continued fractions to obtain the Gamma functions. Convergence to three figure accuracy can be achieved with approximately 100 terms in the series. The results are shown in Figures 1 and 2. These results are consistent with Ref. 20 since $[S_{RC}] + \beta^2/2$ agrees with the tabulated values of $\Phi/2$ in that reference to about 3 significant figures. However, the labor involved in the calculation is greatly reduced compared to that of Ref. 20. Also, the approximate expression for $\Phi/2$ (called $\Phi_3/2$ in Ref. 20) shows good agreement with $[S_{RC}] + \beta^2/2$ for low and intermediate $Z_1$ and large $\beta$ as expected. Ahlen's approximate expression for the Mott correction also agrees fairly well with our calculation of $[S_{RC}] + \beta^2/2$ for large $Z$ and $\beta$ in the limit of $\delta_{\ell} \to 0$.

In summary, the calculations in Ref. (22) shows how the exact scattering amplitudes can be used to obtain the non-relativistic Bloch corrections by using a method much less elaborate than the original paper (4). Our work has extended this method for high order terms at relativistic velocities and the calculation is simpler than the earlier work in computation and interpretation.
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2. N. Bohr, Phil. Mag. 25, 10 (1913); Kgl. Danske, Vid. Sels., Mat-Fys. Medd. 18, 8 (1948).


8. Extensive work on such density effect corrections has been carried out by R. Steinheimer. Summaries of this are available in Refs. (1).


15. The parameter \( \chi \) was introduced by these workers to account for the effects of single-particle excitations in their treatment, which emphasizes collective effects.
23. Note that in references 22, Eq. (1) should be smaller by a factor of 1/2 and the Bohr parameter $\kappa$ following Eq. (3) should be a factor of 2 larger (i.e., $\kappa = 2Z_1^2 e^2/\hbar v$).
25. Jackson and McCarthy\(^{(16)}\) as well as Fano\(^1\) show explicitly how $-B^2/2$ originates from the distant collisions.
Figure 1. The relativistic correction to energy loss vs the atomic number of the projectiles ($Z_1$) for various velocities $\beta$. 
Figure 2. The relativistic correction to the energy loss vs the velocity of the projectiles $\beta$ for various atomic numbers $Z_1$. 
Stopping Powers of Solids for Low-Energy Protons

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Electron gas models are useful approximations for describing the valence electron response of a solid to the passage of a charged particle. A simple free-electron gas model was used by Fermi and Teller\(^1\) to estimate the time required for a "mesotron" to be stopped in various solids. More recent work has employed the Lindhard dielectric response function,\(^2\) or approximations thereto, for calculations of the valence electron contributions to energy loss per unit pathlength for protons. Such calculations have generally shown rather poor agreement with experimental data for low-energy protons (velocity small compared to the Fermi velocity, \(v \ll v_F\)). The purpose of this paper is to draw attention to a recent calculation of the stopping power for slow protons using a density-functional formalism.\(^3\) These new results have been shown to give good agreement with experimental data and thus should provide valuable theoretical guidance in estimating stopping powers of solids for which no experimental data are available.

The density-functional approach was used to calculate the self-consistent potential of a static proton in an electron gas.\(^3\) The energy loss rate for a slowly moving proton of speed \(v\) calculated from this potential is shown as curve ENR in Fig. 1 in the form "stopping power/\(v\)" as a function of \(r_s\). All quantities are in atomic units and \(r_s\) is related to the valence electron density \(n_o\) through \(4\pi r_s^3/3 = 1/n_o\). For comparison we show
the Lindhard-Winther result\(^5\) (curve LW) given by

\[
\frac{1}{V} \frac{dW}{dR}_{\text{LW}} = \frac{2}{3\pi} \left[ \frac{1+2\chi^2/3}{\chi^2} - \frac{1-\chi^2/3}{1+2\chi^2/3} \right] (1-\chi^2/3)^2
\]

with \(\chi^2 = 0.166r_s\). This form is a very good approximation to the values predicted numerically using the exact Lindhard dielectric response function for an electron gas. The dramatic increase of the non-linear, density-functional results over those of the linear response theory is shown in Fig. 2. For many solids commonly used in experiments (1.5 \(\leq\) \(r_s\) \(\leq\) 2.5) the new results show increases of \(\sim 65\%\) over the Lindhard-Winther predictions.

![Figure 2](image)

**Figure 2**

Comparisons of the density-functional predictions with experimental data were recently made by Mann and Brandt.\(^4\) They collected data on targets covering a wide range of atomic numbers and plotted reduced stopping powers \(S_p/f_{\text{LW}}\), where \(S_p = dW/dR\) and \(f_{\text{LW}} = (v_F/v)(dW/dR)\)\(_{\text{LW}}\), as a function of \(v/v_F\). The values of \(r_s\) were taken from Isaacson's tables.\(^6\)
Their comparisons are shown in Fig. 3 with the density-functional results given by the curve ENR and the Lindhard-Winther prediction by LW. The curve FR is a theoretical prediction by Ferrell and Ritchie in which electrons at the Fermi surface are viewed as scattering from the screened potential of a proton. As a guide to the comparison of data with theory, short-dash lines are included giving a ±15% variation around the line ENR. Mann and Brandt conclude that, within the uncertainties in the data: (1) the density-functional predictions give good agreement with the data, (2) the linear dependence on velocity holds up to \( v \approx v_F \), and (3) a proton effective charge less than 1 is not required.

Additional comparisons of theory and experiment may be made using results compiled by Anderson and Ziegler in a book on stopping powers. They collected experimental data for a wide range of energies and employed simple analytic forms to obtain
a "best-fit" for several elemental solids. For energies below 10 keV, a simple, velocity-proportional expression for stopping power is given. These data-based fits are compared with the density-functional results as a stopping power ratio. The ratios are shown by circles in Fig. 4 for 16 elements specified by their atomic number $Z_2$. With the exception of Pb($Z_2 = 82$) the agreement is similar to that shown in Fig. 3 since many of the same data sets are employed in References 4 and 8.

Anderson and Ziegler used an interpolation scheme, described in Ref. 8, to predict stopping powers of materials for which no data were available. Comparisons for several solids are shown as "x's" in Fig. 4. Substantial variations about the density-functional predictions are seen.

In conclusion, the recent density-functional calculation of the stopping power of an electron gas for low-velocity protons is well-supported by experimental data. These theoretical results should be useful for estimating stopping powers of solids for which no reliable experimental data are available.

References


Range Distributions and Electronic Stopping Power of Nitrogen Ions in Solids

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Range Distributions and Electronic Stopping Power of Nitrogen Ions in Solids

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INTRODUCTION

In this presentation I wish to report on some recent data, developed at the Naval Surface Weapons Center, that pertain to range distributions and electronic stopping powers of nitrogen ions in solid elemental targets. We had previously inferred electronic stopping powers of nitrogen in the energy region to 800 keV in targets having atomic numbers \( Z_2 \) = 22 to 32 and 40 to 52, as well as in C, Si, Ta, Au, and Pb. The general feature of the \( Z_2 \) oscillations were observed, with minima occurring at the positions of the closed d shells. In particular, the measured data show minima occurring near Cu and Ag. The motivation of the present investigation was to determine if a minimum also occurs near Au, as theory predicts, as well as to study the regions about the rare earth materials, to verify the maxima predicted.

EXPERIMENTAL RESULTS

The general experimental procedure is to implant 800 keV \( ^{15}N^+ \) ions in selected target materials, in the present instance from \( Z_2 = 62 \) to 90 and in Al and Th. The range distributions are
determined by measuring the gamma-ray yield from the $^{15}$N(p,$\alpha$γ)$^{12}$C nuclear resonance reaction at 429 keV as a function of incident proton energy and by deconvoluting the yield curve, taking into account the width of the resonance and the width of the incident proton beam. The electronic stopping power is inferred by forcing a fit between the experimentally determined projected range and that calculated with the LSS transport theory. In the analysis to date, the electronic stopping power is taken to be proportional to projectile velocity. In our earlier work we studied the range distributions of $^{14}$N implants. The resonant reaction involving the $^{15}$N ions is much narrower and has a much larger cross section than that involving $^{14}$N ions, and consequently we were able to obtain more precise data with less effort than previously.

The experimentally inferred electronic stopping power is shown in Fig. 1, where the circles and the crosses represent the $^{14}$N and $^{15}$N data, respectively. The structured curve shows the results of calculations by Land and Brennan which are based on the Firsov model, in which atomic wave functions for free atoms are used to calculate charge densities and electronic velocities. Also shown for comparison are the Lindhard-Scharff values. All results are presented for a projectile velocity of $v = v_0$ (recall that we are assuming velocity-proportional stopping in the data analysis). A single parameter in the Firsov model, a minimum impact parameter, was chosen to fit the data in the $Z_2 = 22$ to 32 and 40 to 52 regions. The data clearly show the minima near Cu and Ag and also verify a minimum near Au,
although the theoretical values are larger than the experimental results in the region above $Z_2 = 62$.

In order to investigate the consistency of the present data, we consider the straggling in projected range. In Fig. 2 we show the ratio of the experimentally-determined to the (LSS)$^3$ theoretically-calculated straggling for incident 800 keV nitrogen ions. The calculated values are based upon the electronic stopping inferred from the measured projected range. In the figure the solid circles show $^{15}$N data and the open circles $^{14}$N data. We note that, with the exception of the point at $Z_2 = 62$ (Sm), the values for the $^{15}$N data lie close to a ratio of unity with very little spread. In fact, there is even a suggestion of an overall trend of the data with respect to this theory, that the ratios for lighter targets lie above unity and those for the heavier targets lie below unity. We have no reason for this. By contrast, the ratio values for the $^{14}$N data show considerable spread, with most values below unity. It had always been a puzzle to me why so many of the experimentally measured widths were smaller than what was predicted theoretically. It became apparent that the reason for this lie in the broad width of the $^{14}$N resonance reaction (4.8 keV), along with the relatively large statistical errors of these data. The background levels for the measured gamma-ray yield curves could not be well determined for the $^{14}$N data; they were much better determined for the $^{15}$N data.

As part of the present investigation, we measured the range distributions of $^{15}$N implants for a few target elements for which we had previously measured the range distributions of $^{14}$N ions.
Comparison of the results are shown in Fig. 3 in which the ratios of the inferred electronic stopping powers for $^{14}$N to $^{15}$N ions are presented when referred to a common velocity. We see that these ratios are within 5% of unity for all elements except C (8%) and Cr (7%), two elements showing widely varying straggling values for the $^{14}$N implants (see Fig. 2). Thus we conclude good agreement between the two data sets for electronic stopping power.

THEORETICAL COMPARISONS

In the time period since we had performed the calculations of electronic stopping power based on the Firsov model, there has been considerable progress in the development of the effective-charge theory of stopping, work pioneered by Werner Brandt.\(^5\) According to this theory, the electronic stopping power, $S(Z_1,Z_2,v)$, for any projectile, $Z_1$, on any target, $Z_2$, at the velocity $v$ is given by

$$S(Z_1,Z_2,v) = Z_{1,\text{eff}}^2 S(Z_1=1,Z_2,v).$$

The effective charge, $Z_{1,\text{eff}}$, can be established either theoretically from the investigations of Brandt and his collaborators or empirically, as was just described by Jim Ziegler.\(^6\) The proton stopping, $S(Z_1=1,Z_2,v)$, can similarly be estimated either theoretically, based on the calculations of Echenique, Nieminen and Ritchie,\(^7\) or from experimental data.\(^8\)

In Fig. 4 we show a comparison of the present experimental values of electronic stopping for incident nitrogen ions with a
theoretical curve, based on the empirical effective charge and proton stopping of Ziegler. The stopping power is shown at the energy 56 keV/amu, which is approximately 800 keV for either $^{15}\text{N}$ or $^{14}\text{N}$ projectiles. The agreement is quite remarkable. The minima at Cu, Ag, and Au are well reproduced and there is good consistency in the regions tending toward maxima. The region around $Z = 57$ in the theoretical curve displays a genuine peak as opposed to a spike. We plan to measure the range distributions in targets from $Z = 57$ to 61 to test the prediction of the calculated curve. (Our initial efforts to do this failed when we discovered that our target samples had completely disintegrated because of outdated shelf life.)

If the stopping power values plotted in Fig. 4 at 56 keV/amu were shown instead at 25 keV/amu, we would note some differences in the shape of the theoretical curve, although the overall agreement would still be satisfactory. These differences indicate departures from strict velocity-proportional stopping. The velocity dependence implied by the empirical curves of Ziegler could be invoked in the analysis of the present data and we plan to do this.

As a theorist, it is of interest to me to see what the electronic stopping would be like if determined strictly by theoretical considerations. Figure 5 shows a calculation of the stopping power for 25 keV/amu nitrogen ions in which the effective charge is determined from the work of Brandt, although the proton stopping is still taken from Ziegler. (The Ziegler values for the proton stopping at 25 keV are generally not much
different from those predicted by Echenique et al.\textsuperscript{7} for all
target elements of the present experimental investigation.) We
find many spikes in the theoretical curve and the question arises
in my mind what is their origin. This theoretical curve is
basically a function of just one quantity, the Fermi velocity of
the electrons of the target solid. The Fermi velocities used in
this calculation were taken from the compilation of Isaacson,\textsuperscript{9}
for those elements for which a value is available, and from the
calculated values reported by Ziegler and others. The values
compiled by Isaacson are based upon experimental measurements of
the plasma frequency of a given solid. These values often show
considerable variation from one element to the next and it is
this variation which is mainly responsible for the spikiness of
the theoretical curve shown in Fig. 5.

CONCLUDING REMARK

This point, then, leads me to conclude this presentation
with a question to this workshop. By Fermi velocity, what we
really mean in the present context is the effective number of
conduction electrons that participate in the dynamic response to
the intruding projectile. So, what is the relevant Fermi
velocity that should be used for stopping power calculations?
What measurement should be made to obtain it?
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FOR NITROGEN PROJECTILES WITH $v = v_0$

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RATIO OF EXPERIMENTAL TO (LSS) THEORETICAL RANGE STRAGGLING
\( \Delta R_p^{ \text{exp}} / \Delta R_p^{440} \) vs TARGET ATOMIC NUMBER \( Z_2 \)
FOR INCIDENT 800 keV NITROGEN IONS

RATIO OF ELECTRONIC STOPPING POWER
FOR \(^{14}\text{N} \) TO \(^{15}\text{N} \) INCIDENT IONS AT A COMMON VELOCITY
FOR SEVERAL TARGET ELEMENTS \( Z_2 \)
The Stopping and Range of Ions in Solids*

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*The contribution of J. F. Ziegler is abstracted on the following page. A full account of this work will appear as Volume I of "The Stopping and Range of Energetic Ions in Matter" by J. F. Ziegler, J. Biersack and U. Littmark.
THE STOPPING AND RANGE OF IONS IN SOLIDS

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This chapter covers the physical phenomena associated with the penetration of energetic ions into solids. It is primarily concerned with the quantitative evaluation of how the ions lose energy to the solid and the final distribution of these ions after they stop within the solid. Also considered are the first order effects on the atoms of the solid, particularly the electronic excitation of the atoms, the displacement of lattice atoms by energetic collisions (lattice damage) and the production of plasmons and phonons in the solid by the passing ions. No evaluation is made of thermal effects in the solid, especially redistribution of lattice atoms or implanted ions by thermal or vacancy induced diffusion.

