FORMATION OF EXCITED HYDROGEN ATOMS
BY CHARGE TRANSFER AND DISSOCIATION

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I. Title

Formation of Excited Hydrogen Atoms by Charge Transfer and Dissociation

II. Introduction

This report summarizes work performed on excitation phenomena under contract AT-(40-1)-2591 for the U. S. Atomic Energy Commission. The present report covers the period 1 December 1972 to 30 November 1973 which corresponds to the first 9 months of the 12 month period covered by modification No. 15 to this contract, plus the final three months of the preceding contract period.

III. Summary

This work represents a continuation of our program to study formation of excited hydrogen atoms in various collision situations. The principal effort has been the excitation and scattering of hydrogen atoms by collision on rare gases. A second area of study is the excitation and scattering associated with proton scattering from metallic surfaces.

In the study of hydrogen atom collisions with gas targets we have considered formation of the metastable state;

\[ H + X \rightarrow H(2s) + X \]  (1)

The target X, was helium or argon, the projectile energy was 1.5 to 25 keV; measurements were made of the cross section as a function of projectile scattering angle. The objective here was to test the applicability of new coupled state theoretical calculations of this cross section and also to determine the limitations of the more frequently used Born approximation. The conclusion is that the Born approximation is very
inaccurate at low energies although it does, fortuitously, predict the
total cross section fairly well. By contrast the coupled state calcula-
tions are in rather good agreement with experiment and are clearly a
better representation than the Born. It is interesting that these pre-
dictions for neutral particle collisions agree with experiment down to
energies of \(4\) keV or lower while for charged particle collisions (e.g.
proton impact) the theoretical predictions are accurate only at energies
above 200 keV. The reason for this is that charged particles exhibit
very long range interactions which are difficult to handle theoretically;
neutral particles do not exhibit these long range forces.

Our experiments with gaseous targets have also included stripping
of electrons from \(H\) atoms to give protons and also elastic scattering
of neutral atoms. These have been carried out with He and Ar targets
at energies from 5 to 25 keV.

In our studies of particle impact on solids we have been analyzing
the photon emission induced by \(H^+\) and \(He^+\) on copper targets. We have
shown previously\(^1\) that spectral lines from scattered neutralized
projectiles exhibit a characteristic shape that may be interpreted as
a Doppler effect associated with the speed and direction of the reflected
projectiles. Analysis of the line shape provides information on the
fate of projectiles that strike a surface. Some preliminary measure-
ments of line shapes induced by 150 to 1000 keV \(He^+\) on copper are
presented. They indicate that excited projectiles arise principally
from scattering at the first few monolayers of surface and are there-
fore reflected with approximately the same energy as they were incident.

The probability of scattering with excitation has been shown to decrease with increasing energy above 150 keV. All these observations have been made in a relatively "dirty" vacuum system with a base pressure of around $10^{-7}$ Torr; this is not a satisfactory environment for surface studies and we have recently completed construction of a ultra-high-vacuum system that will permit studies in a totally clean environment. With this new facility we are currently repeating and extending these observations.

The body of this report is organized into two sections, the first dealing with neutral particles in gases and the second with ion impact on solids. These are followed by sections concerning the usual administrative matters.

IV. Low Energy Differential Cross Section Measurements in Gaseous Targets

(a) Introduction

We have recently concentrated on the study of how neutral ground state hydrogen atoms interact with targets of helium and argon. The specific mechanisms studied have been excitation of the incident hydrogen

$$H(1s) + X \rightarrow H(2s) + X ,$$  \hspace{1cm} (1)

stripping of the hydrogen

$$H(1s) + X \rightarrow H^+ + e + X ,$$  \hspace{1cm} (2)

charge transfer to form negative ions
\[ H(1s) + X \rightarrow H^- + X^+ \] \hspace{1cm} (3)

and elastic scattering of the hydrogen atoms

\[ H(1s) + X \rightarrow H + X \] \hspace{1cm} (4)

Experimental measurements have been carried out at projectile energies from 1.5 to 25 keV; in all cases we have been studying the cross section for the reaction as a function of scattering angle and for the inelastic events we have also studied the total cross sections.