This broad field is presented in an historical context so it can be seen how various aspects became important because of specific scientific interests in atomic theory, quantum mechanics, radioactive atoms, nuclear physics and ion implantation. Finally, examples of modern theory are presented with comments about how to interpret the physical phenomena which are predicted and observed.

CHAPTER SUMMARY

1 - INTRODUCTION AND HISTORICAL REVIEW

2 - NUCLEAR STOPPING CROSS-SECTIONS

3 - ELECTRONIC STOPPING CROSS-SECTIONS

4 - MONTE CARLO CALCULATIONS OF RANGE DISTRIBUTIONS
Particle Tracks

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Particle Tracks

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The term particle track is used by different authors to describe a variety of phenomena. Frequently the word track is used to infer the geometric array of excitations and ionizations generated by the passing ion and its secondary electrons in the initial physical stage of track formation. It is this which is generated by Monte Carlo calculations, making use of known cross-sections. A second use of the word track refers to an observable end-point, like the array of developed grains in a nuclear emulsion, initiated by the passing ion, produced in the neighborhood of its path, and attained after a number of complex internal and external processes that are poorly understood. In relation to this second use of the word track two points must be made. First, tracks are delta ray phenomena. Second, the properties of the detector are fully as important as the initial atomic physics in any description of track properties. One must deal with the properties of the ion, its charge, its speed, the details of its electronic clothing, One must deal with the medium through which the ion passes, as it is a target for atomic collisions, and for the subsequent delta ray interactions. Finally, one must deal with the properties of the medium as detector: which events, which combinations of events lead to the observed end-point. A cautionary note: the parameters which describe the ion and those which describe the detector are not separable variables. Track phenomena do not scale.

These statements are illustrated by Figure 1, in which we display sections of the tracks of energetic argon ions in a series of nuclear emulsions designated as Ilford K.5 to K-3 emulsions. These are all made from a parent material called K.0, and so all have essentially the same composition, the same grain size. The addition of a sensitizer, thought to be gold and sulphur, yields maximum sensitivity at about 25 ppm of gold, in K.5 emulsion. The addition of a proprietary desensitizer yields minimal sensitivity in K-3 emulsion. All of these emulsions were coated onto glass plates, were exposed to the same Ar beam, and were developed later at the same time in the same bath. At equal distances from the stopping end of the particles, at left, the particle speeds are the same, the atomic physics is the same, the energy deposition is the same, yet the tracks are vastly different.
The stopping power is a maximum at about 15 microns from the stopping end. The tracks give no evidence of the location of this maximum. At about 110 microns from the stopping end the particle energy is about 10 MeV/amu, at 2 mm the energy is about 67 MeV/amu, at 10 mm it is 175 MeV/amu.

The track appearance does not scale, especially as between the top panel, in K.5 emulsion and the bottom panel, in K-3 emulsion. We can take the cross-sectional area of a thick track to approximate the cross-section for the production of developed grains when this cross-section is greater than the cross-sectional area of a grain. We can take the fractional linear density of developed grains (the ratio of developed grains to grains intersected by the ion) to represent the cross-section for grain activation when the track looks like a string of beads. In the last 500 microns of path length in both K.5 and K.2 the activation cross-section decreases with a decrease in range (or an increase in stopping power). The cross-section remains about the same in the last 200 microns for K.0 to K-2 emulsions. The cross-section increases as the range decreases for K-2.5 and K-3 emulsions. This failure of track appearance to scale emphasizes the statement that track parameters and emulsion parameters are not separable variables. One cannot simply multiply the K.5 track by a Quality Factor, or a Relative Effectiveness, to generate the K-3 track. Typically track forming systems do not yield a maximum in response at the Bragg peak. Witness the track in K.5 emulsion.

A second important property of detectors is shown in Figure 2. Here we have exposed two different nuclear emulsions to an alpha particle source. The upper pair of panels are from Kodak NTA nuclear emulsion exposed to the center and to the periphery of an alpha particle source for 15 minutes and then normally developed in D-19 developer for 5 minutes. The lower pair of panels is from a 30 hour exposure of K-1 emulsion developed in a very weak developer which we call Stevens H for 40 minutes. Note how different the upper and lower panels appear. In the upper panels we see individual particle tracks whose initial positions are random, but whose subsequent grain formation is not entirely random for we can make a pretty good prediction of the location of subsequent grains along a track from knowledge of the position of the first two grains. In the lower panel the grain distribution shows no evidence of individual tracks. We are seeing a truly random distribution of track intersections.

Simply to underline what has been said, there is more to track formation than the initial atomic physics is able to tell us.
Reference.

Figure 1
Figure 2
Electronically Induced Sputtering and Ion Etching of Viruses

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Electronically Induced Sputtering and Ion Etching of Viruses

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This discussion is composed of two parts: (1) A summary of the work that has been done on the processes that lead to the sputtering of insulating solids, and (2) the results of using ions as a technique to etch viruses so that information can be gained about their structure by viewing them in the transmission electron microscope.

Sputtering of Insulators

Over the years many investigations have been aimed at studying the sputtering of metals and semiconductors. The bulk of the results of these experiments can be explained by considering that the incident ion's energy is deposited in two forms: (1) electronic – excitation and ionization of atoms of the solid, and (2) nuclear – the creation of nuclear motion by virtue of quasielastic collisions of the incident ion and recoils with atoms of the solid. For metals and semiconductors the sputtering appears to be produced solely by the nuclear deposition, even though this might be small compared to the electronic deposition. The theoretical description of the resulting ejection of atoms from the surface is generally called the collision cascade theory (sometimes the Sigmund theory). Fig. 1b shows that for copper the experimental results for the sputtering yield agree reasonably well with the predictions of collision cascade. However, when one considers an insulating solid such as $H_2O$, it is seen in Fig. 1a that for incident protons (for which the energy deposition is largely electronic) the experimental results and the cascade theory differ widely. In metals the free electrons are capable of rapidly dispersing the electronic energy, so that no contribution to sputtering results. However, in insulators it is quite possible that the electronic energy is somehow converted to nuclear motion, thereby contributing to the sputtering.

We see in Fig. 2 the number of ejected particles from solid $D_2O$ plotted vs. temperature. It is seen that the parent molecule (mass 20) has a yield that is independent of temperature up to 140K, whereas the number of ejected $D_2$ (mass 4) and $O_2$ (mass 32) depend very strongly on temperature. This indicates that considerable chemistry and diffusion is occurring in these cold solids. If we now investigate the energy of the ejected $D_2O$ molecules, we obtain the result shown in Fig. 3. For the low energy, heavy ions (50 keV $Ar^+$) for which the deposition is largely nuclear, the shape of the spectrum is seen to be approximately the same as predicted by collision cascade, but with an assumed surface binding energy that is about a factor of ten less than the sublimation energy for ice. We also see that the shape of the spectrum tends toward more low energy particles as we increase the amount of electronic deposition by going to 1.5 MeV $Ar^+$ and then to 1.5 MeV $He^+$.

In Fig. 4 we show results for solid $D_2O$ for incident 1.5 MeV $Ne^+$. Here the number of ejected $D_2$, $D_2O$, and $O_2$ molecules is plotted vs. fluence of Ne ions. In the case of $D_2O$ (mass 20) the yield increases suddenly as the beam is turned on, but in the case of $O_2$ (mass 32) we see that the number of ejected molecules gradually builds up, indicating that it is necessary to produce fragment molecules in the solid (radicals) before the $O_2$ can be formed and ejected. The case of $D_2$ seems to be a combination of a sudden increase followed by a slow rise of the type seen for $O_2$.

In considering the processes whereby electronic excitation leads to sputtering (which after all is nuclear motion) one looks for mechanisms for converting excitation to nuclear kinetic energy. One possibility is to consider potential curves between neighboring atoms. In Fig. 5 we show several curves for solid Ar. If one has excitation to one of the upper curves, then it is likely that through curve crossings the system relaxes non-radiatively (sometimes moving on repulsive curves) to the lowest excited state from which it emits a 9.8 eV photon to the ground state, which is also a
repulsive curve. This allows the possibility that for every excitation of an argon atom one gets 3eV of kinetic energy. If this energy is released near the surface of the solid it could well contribute substantially to the sputtering, since the binding energy of argon atoms is only 0.08eV.

**Ion-Etching of Viruses**

The exterior appearance of viruses is known from electron microscope studies and the component proteins and DNA are known from chemical analysis. However, we have very little information concerning the interior structure of viruses. The discussion here is of a simple technique for adding to our information concerning this interior structure—namely, etch the viruses with energetic ions to remove some of the outer coating, so that one can view the interior with a relatively small amount of disturbance created by the etching.

If one considers Adenovirus 2 it is known that the exterior is an icosahedron (20 equilateral triangles) but the architecture of the interior has only been guessed at. If one etches these viruses for several minutes with 1 keV Ar$^+$ ions and uses rotary shadowing (deposition of a heavy metal onto the surface from all sides) the result is shown in Fig. 6. It is quite clear that some of the interior has been exposed. In particular, one sees the appearance of three spherical objects within the shell of the virus. These pictures lead to the development of the model shown in Fig. 7, where each of the twelve spheres in the interior is located at one of the vertices of the icosahedron.

Since there are very few techniques for investigating the interior structure of viruses and similar biological systems, it appears that ion etching is a valuable addition to the electron microscopic study of these systems.
Fig. 1b
1.5 MeV He$^+$ ON D$_2$O
MASS 20

Fig. 2
Fig. 3
Fig. 4

Mass 4

Mass 20

Mass 32

\( \Phi (1.5 \text{ MeV NEON IONS/cm}^2) \)
Fig. 5
Studies of Charged-Particle Tracks in Liquid Water

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We have made detailed Monte Carlo calculations of the transport of a charged particle and all of its secondary electrons in liquid water. The computations use a dielectric response function, which includes collective effects, that we developed specifically for liquid water. The electrons are followed until their energies fall below the threshold (~ 7.40 eV) for producing electronic transitions. As an example of this work, Fig. 1 shows a stereoscopic view of the complete track of a 5 keV electron that stops in water. Each dot represents the position of an inelastic event (i.e., an ionization or excitation) caused by the primary electron or one of its secondaries.

The purpose of the work presented here is to show various appearances of track structure when considered on different spatial scales. To be specific, in Figs. 2-6, we show different "pictures" of parts of the same track produced by a 1 MeV proton in water. Details for each figure are given in the captions. Depending on the length of track segment chosen and the scale for viewing, a dense region resembling a track "core" may be evident or absent.

References


Fig. 1. Stereoscopic view of the track of a 5 keV electron that stops in liquid water. The electron started at the left end of the horizontal line, initially traveling upward in the plane of the paper. Each dot represents the location of an inelastic event (ionization or excitation) in the water caused by the primary electron or one of its secondaries. The length of the horizontal line below the track is 1000 Å.
Fig. 2. Segment of track of a 1-MeV proton traveling upward. The beginning portion of this track is shown at three "magnifications" on scales of (a) 2,000 Å, (b) 200 Å, and (c) 20 Å.

Fig. 3. The 1-μm segment of the 1-MeV proton track in Fig. 2(a) viewed end-on at three "magnifications" on scales of (a) 1,000 Å, (b) 100 Å, and (c) 10 Å.
Fig. 4. End-on view of the same 1-MeV proton track as in Fig. 3 with length of segment increased from 1 μm to 5 μm. Scales are the same as in Fig. 3: (a) 1000 Å, (b) 100 Å, and (c) 10 Å.
Fig. 5. End-on view of segments of different lengths for the same 1-MeV proton track on a scale of 100 Å. Segment lengths are (a) 5 µm, (b) 1 µm, (c) 0.1 µm, and (d) 100 Å.
Fig. 6. End-on view of segments of different lengths for the same 1-MeV proton track on a scale of 10 Å. Segment lengths are (a) 5 μm, (b) 1 μm, (c) 0.1 μm, and (d) 100 Å.
Cluster Ion Impacts on Solids

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Cluster Ion Impacts on Solids*

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Experimental methods for the production of cluster ions by expansion of weakly ionized plasmas through a supersonic nozzle and skimmer were described. Techniques for the production of relatively narrow mass distributions of singly charged ions containing as many as thousands of molecules of Hydrogen, Argon, Water, Alcohols and Hydrocarbons were reviewed with an explanation of the dependence of the mean cluster ion size on stagnation conditions in the ion source and the orifice geometry in nozzle or free jet expansions. Diagnostic techniques for the mass analysis and detection of these high molecule weight cluster ions were reviewed. A description of the BNL 400 kilovolt post-acceleration detection system and the advantages of secondary electron pulse distributions were presented and discussed.

The application of energetic cluster ion impacts for deposition of large amounts of translational energy in thin films and solid surfaces was the main topic of the presentation. Cluster ions can be used to generate assemblies of atoms in solid surfaces with energies determined by available acceleration facilities. The production of assemblies of thousands of atoms with energies of in excess of several hundred volts per atom is readily achieved. The consequence of the ability to generate high energy densities is among other things the production of craters, cavities and in thin films holes of sizes that are smaller than those achievable by atomic ion bombardment and wet etching techniques. Examples of such results were presented showing holes in thin carbon films obtained by transmissions electron microscopy.

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Wakes in Large Hydrocarbon Molecules

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Electron capture-and-loss-to-the-continuum processes (ECC and ELC, respectively) for a fast projectile traversing atomic, molecular or solid targets produces a forward directed beam of electrons roughly velocity matched with the projectile, i.e. $v_e \approx v_p$. These continuum electrons, detected at $\sim 0^\circ$, form a cusp, such as is shown in Figure 1, with a peak at $v_e = v_p$. Cusp shapes for atomic gas and solid targets differ in their skewness and widths. There is also a difference in skewness, width and cross sections between ELC and ECC cusps for atomic (and molecular) gas targets, as can be seen in Figure 1 for 0.6 MeV/u He$^+$ and He$^{++}$ on CH$_4$. These cusp shape differences are a clue to the physical origin of the continuum electrons [1]. In an attempt to understand the difference in cusp shapes between atomic and solid targets, we have "bridged the gap" by systematically varying target "size", using hydrocarbon ($C_{m n}^m$) molecular gas targets ranging from $m = 1$ to $m = 7$.

Our cusp shapes for 0.6 MeV/u He$^{++}$ on $m = 1, 2, 3, 4, 5,$ and $7$ molecules are shown in Figure 2. The $m = 1, 2$ and $3$ cusp shapes are quite similar in shape and show the characteristic low energy tailing associated with ECC processes; the cusp peaks acquired with equal velocity H$^+$ projectiles resemble these quite closely. For $m = 4, 5$ and $7$, however, there is a small but distinct change in cusp shape in the region of the peak as observed in the increased pointedness of the cusp. To emphasize the change in cusp shape for $m \geq 4$ targets, we have subtracted a scaled CH$_4$ cusp from the $m \geq 2$ cusp shapes. Our scale factor is determined by the requirement that the slowly-varying "wings" of the cusp should cancel out in
the subtraction. These difference spectra, which we call $\Delta C_m$ spectra 
($C_{mn} - CH_4$), are presented in Figure 3a (for $H^+$) and 3b (for $He^{++}$) for $m = 2, 4(5)$ and 7. The sudden appearance of a significant peak in the $\Delta$ spectra for $m \geq 4$ is clearly seen. Between the $H$ and $He$ $\Delta$ spectra, the width of the peak in the residue spectrum, which we will hereafter call the "W" peak, increases by a factor of $1.8 \pm 0.4$.

When a 0.6 MeV/u $He^+$ beam bombarded these same hydrocarbon gases much greater yields were measured. In addition, the cusp peak was now skewed toward the high energy side. This difference in cusp shape and yield, as seen in Figure 1, is characteristic of ELC processes. Our $He^+ \Delta C_m$ spectra were complementary to those for the bare $H$ and $He$ projectiles in that the "W" peak was largest for $m = 2$ and completely disappeared for $m = 4$. The "W" peak yields, normalized to $m$, are shown in Figure 4 for 0.6 MeV/u $H^+$, $He^{++}$ and $He^+$. Similar results are obtained at higher projectile energies, although the $\sim E^{-3}$ dependence of the overall cusp cross sections gives somewhat poorer difference spectra.

Finally, the "W" peak contribution should show up in the overall cusp yields. To show this, we present the normalized cusp yields, $R$, calculated from the relationship

$$R = \frac{Y(C_{mn})}{m \cdot Y(CH_4)}$$

The quantity $R$ is a measure of the validity of atomic cross section additivity in ECC [2] and also in electron capture to bound states (ECB) [3] in hydrocarbons, due to the very small relative contribution of the $H$ atoms to the overall total electron capture cross section from the various $C_{mn}$ target gases. It is also quite useful in the ELC results since
n = 2m. If additivity truly holds and $\sigma_{\text{ECC}}(C) \gg \sigma_{\text{ECC}}(H)$, then $R \approx 1$. Experimentally, we have always observed that: i) $R < 1$; ii) $R$ decreases as $m$ increases ($E_p$ held constant), and iii) $R$ decreases as $E_p$ decreases ($m$ held constant). In Figure 5, we show the behavior of $R$ vs. $m$ for $E_p/A = 0.6 - 2.5 \text{ MeV/u H}^+$ and $0.2 - 0.8 \text{ MeV/u He}^+$. The equal velocity He results all agreed within error with the H R values. Since these bare projectiles should all be undergoing ECC processes only, the mechanism that causes additivity failure appears to be the same for both. The experimental results for He$^+$ were all within experimental error of one another, so we have averaged the R values at each m. It is in the He$^+$ data that the clearest evidence for something unusual happening occurs. There is a distinct slope change in the R vs. m curve at $m = 3 - 4$, which can also be seen, albeit less definitely, in the ECC data.

Our additivity failure can be explained quantitatively [2,3] by the presence of intramolecular scattering collisions, succeeding the original charge transfer collision. The magnitude of this "outscattering" increases both with the size of the molecule and the relevant "outscattering" cross sections. There is no a priori reason to expect any large change in the "outscattering" cross sections per atom as the molecule size is increased nor is any observed in the case of electron capture, $\sigma_{10}$, or loss, $\sigma_{01}$, cross sections with H projectiles [4]. The break in the R vs. m curve between $m = 3$ and 4 seems then to signal the advent of some other, new process occurring in the molecule, as does the change in the cusp shape observed for molecules with $m \geq 4$.

What other processes could lead to forward-directed electrons? One possibility is "Rydberg pumping" (RYD) which occurs in solid targets and leads to highly-excited occupied levels of the projectile which are then field-ionized by the detector [5]. Another that has not been mentioned
here, but which is expected to be of importance in solid targets is the
production of a oscillatory potential wake accompanying the projectile
through the target [6]. If this wake can capture electrons they will
accompany the projectile to the edge of the target where the wake collapses.
It seems possible that some of these electrons will then exit the target,
velocity-matched with the projectile, and be detected in a forward direction.
Experimental evidence for wake-riding electrons in cusp spectra from solids
is somewhat contradictory to date [7,8,9]; nor would a wake be an obvious
possibility for large hydrocarbon molecules, even though there is
alkanes \( (C_{m}H_{2m+2}) \) with \( m \geq 3 \). However, the "W" peak width is
dependent on the \( Z \) of the projectile, \( Z_p \), as is the depth of the wake,
whereas neither ECC or ELC have a \( Z_p \)-dependent cusp width [1].

Considering each of the possible mechanisms which lead to
forward-directed electrons, and examining each for consistency with our
experimental data, a brief summary of our understanding of the origin of the
"W" peak appears in the following table.

<table>
<thead>
<tr>
<th>Experimental Results (this work)</th>
<th>POSSIBLE MECHANISMS</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>ECC</td>
</tr>
<tr>
<td>1) shape - symmetric/narrow</td>
<td>no</td>
</tr>
<tr>
<td>2) Bare H, He ( \text{FWHM(He)/FWHM(H)} \approx 1.8 )</td>
<td>no</td>
</tr>
<tr>
<td>3) threshold at ( m \geq 4 )</td>
<td>no</td>
</tr>
<tr>
<td>4) ( \text{He}^+ ) - no peak for ( m &gt; 3 )</td>
<td>no</td>
</tr>
<tr>
<td>5) Slope change in ( R ) vs. ( m ) curves at ( m = 3 - 4 )</td>
<td>no</td>
</tr>
</tbody>
</table>
From Table 1 we can see that wake production is the only mechanism that has no "no's" against it. If Table 1 is exhaustive, there is no presently known mechanism that produces electrons into a forward cone that satisfactorily explains the totality of our data except that of wake production in large hydrocarbon molecules.
REFERENCES


Figure Captions

1) Electron cusp spectra for 0.6 MeV/u He$^+$ and He$^{++}$ on CH$_4$. Cusps have been normalized at the peak channel (He$^{++}$ data multiplied by 16.1). The He$^+$ cusp is primarily ELC, while the He$^{++}$ cusp is due to ECC.

2) Cusp spectra for He$^{++}$ on hydrocarbon gases, m = 1 - 7. On the low energy side of the cusp are the C K auger electrons. The m = 1, 2, 3 cusps have a slightly rounded top compared to the m = 4, 5, 7 cusps.

3) The difference (ΔC$_m$) spectra for:
   a) 0.6 MeV/u H$^+$ - m = 2, 4, 7.
   b) 0.6 MeV/u He$^{++}$ - m = 2, 5, 7.
   (errors shown in peak region are ± 2σ, arrow at side denotes digital offset to eliminate negative numbers)

4) Yields for the He$^+$, H$^+$ and He$^{++}$ "W" peak, normalized to m vs. m. Errors shown are ± 2σ.

5) R vs. m for H and He on m = 1 - 7 hydrocarbons. The equal velocity bare H and He have R values within error. The He$^+$ data at all energies were within experimental error of one another and have been averaged together.
1) Electron cusp spectra for 0.6 MeV/u He\textsuperscript{+} and He\textsuperscript{++} on CH\textsubscript{4}. Cusps have been normalized at the peak channel (He\textsuperscript{++} data multiplied by 16.1). The He\textsuperscript{+} cusp is primarily ELC, while the He\textsuperscript{++} cusp is due to ECC.
2) Cusp spectra for He$^{++}$ on hydrocarbon gases, $m = 1 - 7$. On the low energy side of the cusp are the C K auger electrons. The $m = 1, 2, 3$ cusps have a slightly rounded top compared to the $m = 4, 5, 7$ cusps.
3) The difference ($\Delta C_m$) spectra for:

a) 0.6 MeV/u $H^+ - m = 2, 4, 7$.

b) 0.6 MeV/u $He^{++} - m = 2, 5, 7$.

(errors shown in peak region are $\pm 2\sigma$, arrow at side denotes digital offset to eliminate negative numbers)
Yields for the He⁺, H⁺ and He⁺⁺ "W" peak, normalized to m vs. m. Errors shown are ±2σ.
5) R vs. m for H and He on m = 1 - 7 hydrocarbons. The equal velocity bare H and He have R values within error. The He data at all energies were within experimental error of one another and have been averaged together.
Charge States of Ions Traversing Thin Carbon Foils Inferred from 7 MeV \( C^+_2 \) Coulomb Explosion Experiments

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The charge state of fast C\(^+\) ions is assumed to reach some equilibrium value within a few atomic layers after entering a thin carbon stripping foil and then maintain that average charge state while passing through the rest of the foil. The final distribution of different charge states is then assumed to occur at or near the exit surface of the foil by various pickup and stripping processes. Experimentally, the final charge state distribution is easily measured; however, the average charge state inside the foil is difficult to measure in any direct way. This paper describes the experimental method and results of measuring this internal average charge state for 3.5 MeV carbon ions by utilizing the Coulomb explosion of 7 MeV C\(_2^+\) molecular ions. The interesting result is that the internal average charge state is 4.2±x, and the final charge state distribution is not established at the exit surface of the foil, but beyond, approximately 100 \(\mu\)m, a distance comparable to the foil thickness in this measurement.

**EXPERIMENTAL METHOD**

The method of this experiment is to simultaneously measure the angular separation and charge state of the two 3.5 MeV carbon ions resulting from the Coulomb explosion of 7 MeV C\(_2^+\) molecular ions. The Coulomb explosion begins a few monolayers into the carbon foil where the two fast-moving carbon ions start separating because of the Coulomb repulsion of their identical average charges inside the foil.
This average charge Coulomb force persists while the ions are inside, providing the initial phase of the Coulomb explosion. Upon exiting the foil, the two carbon ions assume their final charge states which then provide the repulsion force which completes the Coulomb explosion. The two ions are then simultaneously detected after magnetic analysis which is arranged to separate all of the different charge states from \( C^+ \) to \( C_5^+ \) without perturbing the relative spatial relationship between the two carbon ions produced by the Coulomb explosion (non-focussing). The molecular \( C_2^+ \) ions are produced inside a sputter ion source and then extracted and accelerated to the high voltage terminal of an MP tandem accelerator. They then pass through a gas stripper canal where they are charge exchanged to \( C_2^+ \) ions by gas collisions and then accelerated out of the machine to ground potential. The \( C_2^+ \) fraction of accelerated ions are separated magnetically by a 90\(^\circ\) analyzing magnet and transported to the Coulomb explosion apparatus diagrammed in Fig. 1. The beam is collimated to a diameter of 50 microns and an angular spread of approximately 0.01\(^\circ\), magnetically (cleaned) after collimation and then directed into the 2 \( \mu \text{g/cm}^2 \) carbon stripping foil as shown. A non-focussing bending magnet in close proximity to the foil then separates the carbon ions according to the charge state so that they impinge on the phosphor detector surface of an image intensifier with sufficient separation for unique identification.
The phosphor detector image intensifier system has been described elsewhere. Briefly, fast heavy ions impinging on the monolayer phosphor crystalline surface scintillate phosphor grains. The scintillation flash is then amplified 185,000 times and displayed on the output screen. A sensitive TV camera then views this image and a small computer digitizes the picture into a 128 x 128 set of pixels with 4-bit level sensitivity. After digitization and storage, the computer then scans the stored information for all pixels above a preset level of intensity and stores such events in memory for eventual filing on a floppy disk. These data are later transferred to a magnetic tape and analyzed in a large computer.

As the 7 MeV $C_2^+$ ions enter the carbon foil, the immediately formed charged carbon ions repel each other symmetrically about their center of mass. However, the orientation is random with respect to the geometry of the detector system. In a few cases the two ions could have the same charge and also be oriented with the $C_2^+$ axis, parallel to the beam direction. The two ions would then follow each other and impinge on the same phosphor grain detector and produce only one scintillation flash. If the charges were different the two ions would each impinge on the detector at the centroid locations of the two charge distributions. Figure 2 is an illustration of the TV display that is digitized and indicates how different charge state combinations would appear on the screen. Figure 3 is an isometric plot of a large set of Coulomb explosion events which shows a portion of the overall charge distribution as well as the actual Coulomb explosion distribution within each charge state.
Every event that is analyzed must be a binary event or else it is rejected. In addition, both carbon ion scintillations must be symmetric about the centroid within the errors allowed by multiple Coulomb scattering. For different charge states the symmetry would be checked in terms of the centroids of the different charge state distributions. A simulated set of conditions illustrating the different possible combinations is shown in Fig. 2.

DATA ANALYSIS AND DISCUSSION

The analysis then leads to a distribution plot of the linear distance between each pair of events, symmetric about the centroid in terms of the spreading angle $\alpha_f$ of the two carbon ions, as shown in Fig. 4. This distribution can be fitted quite well by assuming a random orientation of the $C_2^+$ axis in respect to the beam direction for a given charge state combination. The rapid decrease on the high $\alpha_f$ side of the distribution indicates that the effects of both molecular vibrations as well as multiple scattering are small (an idealized rigid molecule and no scattering would show a sharp cut-off.) The $\alpha_f$ angle corresponding to the half height of the rapid decreasing portion of the distribution best characterizes the position of the idealized sharp edge.

This average opening angle, $\alpha_f$, is obviously larger for higher charge state ions and can be measured by appropriate sorting of the data for both symmetric and asymmetric charge state combinations. A large collection of these data were measured for all possible
combinations and the results are shown in Fig. 5. Here, the average opening angle, \(\alpha_f^2\), is plotted as a function of the product of the two charge states involved, \(Q_a \times Q_b\). It is interesting to note that the functional dependence is a straight line, within errors, and that the \(\alpha_f^2\) intercept for 0 charge (not measurable) corresponds to \(\alpha_f = 6.2 \pm 0.2\) mrad.

In order to analyze these data for the average charge inside the foil, Fig. 6 illustrates in a schematic diagram what may be happening during a typical Coulomb explosion for a \(C_2\) molecule with its axis oriented perpendicular to the beam direction and in the plane of the paper. Shortly after entering the foil, two ions are each at \(Q\) charge states, and begin to separate by Coulomb repulsion. After they exit their opening angle is \(\alpha_i\) and their separation is \(R_i\) at a point, where the final measured charges, \(Q_a\) and \(Q_b\), are achieved. The measured angle, \(\alpha_f\), is the result of the two stages of Coulomb explosion. Considering the energy balance outside of the foil after penetration, the energy of the two carbon ions that exit the foil is:

\[
M \left(\frac{\alpha_i V_b}{2}\right)^2
\]

where \(M\) is the carbon mass, \(\alpha_i\) the angle between the two carbon trajectories at the foil exit, and \(V_b\) is the beam velocity. The potential energy of the two carbons at the exit of the foil is:

\[
\frac{1}{4\pi \varepsilon_0} \frac{e^2 Q_a \times Q_b}{R_i}
\]
where $R_i$ is the interatomic distance of the two carbons at the foil exit, and $e$ is the electron charge.