Our principal effort has been on excitation of H to the metastable 2s level [Eq. (1)] and elastic scattering of hydrogen [Eq. (4)]. The objective has been to test the detailed theoretical predictions that are now available. Since both colliding particles are neutral, there are no long range Coulomb forces for the theoretician to contend with in calculating cross sections. A theoretical calculation must be cut off at some finite impact parameter. For a neutral-neutral collision it is quite simple to choose a cut off point where the interaction is essentially zero; for a charged particle collision the interaction will not be zero for any finite cut-off point. It follows that a neutral-neutral collision is an excellent vehicle for testing the formulation of a complex theoretical prediction.

The experimental system is basically the same as that we have used previously and discussed in the open literature\(^2,3\); consequently we shall give only a short description here. The data obtained is

\begin{itemize}
\end{itemize}
Figure 1. Schematic diagram of the apparatus.
extensive and we shall summarize it briefly in this report. A detailed publication of method and results is in course of publication.

The experimental arrangement is shown in Fig. 1. The atomic hydrogen beam was provided by charge transfer neutralization of protons. A 1 to 30 keV accelerator provides a mass analyzed, energy selected, proton beam that is directed into a large tank which contains the experimental system. After passing through the first collimating aperture, the beam enters a differentially pumped gas cell where part of the beam was converted to neutral atoms. Charged particles remaining in the beam were removed by electrostatic deflection; this electric field also Stark quenches any metastable neutral hydrogen atoms. After a further collimation, the remaining hydrogen atom beam enters a cylindrical cell containing the target gas of interest.

Scattered projectiles exit the target cell through a pair of apertures which define the angle of scattering; the width of these apertures limits the angular resolution of the scattered particle detection system. Beyond this last set of apertures are a series of detectors which are used to assay the composition of the scattered particle flux. Metastable hydrogen was monitored by electric field quenching of H(2s) to give Lyman alpha photons; these were then detected by a channel electron multiplier operating in a counting mode. The field used to quench H(2s) also electrostatically separates the fluxes of H°, H+ and H− which are separately monitored on Faraday cups; the H+ and H− produces a current that may be recorded directly; the H° component is used to eject secondary electrons from the base of a Faraday cup and that current is used to monitor H° flux. At scattering angles
larger than 3° the flux of scattered projectiles was very small and could not be conveniently measured on the Faraday cups. To extend the data to 10° scattering angle the cups were replaced by channel multipliers which could count single particles. The signals from the single particle detectors were normalized to the Faraday cup signals at 3°; consequently there was no need to separately calibrate these devices.

The differential cross section for scattering of any particular component at an angle θ is given by the equation:

\[
\frac{d\sigma}{d\Omega}\bigg|_\theta = \frac{N_i}{N_B N_T \int_{\Delta x} \omega(x) dx}.
\]

Here \( N_i \) is the flux of the scattered component under study (particles/sec); \( N_B \) is the incident beam flux (particles/sec) and \( N_T \) is the target gas density (particles/cc). The geometric term \( \int_{\Delta x} \omega(x) dx \) represents the product of effective beam path length that the detectors view, with the effective solid angle subtended by the detectors at the beam path. The method of evaluating the geometric term follows the analysis by Fillipenko\(^4\) and has been previously described by us\(^3\). It is clear from equation (5) that for a constant projectile beam flux and constant target density the relative variation with angle, of signal divided by geometric parameter, will immediately give the differential cross section in arbitrary units.

The apparatus can also be readily modified to determine total cross sections. With the detector systems set for a scattering angle of

of zero degrees (i.e. \( \theta = 0^\circ \)) and the "angle defining apertures" (see Fig. 1) opened up, it is possible to accept into the detector system all projectiles scattered through angles of 5° or less; preliminary tests show no significant flux of particles of angles greater than 5°. The total cross section for production of a state is given by

\[
\sigma = \frac{N_i}{N_B N_T l}.
\]  

(6)

Where \( N_i \) is the flux of particles in the state of interest (particles/sec), \( N_B \) is the flux of incoming projectiles (particles/sec), \( N_T \) the target density (particles/cc) and \( l \) is the length of the target cell. The relative variation with impact energy of the exiting particle flux \( N_i \) gives directly the relative variation of cross section.