The sum of these two terms must be equal to the final energy of the two carbons at infinity, which is:

$$M\left(\frac{\alpha_f v_b}{2}\right)^2 = M\left(\frac{\alpha_i v_b}{2}\right)^2 + \frac{1}{4\pi\varepsilon_0} \frac{e^2 Q_a x Q_b}{R_i}$$

or

$$\alpha_f^2 = \alpha_i^2 + \frac{e^2 Q_a x Q_b}{\pi\varepsilon_0 m v_R^2 R_i}$$

(1)

From Fig. 6, the $Q_a x Q_b = 0$ intersect is $\alpha_f = 6.2\pm0.2$ mrad. This $\alpha_f$ corresponds to the final angle the two carbon ions would have from their average charge inside the foil, $\overline{Q}$ and 0 final charge or neutral atoms. In other words, at $Q_a x Q_b = 0$, $\alpha_f = \alpha_i$, the angle at the foil exit. Similarly, from Fig. 6 the slope for $\alpha_f^2$ per unit charge product, $Q_a x Q_b$, is:

$$\frac{\alpha_f}{Q_a x Q_b} = 4.2\pm0.1 \times 10^{-6} = \frac{e^2}{\pi\varepsilon_0 m v_R^2 R_i}$$

or

$$R_i = 1.96\pm0.1 \AA .$$

The energy balance inside the foil, assuming a mean squared charge state $\overline{Q}$:

$$\frac{e^2 Q}{4\pi\varepsilon_0 R_0} = \frac{e^2 Q}{4\pi\varepsilon_0 R_i} + M\left(\frac{\alpha_i v_b}{2}\right)^2$$

(2)

where $R_0 = 1.31 \AA$ is the interatomic separation for the $C_2^+$ molecular ion in the ground state.
This results in $Q = 4.3$ and if $R_0$ were larger (excited state) like 1.5 $\AA$, then $Q$ would be 5.5 or close to a fully stripped $C_6^+$ ion. With these data, the effective thickness, $T$, or where the final charge occurred, may be calculated. Inside the foil,

$$M \left( \frac{R}{2} \right)^2 + \frac{Q}{Q_0} = \frac{Q_2}{Q_0},$$

or

$$R = \sqrt{\frac{4Q}{M} \left( \frac{1}{R_0} - \frac{1}{R} \right)} = R_\infty \sqrt{1 - \frac{R}{R_0}} \quad (3)$$

where $R = 2Q/\sqrt{MR_0}$ is a slope if $Q_a = Q_b = Q$ (i.e. the charges stay the same outside as inside).

Multiplying both sides of 3 by $dt/dx$,

$$\frac{dR}{dx} = \alpha_\infty \sqrt{1 - \frac{R}{R_0}} \quad (4)$$

where $\alpha_\infty = (dR/dx)_\infty$ is a Coulomb explosion final angle, $\alpha_f$ at $\infty$, if there were no change in charge. From 4, the effective thickness is:

$$T = \int dx = \frac{1}{\alpha_\infty} \int_{R_0}^{R_1} \frac{dR}{\sqrt{1 - \frac{R}{R_0}}}$$

$$= 1.82 \times 10^{-6} \text{ cm} \quad (5)$$

which corresponds to an areal density of 4.1 $\mu$gm/cm² for a density of 2.25.

Since the foil used for these measurements was only 2 $\mu$gm/cm² the final charge is apparently not reached until the ion pair is a full foil thickness, or approximately 100 $\AA$ beyond the exit foil. The number of convoy electrons originating from the surface are known to be independent of the charge state of the exiting ion which was
puzzling and without explanation. However, these results provide a simple explanation. The mechanism for achieving the final charge state (recombination with the convoy electrons, etc.) must be some statistical process that extends well beyond the average distance of 100 Å. However, it is not clear how this recombination occurs at such distances.

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Fig. 1 Diagram of Coulomb explosion apparatus for magnet separation of all carbon ion charge states.
Fig. 2 Illustration of the television display showing how different combinations of Coulomb exploded carbon ion charge states appear.
Fig. 3 Isometric display of computer processed Coulomb explosion data showing a portion of the carbon ion charge state distribution and the Coulomb explosion distribution associated with each charge state.
Fig. 4 A plot of the $^{3+}$,$^{3+}$ ion separation distribution. The analysis cutoff is due to the fact that two overlapping events are recognized as one and rejected.
Fig. 5 A plot of $\alpha_i^2$ versus $Q_a \times Q_b$ for different ion charge state combinations.

$\alpha_i = 6.2 \pm 0.2 \text{ mrad}$

$R_i = 1.96 \pm 0.10 \text{ Å}$
Fig. 6 A diagram showing schematically the two stages of Coulomb explosion: inside and outside of the stripping foil.
Beam-Target Interaction of Intense Ion Beams: 
Theory and Experiment

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Abstract

Work has continued toward the goal of developing and experimentally verifying a numerical model that can accurately predict the stopping power of any ion in any target material at any density, temperature, and degree of ionization. We will review our most recent theoretical work to compute the stopping power of low-Z ions in partially ionized material. We are presently attempting to develop an accurate description of the atomic-electron contribution to the stopping power, especially around the maximum of the curve. Our atomic-electron stopping-power models (e.g., the Local Oscillator Model, the Generalized Oscillator Strength Model, etc.) use realistic descriptions of atomic and solid-state charge densities and energy levels. Such modeling was successfully used in coupled deposition-hydrodynamic simulations of the NRL enhanced stopping-power experiments that were performed last year. These experiments provided the first verification of ion-stopping power enhancement in partially ionized targets and this was also the first definitive test of our stopping-power models. Further stopping-power experiments are in progress on Sandia's PROTO-I accelerator and are producing the first time-dependent measurements of the energy loss of intense ion beams via Rutherford scattering into a Thomson Parabola analyzer. We will also discuss the role of enhanced ion-stopping power in the interpretation of beam intensity diagnostics. Finally, the effects of enhanced stopping power on the design of PROTO-I and PBFA-I target experiments will be summarized.

Introduction

Three main applications of targets in ion-driven ICF are:

1) diagnostic support
2) demonstration of achievement of program milestones, and
3) the ultimate attainment of ignition, burn, and breakeven.

Accurate target simulation models are essential to fulfill these tasks in an efficient manner. Many aspects of target modeling are generic to ICF as a whole. The need to model the target interaction of an intense ion beam, however, is unique to the ion beam fusion program. The primary interaction of interest is the energy deposition of the ion beam within the target. Theoretical models for ion-stopping power are being developed. Moreover, experiments to corroborate these models are being explored.

In 1978, Nardi, Peleg, and Zinamon [1] published the first paper showing enhanced ion stopping in ionized targets. (Earlier work on this subject consists of unpublished reports [2,3].) The degree of enhancement depends on the ionization state of the target and on the plasma electron temperature. Enhancement occurs when free electrons are more efficient at absorbing energy in the distant collisions typical of Coulomb interactions; i.e., when the electron thermal velocity is less than the ion velocity and when the electrons are not strongly degenerate [4]. Concurrent with ionization, the average ionization potential, \( I(Z,q) \), of the remaining atomic electrons increases. (Here \( Z \) is the target nuclear charge and \( q \) the degree of ionization.) Thus, the stopping power per atomic electron decreases with ionization.

For light ion fusion, ion energies of a few MeV/amu require shell corrections along with \( I(Z,q) \) [5]. This implies the need for accurate atomic physics modeling and good interaction models near the stopping-power peak. Moreover, present low-power target behavior can be dominated by the atomic electrons. Therefore, we are currently concentrating on the atomic physics and interaction modeling necessary to describe ion-stopping power from the maximum of the curve out to the high-energy limit.

Next we motivate our work by describing the
effect of ion-stopping modeling on idealized target behavior. Then we review our recent theoretical work. Finally, we describe efforts to measure the stopping powers of an ionized target.

**Motivation**

Optimal target design will be affected by variations in the calculated energy absorption efficiency. Different stopping-power models can predict different absorption efficiencies for the same interaction configuration. In this section we will introduce four such models and demonstrate their predicted absorption efficiencies for a simple target.

The first model is standard neutral target stopping power. Ziegler [6] is one source of such tabulated information. These values serve as a reference point in measuring the degree of enhancement of an ionized system.

The second model, described in Ref. 7, is representative of attempts to describe $I(Z,q)$ by a simple scaling formula. This model, denoted "hydrogenic scaling" on the accompanying plots, proposes the formula

$$I(Z,q) = Z^2 I(Z,0) .$$

The third model is the Generalized Oscillator Strength (GOS) model of McGuire [8]. The GOS model is based on the plane-wave Born approximation. It is one of the most mature models for high-energy stopping powers, but is somewhat cumbersome to use for general ions.

The final model is the Local Oscillator Model (LOM) of Lindhard and Winther [9]. Within the standard Lindhard formalism, $I(Z,q)$ is given by:

$$I(Z,q) = I(Z,0) \left[1 - \frac{(Z-q) \ln I(Z,q)}{\gamma} \right]$$

where $\rho(r)$ is the radial charge distribution of the atom, $\gamma$ is a parameter, and the other symbols have their standard definitions.

Figure 1 shows the predicted variation in absorbed energy efficiency for these models. The results are for 1-D hydrodynamic simulations with a 2-D ion ray tracing package and for PROTO-I input parameters (described in the experimental section). The target was a simple, thin, cylindrical gold foil.

![Figure 1. Variation of time history of absorption efficiency for different deposition models.](image)

We see agreement between the models early in the machine pulse before significant heating occurs. Once appreciable ionization occurs, they diverge. Note the substantial enhancement in absorbed energy for the models with ionization effects. The difference in enhancement between even these two models can be significant to the evaluation of actual target performance. Table 1 summarizes these results as well as presenting similar results for aluminum and CH$_2$. Note that the variation with model is most pronounced for higher-Z materials for these source parameters.

Table 1.

<table>
<thead>
<tr>
<th>Substance</th>
<th>CH$_2$</th>
<th>Al</th>
<th>Au</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neutral Atom</td>
<td>.85</td>
<td>.86</td>
<td>.68</td>
</tr>
<tr>
<td>LOM/GOS</td>
<td>--</td>
<td>.90</td>
<td>.86</td>
</tr>
<tr>
<td>Hydrogenic Scaling</td>
<td>.98</td>
<td>.94</td>
<td>.93</td>
</tr>
</tbody>
</table>

**Theoretical Modeling**

Consistent with the scope of our long-term stopping-power modeling goals, we need to develop a theoretical model that is

1) convenient to use
2) accurate
3) general.

Most of our ongoing theoretical model development is associated with Lindhard's Local Oscillator Model [9] because it shows promise in all three areas. Therefore, we will devote most of the following discussion to this topic. However, some additional work on alternative methods of calculating
accurate a priori values of I(Z,q) is also being pursued [10]. This technique, perforce, requires more information than just the electron density distribution of the atom. However, these values are expected to be reasonably accurate within the validity of the atomic-physics data obtained, typically, from a Herman and Skillman structure code.

The only detailed target structure information that the LOM model requires to calculate stopping powers and average ionization potentials is the electron density distribution of the atom, \( \rho(r) \). However, there are certain shortcomings, ambiguities and general questions that need to be answered in order to make this method quantitative rather than qualitative as is now the case. That is, this method has been used to predict trends and general scaling relations rather than to calculate accurate stopping powers and average ionization potentials.

A list of our most important observations and questions relating to LOM model follows:

1) The LOM does not reproduce experimental values of I(Z,0) without ad hoc adjustments of the parameter \( \gamma \) (see Eq. 2), e.g., with \( \gamma = 1 \) the LOM gives \( I(2,0) = 119 \) rather than the experimental value of 162 eV for aluminum.

2) A consistent treatment of the low and high velocity limits of the Lindhard interaction function with respect to the parameter \( \gamma \) has not been put forward.

3) The LOM does not reproduce the proper one electron limit, I(Z,-1), for ions. That is, Peek has shown [11] that unless modified, the LOM scales as \( Z^{3/2} \) rather than with the proper \( Z^2 \) dependence.

4) The target information usually comes from an atomic structure code for free atoms. What effect does the accuracy of \( \rho(r) \) have on the calculation of \( dE/dx \) and I(Z,q)? In particular, how important are solid state effects? e.g., \( I(5,0) = 106.8 \) eV for free aluminum atoms as compared to 119 eV for a solid state distribution.

5) It is clear that some of these problems are unrelated to the structure calculation. We are therefore studying the standard dielectric response function used in the Lindhard model. Specifically, we are questioning the accuracy of the RPA (Random Phase Approximation) treatment of the interaction. In this regard we are presently investigating the relevance of the Local Field correction as first suggested by Sayasov [12].

We are closely studying the LOM both from a theoretical as well as a practical, computational view. Preliminary results of work in progress aimed at resolving some of the issues mentioned in the previous list are summarized in Fig. 2. Shown are the results of various calculations of stopping power versus proton energy as compared to Ziegler's best fit to experimental data [6] for neutral aluminum. The diamonds are point calculations by McGuire using the GOS method [8].

![Figure 2. Proton stopping power of neutral aluminum for various models.](image-url)
suggest applying this correction in calculating the low-velocity stopping power of ions. We have extended his work by using a structure code to calculate the detailed stopping-power curve shown in Fig. 2. Moreover, we are investigating the application of the Local Field Correction at all ion velocities. In particular, we are interested in the high-energy limit where modifications to \( I(\varepsilon, q) \), as given by Eq. 2, might occur. We will continue to investigate the usefulness of this correction in conjunction with our general study of the Local Oscillator Model.

Finally, we are also investigating the use of the Gryzinski model [16] for the low-energy stopping powers coupled with a Bethe-Born model for the high-energy stopping power. In addition to comparison with experimental, we can theoretically check the compatibility of such hybrid models through the use of a low-to-high energy relationship deduced by Peak [17].

### Ion Stopping Power Experiments at ICF Intensities

The first measurements of enhanced ion stopping power in partially ionized ICF targets were made at the Naval Research Laboratory [18]. These measurements provided the first verification of our stopping-power models. In the experiment a 1 MeV, 200 kA\( \cdot \)cm\(^{-2} \) deuteron beam was focussed onto a composite, sub-range diagnostic target (shown in Fig. 3). A planar anode produced an approximately normally incident beam with an intensity of 50 kA\( \cdot \)cm\(^{-2} \). A curved anode focussed the beam to about 200 kA\( \cdot \)cm\(^{-2} \) at an average angle of incidence of 20°. Neutrons from \( \text{d}(d, n)\text{He}^3 \) reactions between the deuteron beam and the \( \text{CD}_2 \) layers of the target were time-of-flight (TOF) analyzed to determine both the incident deuteron energy as well as the deuteron energy loss in the stopping foil.

![Target schematic from NRL stopping power experiment.](image)

Figure 3. Target schematic from NRL stopping power experiment.

We performed coupled deposition/hydrodynamic simulations of this experiment to test our stopping-power models. Key elements of this experiment included: 1) use of a 2-D ion ray tracing to account for the angles of incidence, 2) use of GAMMA-II voltage and current waveforms scaled to the peak measured deuteron energy, 3) use of a 50/50 mixture of protons and deuterons as experimentally measured. Measurements were made and simulation results were tallied for ion energy loss at the time of peak power on target.

Figure 4 shows the measured and calculated results for these experiments. Both Mylar and aluminum stopping foils were studied at both 50 and 200 kA\( \cdot \)cm\(^{-2} \). We note that the low-intensity Mylar experiment (a) showed cold stopping. However, a definite enhancement was seen for the high current density case (c). A small enhancement was seen for the low intensity beam on aluminum (b), while once again a strong enhancement was seen at 200 kA\( \cdot \)cm\(^{-2} \) (d). We note that good agreement was found between simulation and experiment for all four cases.

![Comparison of calculated energy loss enhancement versus experiment.](image)

Figure 4. Comparison of calculated energy loss enhancement versus experiment.

In the simulations, a hydrogenic scaling for \( I(\varepsilon, q) \) was found to be adequate for the Mylar targets. However, this scaling overpredicted the...
stopping-power enhancement in the aluminum targets. For this case we obtained good agreement using the GOS/LDN values discussed in Ref. 5. Subsequent comparisons of GOS stopping powers with those obtained using the hydrogenic scaling confirm the applicability of the latter scaling for low-Z materials.

A new experiment aimed at expanding this initial data base is presently being fielded on the PROTO-I accelerator at Sandia. This experiment is studying ion-stopping power up to about 1 TW/cm$^2$. Moreover, a modified time-resolved, electric-magnetic analyzer [19] used in conjunction with Rutherford-scattering foils, provides a time-history of the energy-loss process. A complete description of the experimental aspects of this measurement can be found in a companion paper in these proceedings [20].

This experiment has already demonstrated enhanced ion stopping in partially ionized plasmas. We seek to obtain agreement between coupled deposition/hydrodynamic simulations and experimental data. To perform such a simulation one must obtain a description of the time-dependent source terms. This information includes: 1) appropriate voltage and current density time histories for the ion beam at the target, 2) beam composition, and 3) incident beam source geometry. However, some variables associated with the experiment that are required for simulation are still being studied. Our goal is to define these three quantities in a general and consistent way such that quantities that may remain relatively constant from shot-to-shot (e.g., beam geometry) will remain fixed once defined while quantities such as the proton voltage history will allow for shot-to-shot variations.

We are using shot #4123, a cylindrical, aluminum stopping target—see Fig. 4 in Ref. 20), as our baseline case. The compatibility of the symmetry of such cylindrical targets with the barrel diode configuration, as well as their relative simplicity, make these targets the most promising. Our initial studies have used a 1-D hydrocode with a 2-D ion ray tracing package. The source geometry is defined by the anode and target radii and the cylindrical symmetry of the experiment. A uniform beam profile within the target radius is assumed. The current density on target has initially been taken without shot-to-shot modification from a recent study by Leeper and Lee [21]. This study showed a peak current density of about 350 kA/cm$^2$ on target. We have used the measured proton energy time-history for this shot with the expectation that it is a better standard than the corrected diode voltage. We have assumed that the ion beam accelerated from the anode plasma is comprised of equal numbers of protons and carbon. Moreover, we assume that the carbon portion of the beam is comprised of equal numbers of $\text{C}^2$ and $\text{C}^3$ ions. These assumptions about ion beam species are consistent with published results [22] as well as recent experiments [23]. We note that no significant $\text{C}^4$ fraction has been seen in any of these experiments.

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Referring to Fig. 5, we see that experimental output beam shows significantly enhanced proton stopping throughout the time history of the shot (compare to neutral atom curve). The proton only simulation, curve 1, with $j = 350$ kA/cm$^2$ underpredicts the enhancement. Addition of the carbon component, curve 2, of the beam brings the simulation into better agreement. Best agreement is found for a 500 kA/cm$^2$ proton beam with an equal carbon component, curve 3. The disagreement at early times may be partly due to uncertainty in the timing measurement for this analyzer bias-setting that was centered on later times. Late-time disagreement is probably due to 2-D motion of the target. That is, the probe beam traverses a decreasing mass-line-integral at later time as the finite axial velocities of material at the focus carries mass out of the field of view. This would manifest itself as a diminished energy loss in the lower areal mass target. We will corroborate this assertion with 2-D hydro-simulations. More experiments need to be done in this geometry with aluminum targets, perhaps in conjunction with other diagnostics, to develop a consistent source definition. Then additional experiments using
different stopping targets (e.g., gold) can be performed to expand our data base to higher-Z elements.

Acknowledgements

The authors wish to acknowledge support and helpful discussions from J. A. Swift, J. R. Lee, D. J. Johnson, and E. J. T. Burns. One of the authors (T.A.M.) is especially appreciative for the preprint of Ref. 10 provided by Prof. Y. Sayasov currently at the Kernforschungszentrum, Karlsruhe, West Germany concerning the Local Field correction to the Random Phase Approximation.

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Ion Stopping in Heated Targets

J. N. Olsen, T. A. Mehlhorn, D. J. Johnson, P. L. Dreike, and L. P. Mix

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Abstract

Ion stopping departs from classical, cold material values as the target material heats to appreciable ionization levels. We are investigating this phenomenon experimentally on the Proto I accelerator with a radial, Applied-B field ion diode. When the beam is focused to 0.5 TW/cm² on thin aluminum foil targets the time-resolved departure from cold stopping is clear. Our observations and early interpretations will be presented.

Introduction

In ion beam driven inertial confinement fusion (ICF) the target will be bombarded with an intense beam of ions. During the power pulse the ion-absorbing layer will reach progressively higher ionized states. With the ionization process, however, there will be a change in the ion stopping as the free electrons interact more strongly and the remaining bound electrons interact less strongly with the incoming ions. Design of ICF targets will require adjustment for the temperature-dependent stopping of the beam. To be able to do that will require accurate modeling of the interaction dynamics of both free[1] and bound electrons[2,3] with the ions. These problems are compounded by the hydrodynamics, ionization and radiation flow processes that are occurring at the same time. Clearly, confirmation of model predictions at progressively higher beam powers in well-characterized experiments will be an important part of understanding the mechanisms.

The first of these experiments[4] has been reported recently. In those experiments at the Naval Research Laboratories, beams of deuterons were focused onto either Mylar or aluminum foils at intensities up to 0.3 TW/cm². Stopping power was measured by observing the time-of-flight of neutrons from a CD₂ layer behind the target. This type of measurement gives the energy loss at one point in time, essentially at peak power. Departures from cold stopping were seen for both the Mylar and aluminum foils.

The experiment reported here differs in two respects. First, with the Applied-B field ion diode[5] on the Proto I accelerator higher ion beam intensity, 0.5 TW/cm², can be attained. This should advance the measurement to an average ionization T = 5-6, stripping aluminum to oxygen-like atomic levels. The correct modeling of the bound electrons is critical and the experimental data are expected to be very helpful. Furthermore, the energy loss measurements are performed with an analyzer recently developed for PBFA voltage measurements.[6] With this device we obtain a time-resolved stopping power, even observing the time of departure from cold stopping during the power pulse.

The Applied-B field diode will be discussed only briefly since it has been described in detail elsewhere.[5] We will emphasize the deposition experiment and the data that is now available. Some preliminary comparisons with stopping power simulations will also be presented.

The Applied-B Field Diode

Referring to Fig. 1, the diode is essentially a short barrel with 4.5 cm radius. The cathode structure has windings for the externally applied axial magnetic field. Protons from the flashover anode are accelerated toward a virtual cathode formed by electrons trapped in the vertical field lines. After traversing this A-K gap the protons

Fig. 1. Schematic of the Applied-B field ion diode.
enter a gas cell that provides for charge- and current-neutralized ballistic propagation to the on-axis target. The Argon gas fill is puffed in to about 1.7 J. Typically, the voltage peaks at 1.7 MV and falls during the time of the 0.4 MA peak diode current. Previous measurements show that at peak current only 10 percent of the current is lost to electron flow. Peak ion power is thus 0.4 - 0.5 TW.

New, very detailed characterization of the focused intensity of this beam is given elsewhere in these proceedings. The focused proton intensity is 0.5 TW/cm² on a surface of 3 mm diameter and 3 mm height. Proper focusing to this level requires careful tuning of the diode voltage and current, as well as optimized gas puff operation. The analysis of Lee and Leeper in Ref. 7 establishes that the peak current density on target is 0.35 KA/cm².

**Ion Stopping Apparatus**

Due to the mechanical constraints the ion stopping experimental apparatus must be designed as a pre-aligned unit. We chose to solve this problem, and to obtain time-resolved data as well, by adapting a new Thomson parabola analyzer to this configuration. Energy analysis is performed in a region of parallel electric and magnetic fields as sketched in Fig. 2. Time resolution is obtained in this design by applying a ramped, rather than static, electric field to the permanent magnetic plates. In Fig. 2 one quadrant of the radially focusing proton beam strikes a target sled, which is composed of two gold scattering foils and a deposition foil. The protons that are Rutherford-scattered from the slanted foils are collimated in two pairs of 0.25 mm apertures before entering the analyzer. The beam from the first foil is designated as the input beam and that passing through the deposition foil to the second scatterer is the output beam. An advantage of this arrangement is that both beams are analyzed in the same apparatus, thereby avoiding several sources of relative errors. The beams are recorded on a disc of CR-39 track recording plastic, which is etched in NaOH to show the proton impact spots.

The method of analysis is as follows: In the known magnetic field, the B-deflection, y, is

\[ y = 0.95/\sqrt{E} \text{ (in cm)} \]  

where E is the proton energy in MeV. The electric deflection, x, due to the pulsed voltage, V(t) in kV, is

\[ x = V(t)/16E \]  

The proton energy is taken from Eq. 1 and used in Eq. 2 to determine the V(t) at the time of arrival at the analyzer. Knowledge of V(t) then gives E as a function of time. Finally, time-of-flight corrections are made back to the A-K gap for each of the input and output beams. Energy losses in the scattering, deposition, and gas cell foils are added back to E(t) according to tabulated cold stopping powers. If cold stopping holds, the diode voltages inferred from the two beams agree with each other. Enhanced stopping due to target heating is seen when the output beam energy falls below the input beam energy, both referred to the A-K gap.

**Ion Stopping Data**

The target shown in Fig. 2 placed the deposition foil on axis; no enhanced stopping was found in that configuration because best focus of the beam is at approximately 1.5 mm radius. Correspondingly, our best data was obtained with the target sleds of Figs. 3 and 4. We will describe these targets now and present the analyzed data.

The sled of Fig. 3 placed the deposition target 3 mm off-axis (left side of sled) and the output beam, labelled "a," was distinct from the input beam "b." In the variation shown here the input beam passed through a moderator foil and stopped in the deposition foil, adding to the heating of that
The deposition foil is 3 mm off-axis. The beams at the plastic are also sketched. The data can be analyzed because the a and b beamlets predominate in the final record. Alternatively, the moderator foil was replaced with a beam stop: In that case no enhanced stopping occurred.

The final sled configuration is shown in Fig. 4. In this variation the deposition target is rolled into a 1.5 mm radius cylinder and attached directly to the scatterer. Thus positioned, the target is at the best focus of the proton beam.[5,7] The beam stop or moderator feature is retained.

The beam energies, referred to the A-K gap, for the flat target with moderator are shown in Fig. 5. The input beam agrees in peak voltage and fall-time with the inductively corrected diode voltage. The output beam has heated and penetrated a 7.6 μm thick aluminum deposition foil. The peak voltage, inferred using cold stopping in the aluminum, agrees with the input beam but quickly deviates to show enhanced deposition. The change of deposition amounts to as much as 250 keV beyond the amount calculated from tabulated values.

A similar shot with the cylindrical target is shown in Fig. 6. In this case the beam stop was placed between the input and output scatter foils. As in the previous case the peak input beam energy agreed with the output beam but quickly showed enhanced deposition. The additional energy loss was as high as 350 keV.

Discussion

We are comparing these loss measurements with calculated losses obtained from modeling of free electron[1] and bound electron[2,3] interactions with the incoming protons. A possible difficulty would be the estimation of the heating of the target in the focused ion beam. We are able, however, to
rely upon previous experiments that have 1) charac-
terized the target temperature [5] and 2) established
the focused intensity. [7] Details of the target simu-
lation become less important as long as the incident
intensity and the resulting target temperature are
close to the observations. A further easing of the
problem is provided by a diagnostic geometry that
only accepts ions scattered from the axis of the
diode. Thus, although grazing incidence ions are
important for target heating, they do not contribute
to the sampled beam.

A preliminary comparison of the computed and
measured ion energies is given in Fig. 7. This
calculation was performed for the case of a normally
incident proton beam on a single side of a planar
aluminum target. The top curve is the ion input
beam from the diode voltage which is used as an
input to the deposition calculation. [9] The lower
curves are the expected output particle energies
for cold stopping and heated stopping. In these
d detailed in connection with Figs. 5 and 6 (shown as diamonds).

Fig. 7. Comparison of computed and measured beam
calculations of ion energies are given for each of six tests, four with
no enhanced stopping and two as detailed
in these proceedings.

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sleds and J. Heise and J. Gergel ran the Proto I
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acknowledge helpful discussions with F. C. Young of
Naval Research Laboratories. This work was sup-
ported by the U. S. Department of Energy under
contract DEAC04-76-DP00789.