In order to establish absolute cross sections it is necessary to monitor these various components absolutely. The target density, \( N_T \), was determined from measurement of target gas pressure with a capacitance manometer\(^2\). Scattered \( H^+ \) and \( H^- \) were measured directly as a current and are therefore absolute. The efficiency of the scattered neutral detector was calibrated using an \( H \) atom beam of known flux in the manner we have described previously\(^2,3\). It was also necessary to monitor the incident primary beam \( N_B \) and this was achieved by temporarily inserting a secondary emission detector into the target cell; this detector was identical to that used to detector scattered \( H^+ \) flux and its calibration was assumed to be the same. These variation techniques for determining parameters absolutely are direct, and quite accurate (say \( \pm 5\% \)). There remains the problem of determining the efficiency of the metastable detector.
This was achieved by measuring the total cross section for the process
\( \text{H}^+ + \text{Ar} \rightarrow \text{H}(2s) + \text{Ar}^+ \) in relative units and then normalizing to the previous
results of Andreev et al.\(^5\). This procedure was discussed and justified in
our previous work\(^2\); the accuracy is no better\(^2\) than \( \pm 60\% \).

Various tests have been made to confirm the proper operation of
the equipment; these are in general the same as those mentioned in our
published reports\(^2,3\) and are too numerous to discuss in detail here. One
new test that is of importance is to confirm that measured cross sections
are not dependent on either the nature or pressure of gas in the neutralizer
cell; this test is taken as proof that the neutral beam contains only a
negligible amount of excited atoms.

(b) Consideration of Angular Resolution

A considerable amount of effort has been expended on the mathematical
treatment of how the limited angular resolution of an experiment distorts
the cross section measurements. Many experiments, including the present,
utilize an angular resolution of the order \( 1/4 \) degree. If a large cross
section change occurs in this interval then the apparent cross section will
be greatly different from the true value. We have developed a computer
program that takes an analytical form of the cross section and predicts
the cross section that would be observed with an apparatus which has a
limited angular resolution; in effect we can predict the apparent cross
section that is measured by the apparatus with the operational definition
of cross section given by Eq. 5. We show, using a simple Rutherford
scattering cross section, that a measured cross section may differ from
the true cross section by as much as \( 50\% \) at angles around \( 0.5^\circ \).

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\(^5\) E. P. Andreev, V. A. Ankudinov, and S. V. Bobashev, Soviet Physics,
We consider this to be a very important factor in the interpretation of measured cross sections and we are preparing a publication that will detail our findings in this matter.

(c) Differential Cross Sections for Excitation to $\text{H}(2s)$

In Fig. 2 we show cross sections for excitation of $\text{H}(1s)$ to $\text{H}(2s)$ by impact on helium at 10 keV energy. Also indicated are a prediction in the Born approximation and in a coupled state calculation involving $1s$, $2s$, $2p_0$, and $2p_{\pm1}$ states. The data bears no resemblance to the prediction of the Born approximation and this type of calculation is clearly incorrect. The experiment gives the same relative behavior as the coupled state prediction but they differ by a factor of four. The discrepancy here is a little puzzling. We have not yet taken into account the effect of finite apparatus resolution; this will reduce the discrepancy somewhat. Also the measurements have an inherent uncertainty in absolute magnitude of $\pm$ 60%. Analysis of this apparent discrepancy is underway to determine whether it indicates that the coupled state calculation is incorrect in some manner. We can say with certainty that the coupled state calculation is a far better representation than the Born.

We have a considerable body of additional data at various energies down to 1.5 keV and including some work with an argon target. The results are very similar to the data in Fig. 2 except that at very low energies some structure is observed. These various additional data are now being prepared for publication and will not be given here.

(d) Differential Cross Sections for Stripping and Elastic Scattering

We have a considerable body of data on stripping and elastic scattering represented by the reaction equations (2) and (4). A sample

Figure 2. Differential Cross Section for Excitation of H(2s) Induced by 10 keV H(ls) Impact on He. Note that Cross Section and Scattering Angle are in the Center of Mass Co-ordinate System.
for 10 keV H(ls) impact on He is shown in Fig. 3. The cross sections decrease rapidly with angle showing approximately a $\theta^{-3.6}$ fall-off. This is almost the same as the $\theta^{-4}$ fall off that would be predicted by a simple Rutherford scattering expression; it may be concluded that the principal interaction is between nuclei of the colliding particles and that electron screening is relatively unimportant.