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Young, in these proceedings.
Time-Dependent Charge State Calculations

D. Bailey, Y. T. Lee and R. M. More

Lawrence Livermore National Laboratory
Livermore, California
Where the stopping numbers $B$ as well as the ionization fraction depend on the $\rho$, $T$ of the plasma. To calculate a pellet implosion using an ion beam driver, we need to evaluate this expression over a wide range of $\rho$ and $T$. Note that all data is for cold matter and solid density usually, so that there is only a single check point in $\rho$, $T$ space.
• BOUND-BOUND EXCITATION & DE-EXCITATION
• BOUND-FREE IONIZATION

ION RATES

B. BOUND-BOUND EXCITATION & DE-EXCITATION

(3-BODY RECOMBINATION)

ELECTRON RATES

BOUND-BOUND EXCITATION & DE-EXCITATION

BOUND-FREE IONIZATION

BOUND-FREE IONIZATION

BOUND-IONIZATION

BOUND-IONIZATION

BOUND-IONIZATION

BOUND-IONIZATION
Improved screening coefficients

\[ E_n = -13.6 \text{ eV} \frac{Q_n^2}{n^2} + E_n^0 \]

\[ Q_n = Z - \sum_m \sigma_{nm} P_m \]

Pressure ionization and continuum lowering +

degeneracy of free electrons

\[ P_n = \frac{2n^2}{1 + \exp (E_n - \mu)/kT} \]

OR use rate equations for non-LTE

\[ \kappa_{BF} = 11.9 \frac{1}{A \nu^3} \frac{Q_n^4 P_n}{n^5} \]

\[ \kappa_{BB} = 6.6 \times 10^4 \frac{f_{nm}}{A} P_n (2m^2 - P_m) I_{nm}(\nu) \]

Energies obtained from consistent thermodynamic formulas
The diagram illustrates the ionization cross section (cm$^2$) as a function of electron energy (keV). The data points represent the reaction $e + Au \rightarrow 2e + Au^+$ (E.J. McGuire) and the theoretical curve from Lotz's formula $P + Au \rightarrow P + e + Au^+$.

- Electron energy (keV) range from $10^{-2}$ to 1.
- Ionization cross section range from $10^{-17}$ to $10^{-15}$ cm$^2$.
Ne = 6 \times 10^{23} \text{cm}^{-3}

E_{\text{ion}} = 46 \text{ MeV/Amu}

Electron - Ion
Equilibrium charge ($Z^*/Z$)

Ion velocity ($v/c$)

$\triangle$ Si

$\bigcirc$ Ca

$\{ \text{Betz empirical formula} \}$
$E_{\text{ION}} = 46 \text{ MeV/amu}$

Target = 0.25 g/cm$^2$
EQUILIBRATION DISTANCE (IN UNITS OF COLD RANGE)

<table>
<thead>
<tr>
<th>Element</th>
<th>Distance</th>
</tr>
</thead>
<tbody>
<tr>
<td>Au</td>
<td>0.16</td>
</tr>
<tr>
<td>Ag</td>
<td>0.1</td>
</tr>
<tr>
<td>Cu</td>
<td>0.025</td>
</tr>
<tr>
<td>Ca</td>
<td>0.01</td>
</tr>
<tr>
<td>Si</td>
<td>0.005</td>
</tr>
</tbody>
</table>
\( \delta \)-function electron spectrum means rate integrals are just proportional to cross-sections.

For electron impact collisions, \( E \sim l_n \) and virial theorem \( \Rightarrow 1/2m_e v_{\theta n}^2 = l_n \) or
\[
1/2m_e v_p^2 = 1/2m_e v_{\theta n}^2 \Rightarrow v_p \sim v_{\theta n}
\]
- Low density (gas phase) \( v_p \sim v_{\theta n} \)
  
  Excited ions decay to ground state between electron impacts

- High density (2-step ionization)
  
  2 electrons/ionization \( \Rightarrow 2E = l_n \)
  
  \[ v_{\theta n} \]
  
  or \( v_p \sim \frac{v_{\theta n}}{2} \)
  
  Excited ions are ionized before radioactive decay can occur

- Ion impact collision: 2 body kinematics \( \Rightarrow E_{\text{max}} = 2 m_e v_p^2 \)
  
  \[ v_{\theta n} \]
  
  or \( v_p = \frac{v_{\theta n}}{2} \)

- "Coronal" type equilibrium, but not with maxwellian spectrum.
  
  \( \delta \)-function both exciting and recombining ions are the same
Universal Equilibrium Charge Functions

Equilibrium $Z^*/Z$

- Betz $f(v)$ (solid)
- Betz $f(\sqrt{2v})$ (gas)

Projectile velocity $v/c$
Ionization state, $Z^*$

$Z = 79$

$n_e = 6 \times 10^{17} \text{ cm}^{-3}$

$1.8 \text{ MeV/amu}$

Low density (~gas)

time dependent ionization
\[ E = 1.8 \text{ MeV/amu} \]

<table>
<thead>
<tr>
<th>Ne</th>
<th>Au</th>
<th>Ag</th>
</tr>
</thead>
<tbody>
<tr>
<td>No BB</td>
<td>30.1</td>
<td>22.3</td>
</tr>
<tr>
<td>$6 \times 10^{17}$</td>
<td>30.9</td>
<td>24.1</td>
</tr>
<tr>
<td>$6 \times 10^{20}$</td>
<td>45.4</td>
<td>34.1</td>
</tr>
<tr>
<td>$6 \times 10^{22}$</td>
<td>49.1</td>
<td>36.1</td>
</tr>
</tbody>
</table>
Monte Carlo Simulation of Electron-Photon Showers

M. Cha, O. Goktepe and N. Rao

Naval Surface Weapons Center
Silver Spring, Maryland
# E-8 Shower Code Calculations

## Benchmark Target 'A'

<table>
<thead>
<tr>
<th>Code</th>
<th>$E_0 \text{(MeV)}$</th>
<th>Reference</th>
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</thead>
<tbody>
<tr>
<td>SA1 EPSC</td>
<td>200/500</td>
<td>NSWC TR 76-23</td>
</tr>
<tr>
<td>LLNL Handy</td>
<td>200/500</td>
<td>LLNL UCRL-52871</td>
</tr>
<tr>
<td>NSWC EGS 3</td>
<td>200/500, 500/1000</td>
<td>NSWC TR 84-104</td>
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</table>

## Benchmark Target 'B'

<table>
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<tr>
<th>Code</th>
<th>$E_0 \text{(MeV)}$</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>LLNL Handy</td>
<td>500</td>
<td>LLNL D 79-14</td>
</tr>
<tr>
<td>NSWC EGS 3</td>
<td>200/500</td>
<td>NSWC TR 84-106</td>
</tr>
</tbody>
</table>
Simplified Geometry Configuration

Fig 1
Benchmark Target 'A' (90g/cm²)

Air
Gaps

\[
\begin{aligned}
\text{\( \sim 46 \text{ cm} \)} \\
\text{\( 58.69 \text{ cm Air} \)} \\
\text{\( \frac{1}{4} \text{" Plastic - TLD #5} \)} \\
\text{\( \frac{1}{4} \text{" Al} \)} \\
\text{\( 5.13 \text{ cm Air} \)} \\
\text{\( \frac{1}{2} \text{" Fe} \)} \\
\text{\( \frac{1}{2} \text{" Plastic - TLD #6} \)} \\
\text{\( \frac{3}{8} \text{" Lucite} \)} \\
\text{\( \frac{1}{4} \text{" Plastic - TLD #7} \)} \\
\text{\( \frac{2}{4} \text{" Lucite} \)} \\
\text{\( \frac{1}{4} \text{" Plastic - TLD #3} \)} \\
\text{\( \frac{1}{4} \text{" Plastic - TLD #2} \)} \\
\text{\( \frac{1}{2} \text{" Al} \)} \\
\text{\( 7.89 \text{ cm Air} \)} \\
\text{\( \frac{1}{4} \text{" Plastic} \)} \\
\text{\( \frac{1}{2} \text{" Al} \)} \\
\text{\( 8.25 \text{ cm Air} \)} \\
\text{\( \frac{1}{4} \text{" Al} \)} \\
\text{\( \frac{1}{4} \text{" Plastic - TLD #1} \)} \\
\text{\( 40.86 \text{ cm Air} \)} \\
\text{\( \frac{1}{4} \text{" Al} \)} \\
\text{\( 30.56 \text{ cm Air} \)} \\
\text{\( 2\frac{1}{2} \text{" Al} \)} \\
\end{aligned}
\]
Fig 2 Benchmark Target 'B' (72 g/cm³)
LONGITUDINAL DOSE PROFILES

Fig 3 Longitudinal Profile Comparison for HANDYL76 and EPSC on Target 'A'
Fig 4  Comparison of Target 'A' Results
Comparison of Radial Profiles

(cf LLNL D79-14)
Fig. 4.6
Comparison of Radial Profiles (cf. LLNL D79-14)
Fig 5  Comparison of Target 'A' Results
Fig 5a  Energy Deposition, Target 'A', Position 6
Fig 6  EGS 3 results for energy deposition in Target 'A'

On-Axis Energy Deposition (MeV/g/cm²)

Longitudinal Distance (g/cm²)
Fig 7
EGS 3 results for 500 MeV electron on benchmark targets

Dots represent calculated values
Fig 8 Comparison of Target B results, 500 MeV

On-axis Energy Deposition (MeV/g/e)

Longitudinal Distance cm
Summary Remarks

- EGS-3 self-consistent with respect to front surface ionization energy loss
- Fair (dis)agreement among the codes (EGS 3, EPSC, HANDYL) and experiments
- Statistics good for moderate thicknesses, e.g. 1000 histories @ 500 MeV @ 20 g/cm²
- Geometry extremely important, i.e. voids
- Codes versatile but costs limit results
- More calculations and experiments needed
The Beam-Density Effect on Energy Loss by a Charged Particle Beam

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The beam-density effect that I will describe has its origin in the vicinage effect which modifies the energy loss of ion-clusters passing through solids. The investigations of vicinage effects and of the related phenomenon of the wake created behind a charged particle traveling in a solid, were pioneered by Werner Brandt, Rufus Ritchie and their coworkers.¹ McCorkle and Iafrate² used the vicinage concept as the basis of their treatment of the beam-density effect and they predicted a surprisingly large enhancement of the beam energy loss to a background plasma. Conceptually, the theory which I am going to discuss is the same as that of McCorkle and Iafrate; however the details of the realization of these concepts and the resulting conclusions are quite different from their results. Some of these results appear in a recent paper by Oakley Crawford and myself.³

In an earlier presentation, Tom Mehlhorn described a very nice set of experiments by Frank Young, as well as his own calculations,⁴ which demonstrated that there is a beam enhancement of energy loss caused by the increased stopping power of a target which becomes highly ionized as the beam enters it. Essentially, the later part of a long, nonrelativistic, high-current pulse of deuterons encounters a target which has been highly ionized by the earlier part of the pulse. This is an incoherent beam effect on single-particle energy loss, since it doesn't depend in an essential way on the details of internal structure of the beam, such as beam particle separation, but it does depend on the change in the ionization state of the
medium through heating by the beam. In contrast, the beam-density effect does
depend on the detailed structure of the beam, including its shape, through the
vicinage function which depends on both the beam particle separations and the
medium properties. The latter effect can then be considered as a coherent
energy loss enhancement mechanism. These coherent and incoherent energy loss
effects can be thought of as different extremes of a general self-consistent
treatment of beam energy loss. As we have seen, the target heating effect is
important for nonrelativistic beams where the stopping power of cold materials
is relatively high and an intense beam can rapidly create a dense plasma. In
contrast, the coherent beam-density effect is important for relatively weakly
ionized targets which can occur when an intense relativistic beam interacts
with material, since stopping powers of materials for relativistic beams are
orders of magnitude lower than in the case of non-relativistic beams.
Depending on the beam's current density, there will be a critical value of
background plasma density at which the beam's particles are effectively
screened from each other so that they cannot interact cooperatively with the
medium. At this plasma density, the coherent beam-density effect becomes
negligible; however, if the background plasma density increases still further,
the incoherent beam heating effect becomes important and the energy loss is
again enhanced in comparison to cold target values.

The approach that I have used in formulating the beam-density effect has
three parts: the target medium was modeled with an appropriate response
function; a relativistic theory of the vicinage function for pairs of beam
particles was developed; and finally the vicinage function was integrated over
a simple model for a beam pulse. A Lorentz dielectric function $\varepsilon(\omega)$ was used
to represent the medium. The origin of the vicinage terms for energy loss by
clusters can be quickly reviewed by starting with the classical expression for energy loss per unit length:

\[ W = \frac{1}{2\pi} \int_{-\infty}^{\infty} db \int_{0}^{2\pi} d\phi \int_{0}^{\infty} \omega \, \text{Im} \varepsilon(\omega) \left| E(\omega, b) \right|^2 d\omega, \]  

(1)

where \( b \) is the impact parameter and \( E(\omega, b) \) is the electric field Fourier transformed with respect to time. The minimum impact parameter \( a \) can be chosen so that for single particles, \( W \) reproduces the Bethe form of ionization energy loss. This compensates for the fact that \( \varepsilon(\omega) \) is really only valid for distant, low-momentum collisions, since it doesn't take into account spatial dispersion through a \( k \)-dependence.

For a cluster of particles, one can decompose the field into terms with each particle as a source, thus obtaining

\[ \left| E \right|^2 = \sum_{i=1}^{n} \left| E_i^+ \right|^2 + 2 \text{Re} \sum_{i < j=1}^{n} E_i^+ \cdot E_j^-, \]  

(2)

In the case when \( n = 2 \), using (2) in (1) gives

\[ W = W_1 + W_2 + W_{12}, \]  

(3)

where \( W_1 \) and \( W_2 \) are the usual single-particle energy loss terms and the vicinage term \( W_{12} \) comes from the second term on the right of (2). For a relativistic pair of particles with charges \( Z_1e \) and \( Z_2e \), it can be shown that

\[ W_{12} = \frac{-4}{\pi} Z_1 Z_2 \frac{e^2}{v^2} \int_{0}^{\infty} \omega \cos(\frac{\omega z}{v}) \text{Im} \left( \varepsilon^{-1}(\omega) - \beta^2 \right) K_0\left( \frac{\omega B}{v} \right) \, d\omega, \]  

(4)
where $B_\gamma = \text{Max}(B, a)$. In (4), particles 1 and 2 are separated by the radius vector $\hat{R} = z \frac{\hat{v}}{v} + B \hat{e}_\perp$, where $\hat{e}_\perp$ is a unit vector perpendicular to the velocity. I have assumed that the velocities of the particles are equal.

The factor $\Gamma$ in the argument of the modified Bessel function $K_0$ is defined as $\Gamma = (1 - B^2 \epsilon(e))^{-1/2}$ and is familiar from the Fermi density effect. Note that from the form of $W_{12}$,

$$\lim_{R \to 0} W = (Z_1 + Z_2)^2 S, \quad (5)$$

where $S$ is the single-particle ionization energy loss for a particle of unit charge. This is the expected result when a pair of charged particles coalesces.

If the nonrelativistic limit of (4) is considered, the vicinage function $W_{12}$ can be analyzed in terms of a resonant part $W_R$ which comes from the poles of $\epsilon^{-1}$ in the complex $\omega$-plane, and the nonresonant remainder $W_N$:

$$W_{12} = W_R + W_N. \quad (6)$$

The resonant contribution is just

$$W_R = 4Z_1Z_2 e^2 \left(\frac{\omega p}{v}\right)^2 \text{Re} \sum_{j=1}^{n} \frac{\alpha_j}{\alpha_j - \beta_j} \exp\left(-\frac{i\omega p}{v}\right) z \left| a_j \right| K_0\left(\frac{B\omega}{v} \alpha_j\right). \quad (7)$$

The frequency $\omega_p$ is defined by

$$\omega_p^2 = 4\pi Ne^2/m_e. \quad (8)$$
where \( N \) is the total electron density of the material, including both bound and free electrons. The poles of \( \varepsilon^{-1} \) occur at \( \omega = \alpha_j \omega_p \) and \( \omega = \beta_j \omega_p \) which are in the fourth and third quadrants, respectively. Further details about \( \varepsilon(\omega) \) are given in Ref. 3.