(e) Total Cross Sections for Excitation to H(2s)

In Fig. 4, we show the measured total cross section for excitation of H(2s) induced by H(ls) impact on He. Bearing in mind the accuracy limitations on the absolute experimental values (± 60%) we may conclude that the experimental data agrees with the four state theoretical predictions down to 5 keV. By contrast the Born approximation predictions show a completely different energy dependence below 10 keV and do not follow the experimental data at all.

(f) Conclusion

The general conclusion from this work is that the Born approximation should not be used for predicting neutral-neutral collisions cross sections below 20 keV energy. The coupled state calculation, does however seem to give an adequate representation down to 5 keV energies.

V. Excitation Induced by Particle Impact on Solids

(a) Introduction

During the present reporting period we have embarked upon a study of particle scattering from surfaces. We have shown previously\textsuperscript{1} that when an energetic proton or He\textsuperscript{+} ion is incident upon a metal target then some of these ions are neutralized into excited states and emit photons as they recoil from the surface. The line emission from the excited
Figure 3. Differential Cross Sections for Formation of $H^+$, $H(2s)$ and Elastic Scattering of $H$, For 10 keV H Impact on a He Target. Cross Section and Angle are Expressed in the Laboratory Co-ordinate System.
Figure 4. Cross Section for H(2s) Formation Induced by H(1s) Impact on He. Curve A is present Data. Curve B, Born Approximation. Curve C is Four State Theoretical Calculation.
projectiles exhibits a Doppler broadening that is related to the velocity distribution of the scattered projectiles. The objective of our work is to determine the angular and speed distribution from an analysis of the line shape.

For any study of phenomena occurring at a surface it is necessary that the composition of the surface be properly specified. In general one requires that the surface be atomically clean so that the surface composition is identical to that of the bulk material; in the event one is interested in contaminated surfaces then one starts with a clean surface and introduces the contaminant in a controlled and reproducible manner. The principal objective for the present reporting period was to construct new equipment that would provide the necessary surface cleanliness and would also have facilities for precise orientation of the target surface with respect to the projectile beam. This construction is now complete and some data obtained with this new apparatus will be discussed; in addition we have also carried out some measurements in an existing chamber in which the vacuum conditions were poor and the target surface may have been contaminated.

Figure 5 shows the general layout of the apparatus. Mass analyzed projectile ions from an accelerator are collimated by a series of apertures and then directed onto the target surface. An optical spectrometer is placed at 90° to the beam path. The target specimen is held in a precision manipulator that can rotate about an axis perpendicular the plane defined by the projectile beam and optical axis; the target may thereby be rotated to vary the angle of incidence on the surface, θ.
Figure 5. Schematic Diagram of the Apparatus Used for Study of Light Emission Induced by Ion Impact on Solid Targets.
We have utilized two separate experimental arrangements of this type. The first was a rather poor quality vacuum environment (base pressure $10^{-7}$ Torr) assembled from existing components. It was attached to the Van der Graaff accelerator and used for measurements at energies between 150 keV and 1000 keV. Since this environment will not guarantee a clean surface, we must bear in mind that the data may be influenced by surface contamination. Our second experimental system has been specially designed for this work and only recently was finally assembled. It has a base vacuum of $10^{-10}$ Torr and is fully bakeable. Currently it is used for studies at impact energies from 5 to 30 keV using our low energy accelerator; it has been constructed so that it may be readily transferred to the Van der Graaff accelerator when desired. With the recent completion of this new system we have ceased using the first arrangement where the vacuum conditions are unsatisfactory.

Some of the more important features of the new system are as follows. It is all metal and fully bakeable with ion, sublimation and sorption pumps to ensure cleanliness. To interface between the rather poor vacuum of our accelerator ($10^{-6}$ Torr) and the high vacuum of the target chamber ($10^{-10}$ Torr) we have built in some three differential pumping stages. The target chamber is now being fitted with a residual gas analyzer to further define the possible background contaminants. The target is held on a precision manipulator which permits rotation about one axis and translation in three mutually perpendicular directions. We also have facilities for heating the target (to 800°C) and cooling it (to -173°C) if this should appear to be desirable. For the moment we have confined ourselves to the use of high purity polycrystalline
metal targets. These are subjected to a preliminary polishing and chemical cleaning before insertion in the vacuum system; before commencing measurements the surfaces are further sputter cleaned with a heavy ion beam.

The main effort this year has been in the development of new facilities; consequently only a small amount of data has been obtained and that has not yet been fully analyzed. Since all construction work is now complete, we anticipate more rapid progress on the research. Our preliminary work has been restricted primarily to the effect of He$^+$ impact on polycrystalline copper; this case has been chosen because it has been researched elsewhere and we have some guidance as to what will occur. Also, there is a rather good body of data on the conduction band structure of copper; it therefore provides a useful vehicle for studying the basic collision mechanism. We summarize below some of our preliminary findings for this case.

(b) **General Spectral Features**

We can observe only transitions from the $^3D$ and $^3P$ levels of helium atoms that result from scattering and neutralization of the He$^+$ ions at the surface; this general observation is valid for all impact energies from 5 to 1000 keV. It is not clear why we see no emissions from singlet levels. This implies some sort of selection rule in the neutralization. For H$^+$ impact on copper we see very clearly the Balmer series of hydrogen. In none of these observations do we see spectral lines from copper. This implies that sputtering of excited atoms occurs only very rarely.

(c) **Line Shapes**

In Fig. 6 we show a detailed line shape for the 5875 Å ($^3D \rightarrow ^3P$) transition of helium measured at 20 and 200 keV impact energies. The
Figure 6. Line Shape of the 5875Å (3^2D \rightarrow 2^3 P) HeI Emission Induced by He^+ Impact on Polycrystalline Cu.
(a) 200 keV Impact Energy, 70° Angle of Incidence.
(b) 20 & 33keV Impact Energy, 85° Angle of Incidence.
structure is quite similar to that which we discovered\textsuperscript{1} for 10 keV He\textsuperscript{+} impact on copper and which has already been reported.

We have shown previously\textsuperscript{1} how such line shapes may be predicted with assumptions concerning the speeds and directional distributions of the scattered atoms; comparison of the prediction with the measurement allows one to decide whether the assumptions are consistent with reality. We determined previously\textsuperscript{1} in a study of 10 keV He\textsuperscript{+} on copper, that the excited atoms had a speed distribution consistent with a biparticle He\textsuperscript{+} on copper atom collision at the surface; moreover the angular distribution of the scattered He atoms was essentially a Rutherford distribution appropriate to the interaction of He and Cu nuclei. This same picture evolves from the analysis of our new data at energies up to 1000 keV.

This general result says simply that excited He is created only when the projectile is reflected from the surface layer in a biparticle, He\textsuperscript{+} on copper atom, collision. Now, it is well known that most projectiles penetrate some distance into the target before undergoing a "violent" scattering event that returns them towards the surface; during passage into and out of the target a considerable amount of energy is lost by small angle collisions so that exit velocity is substantially reduced below the entrance velocity. These "slow" reflected particles apparently are not excited. The reason for this is not clear.

One factor we have not yet included in our analysis is radiationless de-excitation of excited atoms when they are close to the surface; possible mechanisms for the radiationless decay are Auger effects\textsuperscript{7} and

resonant transfer of the excited electron into the conduction band. These mechanisms would tend to destroy excited states and be particularly effective for slow moving atoms that remain in the vicinity of the surface for a long time. With these mechanisms in mind one might postulate that all emerging projectiles, irrespective of energy, have an appreciable chance of being excited but that the slow ones become subsequently de-excited so that the scattered atoms which remain excited are primarily those with high velocity. We are at present reformulating our analysis to include the radiationless decay process. While this explanation does seem to be consistent with our data in the 3 to 20 keV region, it is not consistent with our observations at 200 keV.