The nonresonant remainder can be expressed as an asymptotic series,\(^3\) the lowest term of which is given by

\[
W_N = 8Z_1 Z_2 e^2 \left( \frac{\omega_p^2}{\nu} \right) \text{Re} \sum_{j=1}^{n} f_j \frac{i \alpha_j}{\alpha_j - \beta_j} \left( \frac{\nu}{\alpha_j \omega_p} \right)^3 P_2(\cos \theta) R^{-3},
\]

where \( R = (z^2 + B^2)^{1/2} \) and \( \cos \theta = z/R \). If \( z < 0 \) and \( B > a_s \), \( W_R \) takes the form of the standard, classical, single-particle ionization energy loss (see Eq. (5)). The nonresonant terms are then usually small and are therefore properly neglected in that case.

Since the poles of \( \varepsilon^{-1} \) at \( \omega = \alpha_j \omega_p \) lie in the fourth quadrant, it can be seen that \( W_R \) has the form of a damped cosine function of \(|z|\) times the modified Bessel function which has the asymptotic dependence of \( x^{-1/2} \exp(-x) \). Thus \( W_R \) is short ranged and decays exponentially with a scale set by \( \nu/|\alpha_j \omega_p| \), where \(|\alpha_j \omega_p|\) is roughly equal to the eigenfrequency of the \( j \)th oscillator used to model \( \varepsilon \). In contrast, \( W_N \), the nonresonant contribution, is dipole-like to lowest order for large \( R \). Thus it is very long ranged relative to \( W_R \) and, provided \( \nu/|\alpha_j \omega_p| \) is large enough in comparison to the beam-particle separations, \( W_N \) can make a large contribution to beam energy loss, as I will show. The beam shape dependence which will be discussed also arises from the angular dependence of \( W_N \) contained in the Legendre polynomial \( P_2(\cos \theta) \). Note that an angle average of \( W_N \) would yield zero; therefore, angle...
averaging the vicinage function $W_{12}$ before integrating over a finite beam will yield misleading results for calculations of beam energy loss.

The vicinage function $G = W_{12}/2S$ is shown in Fig. 1 in the case of the conduction electrons of aluminum. The plasmon energy was taken to be 15 eV and the plasmon width used was 1 eV. Note that the transverse and longitudinal distances are scaled by $v/\omega_p$, where $\hbar \omega_p = 32.78$ eV, and is not the plasmon energy (see Eq. (8)). This vicinage function has the qualitative features of the one calculated by George Basbas and Rufus Ritchie\(^6\) using a $k$-dependent dielectric function.

The vicinage function $W_{12}$ of (4) has been used to determine the beam-density effect on energy loss by both relativistic and nonrelativistic beams in weakly ionized gases.\(^3\)\(^7\) For this purpose, a model beam was chosen with a Gaussian radial profile and a step-function current rise. If a test beam particle lies on the beam axis, the total energy loss of this particle consists of the usual single-particle loss $W_s$ plus the beam contribution:

$$W = W_s + W_B,$$

where

$$W_B = \frac{1}{2} \int \int_{-\infty}^{\infty} dz \int_{0}^{2\pi} dB \int dB W_{12} (z, B) n_b e^{-\left(\frac{B}{B_0}\right)^2} \Theta(L - z).$$
Here the beam density is \( n_b \), the Gaussian radial parameter is \( B_0 \), and the Heavyside step function \( \Theta(L - z) \) is appropriate for a test particle a distance \( L \) from the beam head.

The cooperative beam energy loss \( W_B \) has been shown to have a peculiar behavior when the limiting cases of infinitely long and infinitely wide beams are considered. If the beam radius parameter \( B_0 \) is kept constant and the limit \( L \to \infty \) is taken, i.e. we have an infinitely long beam, then

\[
\lim_{L \to \infty} W_B = 0. \tag{12}
\]

On the other hand, for constant \( L \gg v/|\sigma J_B| \), if we let the beam radius become infinite,

\[
\lim_{B_0 \to \infty} W_B = \text{constant}, \tag{13}
\]

where the above constant is \( eJ_B/\sigma \) for conduction electrons, \( J_B \) being the beam current density and \( \sigma \) the conductivity. Eqs. (12) and (13) mean that

\[
\lim_{L \to \infty} \lim_{B_0 \to \infty} W_B \neq \lim_{B_0 \to \infty} \lim_{L \to \infty} W_B. \tag{14}
\]

Thus the concept of an infinite beam is not a useful one when considering the behavior of \( W_B \) unless one specifies the limiting procedure used to achieve the infinite beam. The ratio \( L/B_0 \) during the limit taking must be specified. This surprising result does not seem to be an artifact of the particular
current distribution chosen, but seems to be generally true and traceable to
the long-range, angular-dependent, nonresonant part of the vicinage function,
$W_N$ Eq. (9).

In Fig. 2 the result of a nonrelativistic calculation of $W_B$ is shown for
a 5 MeV, 10 kA proton beam in weakly ionized $H_2$ gas at 1 atmosphere
pressure.\(^3\) The Gaussian beam radius parameter was taken to be 1 mm, giving
$n_b = 6.44 \times 10^{14}$ cm$^{-3}$. The ratio $W_B/W_S$ is plotted versus the distance of a
test beam particle from the beam front. Two different values of the free
electron density $n_e$ were assumed. In the case of $n_e = 1.11 \times 10^{16}$ cm$^{-3}$, $W_B$
was found to be oscillatory near the beam front, with a peak value of about 20
times $W_S$. These oscillations come from the resonant part $W_R$ of the vicinage
function Eq. (8). When $n_e = 1.11 \times 10^{15}$ cm$^{-3}$ was used, the nonresonant term
$W_N$ gave the dominant contribution to $W_B$ even near the beam front; therefore $W_B$
was then always positive and had a peak value of about 60 times $W_S$.

In Fig. 3 the ratio $W_B/W_S$ is shown for a relativistic electron beam in
weakly ionized nitrogen at 1 atmosphere.\(^7\) The beam parameters were 50 MeV,
10 kA, $B_0 = 1$ mm, and a pulse length of 1 meter. In this case, the ratio
$W_B/W_S$ varied from about 10 to 1000 as $n_e$ ranged from $10^{17}$ to $10^{15}$ cm$^{-3}$. Even
50 cm into the pulse, $W_B$ was roughly twice $W_S$. In all three cases, $W_B$
was positive everywhere although the resonant contribution to $W_B$ caused a ripple
near the beam front in the case of $n_e = 10^{17}$ cm$^{-3}$.

In summary, the results that I have presented illustrate that the
nonresonant terms in the vicinage function play a major role in determining
its long range behavior. These terms have a dipole-like angular and radial
dependence and thus are dominant at large particle separations over the resonant part of the vicinage function, which has an exponential fall off. The nonresonant terms give rise to a beam shape dependence of the beam-density effect on energy loss. The beam-density contribution to energy loss by intense beams in weakly ionized gases was found to increase energy loss by several orders of magnitude in the cases examined. Similar results are expected in the case of an intense beam interacting with the transient conduction electrons which it generates as it interacts with an ionic solid.

This theory of the beam density effect includes the Fermi density effect on ionization energy loss by beams, the Cherenkov component of loss by beams (if a suitable dielectric function is used), Ohmic drag on beams by fields induced in a conducting medium, as well as single-particle collisional energy loss and vicinage effects on energy loss. These effects may all be considered as different aspects of the inductive energy loss caused by the interaction of a current with the electric field that it induces in the medium. The next step in developing a more realistic theory of beam-material interactions is to include the modification of the target medium properties as the beam interacts with it and also to include the dynamical effects on the beam of self-fields and induced fields as the beam traverses the medium. The goal is a self-consistent theory.
References


Figure Captions

Figure 1 The two-particle vicinage function for particles with velocity equal to twice the Bohr velocity interacting with the conduction electrons of aluminum.

Figure 2 The ratio of the beam-density contribution $W_B$ to the single-particle ionization energy loss $W_S$ vs the distance of a test beam particle from the beam front. The curves are for a 5 MeV proton beam with beam density of $n_b = 6.44 \times 10^{14}$ cm$^{-3}$, which is interacting with partially ionized $H_2$ containing $n_e$ free electrons per cubic centimeter.

Figure 3 The ratio of the beam-density loss $W_B$ to the single-particle ionization energy loss $W_S$ vs distance from the beam front for a 50 MeV, 10 kA electron beam pulse of 1 meter length in nitrogen at 1 atmosphere. The density of free electrons assumed is $n_e$. The beam current density was $3.18 \times 10^5$ A/cm$^2$ giving a beam density of $n_b = 6.6 \times 10^{13}$ cm$^{-3}$. 
VICINAGE FUNCTION G FOR A DICLUSTER IN ALUMINUM V=2V₀

Fig. 1
The ratio per particle of beam to single-particle energy deposition \( \frac{W_B}{W_S} \) in weakly ionized hydrogen.

**Fig. 2**

- **Analytic Approximation**
- **Exact**
- \( n_e = 1.11 \times 10^{15} \text{ cm}^{-3} \)
- \( n_e = 1.11 \times 10^{16} \text{ cm}^{-3} \)

- \( W_S = 183 \text{ MeV/g-cm}^{-2} \)

- **E = 5 \text{ MeV protons}**
- **I = 10 \text{ kA}**
- **B = 1 \text{ mm}**
- **P = 1 \text{ Atm}**
THE RATIO PER PARTICLE OF BEAM TO SINGLE-PARTICLE ENERGY DEPOSITION, $W_B/S$, IN WEAKLY IONIZED NITROGEN

$E = 50$ MeV
$I = 10$ KA
$b_0 = 1$ mm
$L = 1$ m
$P = 1$ Atm

Fig. 3
Electron Transport and Transient Conductivity of Irradiated Insulators

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Electron Transport and Transient Conductivity of Irradiated Insulators

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Irradiation of an insulator by an intense electron beam causes a large transient increase in the conductivity, due to ionization. In this talk, the development of a theory of this effect and comparison with experiments are described. Predictions of the scaling of transient conductivity over a wide range of beam current are made on the basis of present understanding, and issues for future work are presented.

A detailed treatment of ionization and excitation leads to an electron energy distribution in the irradiated medium, Fig. (1), from source energy down to about 10 eV. This calculation is for NaCl. A treatment of electrons at lower energies requires consideration of the band structure of the medium, for which we use the model shown in Fig. (2). In it, the conduction electrons behave as free electrons. Note that electron energy is measured from the bottom of the conduction band.

Electrons with energy below the gap energy, $E_g$, lose energy by interaction with phonons only. The rates of energy loss to longitudinal optical phonons (by Fröhlich's model) and to longitudinal acoustical phonons (by the deformation potential interaction) are shown in Fig. (3) as functions of electron energy. These models contain an upper limit (the Brillouin zone radius) on the change in electron wave number in electron-phonon scattering. This affects the scattering for energies $\gtrsim 2$ eV, leading to the discontinuity in slope shown in the figure.
The Langevin theory of recombination is used to describe the rate at which electrons disappear from the conduction band.

Using the rates of ionization, energy loss, and recombination described above, one calculates the steady-state energy distribution. This is shown in Fig. (4) for a 4.5 MeV beam of 1 kA/cm$^2$ current density. Note that the distribution divides itself into two groups, the thermal and the epithermal electrons. The electron densities of these two classes of electrons separately, as well as the combined density, are shown as functions of beam intensity in Fig. (5).

The models discussed above for electron-phonon scattering lead to collision frequencies which are given as functions of electron energy in Fig. (6). The three curves are for scattering by optical and by acoustical modes, and for the total rate. At energies $\lesssim 0.1$ eV, the total rate has been reduced by a saturation correction so that the mean free path does not exceed the lattice constant.

The predicted steady state conductivity of irradiated NaCl is shown in Fig. (7) as a function of beam current density. Also shown are the separate contributions of the thermal and epithermal groups of electrons. Vaisburd and coworkers found a 0.4 power law in the conductivity vs beam current (Fig. 8), in the range of 1 to $10^3$ A/cm$^2$, while the present theory gives a 0.5 power dependence in the same regime. It is unclear what the experimental accuracy is, and this difference might well be within experimental error.

The predicted evolution of the conductivity with time is illustrated in Figs. (9) and (10). In the former, where a 0.1 ns beam pulse is assumed, the response time of the conductivity is seen to be on the order
of 10 ps (assuming a 1 kA/cm$^2$ 4.5 MeV beam) which is consistent with the
observations of Vaisburd et al. We must point out, however, that the
decay of the conductivity is not exponential, and has a long tail, so
the concept of response time should be treated with caution. Another
important point is that the response time decreases with increasing beam
current density. This is because the density of holes goes up, thereby
decreasing the lifetime of thermal electrons. This is why the conductivity
response time discussed here is so much shorter than the ones observed
in classical (low-current) photoconductivity measurements.

The conductivity during and after a 20 ns beam pulse is shown in
Fig. (10). On this time scale, the conductivity is predicted to
decrease somewhat over the duration of the pulse, due to heating of the
medium.

We conclude that the theory is consistent with experiments, and
explains the nonlinear dependence with beam current density, and the
fast response. Many important issues remain for future work. Several
of them are listed in Fig. (11).

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Electric Conductivity of Polyethylene During Pulsed Electron-Beam Irradiation at ETA

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The letters "ETA" stand for "Experimental Test Accelerator," the facility at Lawrence Livermore National Laboratory that we used to do this experiment in October, 1983. This paper will focus on our experience at ETA, on how the nature of the electron beam from that machine has complicated our data analysis. We hope to apply the lessons we learned there to future experiments which we are planning to do in May, 1984, at the PHREMEX facility of Los Alamos National Laboratory.

The motivation for this work is two-fold: first, we want to understand the basic physics of the interaction of an intense electron beam with condensed matter. "Intensity" is the key word in the sense that many projectiles might act coherently or incoherently to alter significantly the medium through which they pass.\(^1\) During the pulse of an intense electron beam through a material which is ordinarily dielectric, the conductivity increases by 10 to 15 orders of magnitude.\(^2\) This effect is related to the promotion of charge carriers to the conduction band and to the mobility and lifetime of those carriers in that band.\(^3\)\(^4\) Therefore, there is a physically interesting system which is measurable in real time. It is a system which interrelates
beam energy-deposition, charge-carrier concentration, material temperature, and carrier mobility. We are relying on Oakley Crawford and Rufus Ritchie of Oak Ridge National Laboratory for the theory of this transient conductivity.