VI. Program for the Remainder of the Contract Year

The studies of H(1s) impact on He and Ar at energies from 1.5 to 25 keV will be extended to include the charge transfer reaction that leads to H⁻ formation [Eq. (3)]. That will complete our analysis of this type of collision. The H⁻ formation has a very small cross section and is strongly forward peaked so that little H⁻ is observed at large scattering angles. It will be necessary to make some slight improvements to the apparatus to ensure that the resulting small signals are properly recorded. This will be completed by the end of the contract year.

We shall continue our study of the line spectra induced by 5 - 25 keV H⁺ and He⁺ impact on solids. The objective will be to determine total fluxes of scattered excited atoms and to analyze the spectral line shape to give velocity distributions of such particles. Particular attention will be paid to the question of why no slow excited particles are observed.
We anticipate completion of studies with copper targets by the end of the contract period; this will supplement the results we have already published.

VII. Program for the Future

It is intended to continue our studies of H(1s) scattering in gases by going to an H_2 target; we shall again monitor the formation of H(2s), H^+, H^- and the elastic scattering of H^0. The justification for this comes from the possibility of using the data to model the behavior of neutral beam injectors used on ORMAK. These injectors take an unanalyzed beam of hydrogen ions from a discharge source and send it through H_2 gas; while not an optimum situation for production of H^0 it is very simple because the target H_2 is merely gas from the discharge region that effuses from the exit aperture. The collisions of principal importance in such a source are H^+, H_2^+, H_3^+ and H^0 impact on H_2. We have already performed these measurements for the ionic states and published the data; the measurements for H^0 on H_2 will complete the set of information. This is a very simple thing for us to accomplish since it involves only a further exploitation of an existing apparatus and established techniques; the work is expected to take no longer than six months. The apparatus will then be modified for studies on solids as described below.

We shall continue to exploit the capabilities of our new equipment to study formation of excited states by scattering at surfaces. By using a variety of solid metal targets such as Cu, Mo, Al, Fe, Steel, and various alloys we shall determine the parameters which influence the excited state content of the reflected particle flux; relevant para-
meters which we expect to be of importance are work function, depth of conduction band and population distributions in such bands. We expect that our studies of line shape will confirm our preliminary conclusion that excited atoms arise by bi-particle collisions at the surface and that atoms scattered deep in the solid do not return in an excited state.

We propose modifying the apparatus that we have been using for study of gas target scattering to accommodate a solid target. We shall use an incident beam of $H^+$ or $H^0$ and study the charge and excited state distributions of scattered particles and also the energy distributions of such particles. To accomplish this we will utilize essentially the same detection systems that we have used for particle scattering in gas targets; thus no appreciable development of equipment is necessary.
VIII. Publications and Travel

A total of five papers have been published during the present reporting period.


Four papers have been presented at conferences.

(i) Angular Distribution of H(2s) Formed by Collisional Excitation of Hydrogen Atoms", Contributed Paper, Fourth Annual Meeting of the Division of Electron and Atomic Physics of the American Physical


During the year Dr. Thomas (Project Director) attended the Eighth International Conference on the Physics of Electronic and Atomic Collisions in Belgrade, Yugoslavia (July 1973); associated visits were made to the atomic collisions research groups at Belgrade University and at the University of London, England. This foreign travel was at no cost to the AEC. Dr. Thomas also attended the Physical Electronics Conference in Berkley (March 28, 1973). Mr. Sauers attended the meeting of the Division of Electron and Atomic Physics of the APS in Menlo Park (November 1972).
Dr. Thomas visited Auburn University to give a seminar on the work contained in this report. He has also made periodic visits to Oak Ridge in order to confer with scientists in the Controlled Thermonuclear Research Program.

IX. Personnel

Professor E. W. Thomas has been Project Director and Principal Investigator in this work. He has devoted to it 15% of time in the academic year and 85% of full time during the summer.

Mr. Isidor Sauers has been supported for half of full time on this contract. He has been responsible for the work on atomic collisions in gases and is currently writing his Ph.D. thesis on that material.

Mr. W. Allen Carr has been supported for about one half of full time to carry out the study of atomic collisions on solid surfaces. He continues to work on this project.

Mr. T. W. Nichols was supported for about one quarter time up to July 1973. He has now graduated with an M.S. degree and is no longer with us.

X. Incident Report

There have been no incidents for which a report is required during the performance of the research under this contract in the present reporting period.