The second motivation for these experiments is methodological. If we can understand the relationship between conductivity and energy deposition, we might be able to develop this technique into a diagnostic of the dose-depth relationship in materials subject to very-high dose and high dose-rate irradiation ($\geq 1$ Grad in $\leq 25$ nsec). This relationship is inaccessible to measurement by means of conventional dosimetry.

The idea for our experiment is based on work done by Vaisburd and Tavanov$^2$ (cf. fig. 2). They studied crystals of NaCl and KBr traversed by electrons of energy 0.3 MeV, in currents of density 500 A/cm$^2$, in pulses of duration 2.5 nsec. What one does is apply a d.c. bias voltage across a specimen of a given thickness, say $x_1$. For example, the bias voltage may lie in the range -500 V to +500 V. With a given bias voltage applied, the sample is traversed by the electron beam during a pulse, and the medium that was dielectric becomes conductive. The applied voltage drives a conduction current, and one measures a signal.

For an Ohmic medium, one expects a linear relationship between the signal and the applied voltage. As fig. 2 depicts, the thicker the specimen, the larger its resistance, and the less steep the slope of the relationship between signal and applied voltage. In other words, each slope corresponds to a resistance $R$ for a given thickness of sample. If one plots sample
resistance -- that is the resistance of the specimen induced by the electron beam through it -- one expects a linear dependence on thickness up until a thickness comparable to the range of beam electrons in this material. There should be a "knee" in the curve when samples are range thick, and the slope should increase greatly with thicker samples. Vaisburd and Tavanov measured values of resistance of 100 Ω to 1000 Ω in samples of NaCl. Ordinarily the sample resistance at 35°C would be \( \sim 10^{14} \) Ω when there is no beam-induced conductivity.\(^5\)

The conductivity is inversely proportional to the slope \( dR/dx \) and to the area \( A \) of the specimen through which the electron beam passes. One can determine the charge-carrier concentration \( n \) from the conductivity \( \sigma \) provided that one knows the mobility \( \mu \) of the charge carriers.

The top half of fig. 3 shows a diagram of the circuit we used to do our measurements. A sample is fixed by sandwiching two equally thick halves of material about a foil of titanium, which served as a high-voltage electrode. The sandwich is in a Faraday-shielded module which admits the electron beam through a collimator. (A sandwich configuration was tried in order to reduce any signal that may be induced by the magnetic field of the beam.) There is only one transmission line into and out of the module. It provides the d.c. bias voltage and carries the signal to the oscilloscope (Tektronix 7104).

The samples we used were polyethylene disks which were stacked to make specimens of different thicknesses. Polyethylene was selected because of a practical consideration: it stood up well to the 1-MeV electron beam of current density 2 kA/cm\(^2\) provided by the PULSERAD facility at the Naval
Polyethylene did not crack, whereas range-thick KBr did. At the ETA runs, we tested some sandwiches of range-thin KBr which exhibited in several cases anomalous cracking. In principle, one can obtain electrical information in a nanosecond time scale, way before a sample might crack mechanically milliseconds later. However, in a real experiment, one saves much machine time if a sample has the capability to withstand many electron-beam pulses and not be damaged. A few samples with the ability to assume their pre-irradiation electronic and structural properties between many successive electron-beam pulses enable the acquisition of a sufficient statistical sampling of data in only two days of machine time.

The linear relationship between the signal voltage and applied voltage is written in the bottom half of fig. 3. We would like to extract a value for the resistance of a given sample-sandwich of polyethylene from the slope of this line. This resistance of the medium arises from the non-zero conductivity induced by the exciting electron beam. There is a large zero-bias term to the expression for the signal, and it is dominated presumably by a current generated from a net flux of secondary electrons into the central electrode.

Figure 4 shows oscillograms from two typical shots out of the 200 we ran in two days of accelerator time. The upper two oscillograms were photographed during one shot through a polyethylene sandwich whose thickness corresponded to 7% of the beam-electron range in this material. There was a bias voltage of +100 V applied throughout the duration of the shot. On the left is a profile in time of the signal, and on the right a profile of the beam current measured upstream of the sample. The lower two oscillograms were photographed
during a beam pulse through a polyethylene sandwich whose thickness was about 27% of the beam-electron range. This sandwich was biased to +500 V. The signal is displayed on the left, the beam current on the right.

There are two distinct features to the signal oscillograms: All of the signals were about half as wide as the beam-current pulses — 11.4 nsec vs. 25 nsec. Secondly, each signal displayed either a double-peak or single-peak temporal structure. We know now that the narrowness is an artifact of a spatially wide electron beam misaligned and sweeping across the face of the collimator of the sample module. The variation in sweep of the beam from pulse to pulse may cause the variation between single-peak and double-peak structures. We do not know how the beam-current profile is related to the variable beam sweep. It is the beam sweep which has wreaked havoc on our data analysis.

As a start for the analysis, we naively plot the signal height vs. applied voltage. (For signals with a double-peak structure, we take the height of the signal at the temporal midpoint of the two peaks.) Figure 5 shows a plot of these signals for a polyethylene sandwich 7% of a beam-electron-range thin. This plot is representative of the quality of data obtained with specimens of other thicknesses. The results appear to be disastrous: there is large scatter in the data, and they appear to have the wrong slope from what one would expect (cf. fig. 2). These results call for a careful study of the behavior electron beam in order to characterize its sweep and effect on the signals observed.
The experiments at ETA were conducted in collaboration with Ed Jones, John Kidd, and Alex Stolovy of the Naval Research Laboratory. They had brought with them radiachromic film which, initially transparent, turns blue when exposed to radiation. The optical density is proportional to dose incurred by a specimen for one pulse of the beam. We placed these films over the collimation holes of the sample modules. Figure 6 shows several typical results.

"Wheel P" designates the wheel which held different-thickness samples of polyethylene in nine separate modules. (Wheels 1 and 2 held modules containing KBr.) The film placed over Wheel P, port 6, exemplifies the most salient feature of this experiment. The center of the cross locates the center of the collimator of the module. The dark spot on the left side of the film shows that most of the electron beam missed the collimator hole and wound up to the west of it. Also, the east tip of the film is optically denser than the center. Independent x-ray bow-probe measurements were performed by John Clark of ETA to obtain spatial distributions of the beam which are resolved in time of the current pulse. The measured distribution of the beam is consistent with the optical-density pattern of the exposed film: the beam starts in the east at the beginning of the pulse and sweeps west by some 4.3 cm. The beam sweeps in the same direction during each shot, but there can be a significantly large variation in amount and duration of its sweep. Our collimator is only 0.6 cm wide, and so it sees only a fraction of the beam.

Figure 7 shows optical-density scans of one of the radiachromic films which was exposed to 14 beam pulses. The film was scanned in two orthogonal
directions -- up-down and east-west. In the up-down direction, the optical density varies roughly symmetrically with respect to the collimator location. East and west along the film there is a large asymmetry attributable to the sweep of the beam. (The optical density in the western portion of the film was reduced as the film was warmed sufficiently in that region from repeated pulsing which thermally bleached the color.) From these diagnostics, if one crudely assumes a Gaussian beam profile, one can evaluate the beam radius, current density, and mean centroid shift during an average pulse.

What do these beam parameters mean for our conductivity experiment? They mean that the sweep time, or sweep velocity, or sweep distance across the face of the module fluctuates from beam shot to beam shot to cause a 30-40% scatter in the electron current density or in the amount of charge through the collimation hole and hence through our specimens. The only way to make sense of data exemplified by fig. 5 is to develop a scheme to normalize the signals measured with respect to values of the current density or charge through the collimator for each current pulse. Figure 8 outlines a simple scheme for such normalization, where \( J_{\text{beam}}(\text{meas}) \) represents the beam current density through the collimator hole for a particular shot, and \( J_{\text{beam}}(\text{norm}) \) would be the mean value for a series of shots. The problem is that one cannot apply such a scheme without a detailed analysis of beam observables correlated to signals. Such an analysis would permit evaluation of \( J_{\text{beam}}(\text{meas}) \) for each shot.

Don Rule has come up with a program which might enable us to evaluate the beam current density through the collimator for each beam pulse. We could
then normalize our data properly to account for the effect of the shot-to-shot fluctuation. Although we still hold hope for the data, at this time we cannot provide values for the beam-induced conductivity of polyethylene. Figure 9 summarizes our experience at the Experimental Test Accelerator.

We would like to conclude on an upbeat, tantalizing note. In planning for experiments in May, 1984, at the PHERMEX facility of Los Alamos National Laboratory, we have conducted test runs of prototype modules and samples at the PULSERAD accelerator of NRL. At PHERMEX we are planning to study the beam-induced conductivity of vitreous silica ("fused quartz"), and we simulated this material at PULSERAD with ordinary glass microscope slides. The top oscillogram in fig. 10 shows the signal measured with no applied voltage. The surprise comes in the middle oscillogram, which displays the signal photographed during and after the beam pulsing through a specimen biased to -200 V. Apparently the increase in electrical conductivity develops significantly after the beam has traversed the glass! At this time we have no explanation of this observation. We are looking forward to the interesting physics we expect to learn from upcoming observations at PHERMEX, whose current pulse train is depicted in the bottom plot of fig. 10.

We would like to acknowledge the support of John Clark and Ken Struve of ETA, and we thank them for their helpful assistance with these experiments. In fig. 11 is a list of some names of people on whose help we have relied since the start of the experimental program in July, 1983. This work was supported by the Directed Energy Program Office of the Naval Sea Systems Command.
References


6. We thank J. Robert Greig for the opportunity to use this facility. Test runs were conducted with the guidance of Michael Raleigh on October 5 and 7, 1983, and on April 2, 1984. We are grateful for his help.


14. We thank Marty Ganosczy of NBS for assisting us with these measurements.
ELECTRIC CONDUCTIVITY OF POLYETHYLENE DURING PULSED ELECTRON-BEAM (4.2 MeV) IRRADIATION AT ETA

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Fig. 1
D. I. Vaiburd and É. G. Tavanor, transl. of

NaCl; earlier work: KBr

Slope of a line yields $R$, resistance for a given thickness $x$ of sample.

$$\sigma(x) = \frac{1}{A \frac{dr}{dx}}$$

$$n(x) = \frac{\sigma(x)}{\mu e}$$

Fig. 2
For an Ohmic medium,

\[ V_{signal} = \frac{Z_{line} R}{Z_{line} + R} I_{s.e.} - \frac{Z_{line}}{Z_{line} + R} V_{applied} \]

\[ R = \text{effective resistance of sample sandwich} \]
\[ Z_{line} = 50\Omega, \text{ impedance of transmission line} \]
\[ I_{s.e.} = \text{current induced by secondary electrons} \]
polyethylene
sandwich thickness 6.7% e⁻ range (0.161 cm)
$V_{\text{applied}} = +100 \text{ V}$

\begin{align*}
V_{\text{signal}} & \quad (100 \text{ V} / \text{div}) \\
& \quad (10 \text{ nsec/div}) \\
\end{align*}

Beam Current
(2 A/div)

FWHM \approx 11.4 \text{ nsec}

sandwich thickness 27.1% e⁻ range (0.649 cm)
$V_{\text{applied}} = +500 \text{ V}$

\begin{align*}
V_{\text{signal}} & \quad (500 \text{ V} / \text{div}) \\
& \quad (10 \text{ nsec/div}) \\
\end{align*}

Beam Current
(2 A/div)

FWHM \approx 25 \text{ nsec}

\sim 200 \text{ shots in 2 days}

Fig. 4
Fig. 5

Polyethylene 0.161 cm Raw Data
(0.067 R<sub>ex</sub>)
Chlorostyrene-1, Radiachromatic Film
(a polymerized mixture of 96% styrene monomer and 4% chlorostyrene monomer)

wheel P, port 1, 26 shots
wheel P, port 6, 2 shots
wheel 1, port 7, 2 shots
wheel 2, port 2, 1 shot

Fig. 6
Optical density of chlorostyrene-1 film exposed to 14 shots, wheel P, ports 3

For $J(r) = J_{\text{peak}} e^{-r^2/a^2}$

$a = 1.3 \pm 0.1 \text{ cm}$

$J_{\text{peak}} = 1.2 \pm 0.1 \frac{\text{mA}}{\text{cm}^2}$

dose $\sim 2.2 \text{ Mrad}$

$\Delta T \sim 10 \text{ K/shot}$

east $\rightarrow$ west
centroid shift of 4.3 cm
in $\sim 4-12 \text{ nsec}$

Fig. 7
What is the source of the scatter?

Assume that sweep time, sweep velocity, or both fluctuate from beam shot to beam shot to cause a 30-40% scatter in the current density $J_{\text{beam}}$ through the collimator.

$$V_{\text{signal}} = \frac{Z_{\text{line}}R}{Z_{\text{line}} + R} I_{\text{s.e.}} - \frac{Z_{\text{line}}}{Z_{\text{line}} + R} V_{\text{applied}}$$

Assume $I_{\text{s.e.}} < J_{\text{beam}}$, $R < 1/J_{\text{beam}}$, then

$$V_{\text{signal}}^{(\text{norm})} = \frac{Z_{\text{line}} + R}{Z_{\text{line}} + J_{\text{beam}}^{(\text{meas})}} V_{\text{signal}}^{(\text{meas})}$$

Also, if $R > Z_{\text{line}}$, then

$$V_{\text{signal}}^{(\text{norm})} \approx \frac{J_{\text{beam}}^{(\text{norm})}}{J_{\text{beam}}^{(\text{meas})}} V_{\text{signal}}^{(\text{meas})}$$

Fig. 8
SUMMARY of ETA experience

1. Beam profile \( \sim 1.3 \text{-cm radius} \)
   peak current \( \sim 6.4 \text{ kA} \)
   current density \( \sim 1.2 \text{ kA/cm}^2, \text{max.} \)
   energy \( 4.2 \text{ MeV} \)

2. Beam misalignment \( \sim 1.6 - 2.7 \text{ cm} \)
   beam sweep \( \sim 4.8 \text{ cm/4-12 nsec} \)
   fluctuation, shot-to-shot \( \sim 30\% \)
   reduce effective pulse duration from 25 nsec to
   11.4 nsec and cause much scatter in the data.
   These effects are large because the collimator
   dimension is smaller than the beam-sweep dimensions.

CONCLUSION

Successful conductivity measurements depend
on proper beam alignment on target, small
shot-to-shot variation, synchronized signal and
beam-current diagnostics, and controllable, high
rate of repetition of the beam pulses.

Fig. 9
Pulserad (NRL) - 1 MeV, 2 kA/cm²

silver-coated GLASS, 65% range thick
0 V applied

silver-coated GLASS 65% range thick
-200 V applied
WOW!

Phermex (LANL)

20 MeV, 10-15 kA/cm²
The shape of things to come.

Figure 4. Beam current pulse train.

Fig. 10
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Fig. 11