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ION-SOURCE DEPENDENCE OF THE DISTRIBUTIONS OF INTERNUCLEAR SEPARATIONS IN 2-MeV HeH^+ BEAMS*

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Experiments involving the use of MeV molecular-ion beams have yielded new information on atomic collisions in solids. A central part of the analyses of such experiments is a knowledge of the distribution of internuclear separations contained in the incident beam. In an attempt to determine how these distributions depend on ion-source gas conditions, we have studied foil-induced dissociations of H_2^+, H_3^+, HeH^+, and OH_2^+ ions. Although changes of ion-source gas compositions and pressure were found to have no measurable influence on the vibrational state populations of the beams reaching our target, for HeH^+ we found that beams produced in our rf source were vibrationally "hotter" than beams produced in a duoplasmatron. This was also seen in studies of neutral fragments and transmitted molecules.

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Fast (MeV) molecular-ion beams provide a unique source of energetic projectile ions which are correlated in space and time. The recognition of this property has prompted several recent investigations of various aspects of the interactions of these ions with matter. High-resolution measurements on the fragments resulting from these interactions have already yielded a wealth of new information on such diverse topics as plasma oscillations in solids and stereochemical structures of molecular ions as well as a variety of atomic collision phenomena.

A central part of the analysis of such experiments is a knowledge of the distribution of internuclear separations contained in the incident molecular-ion beam. In the earliest of such experiments, the analyses assumed simple distributions centered at the equilibrium separation of the ground electronic state of the ion. In the case of H_2^+, several groups assumed that direct ionization of the ground electronic and vibrational state of H_2 would populate the vibrational levels of H_2^+ $(1s\sigma_g)$ according to the Franck-Condon principle.[1,2] There are, however, a variety of reasons to expect significantly more complex distributions for the ion source conditions generally used. Important contributing mechanisms include the collisional rearrangement of vibrational state populations, autoionization of high Rydberg states of the neutral molecule and rotational excitation in the ionizing collisions.[3] Furthermore, the usual assumption of the neutral source gas molecules being found in the ground vibrational state would appear to be unfounded in the case of the "hot" ion sources often used for these experiments.

It has previously been demonstrated that by choosing beam energies and target thicknesses to minimize simultaneously the influence of both non-equilibrium charge state effects and multiple scattering, energy-angle

measurements of Coulomb-exploded molecular dissociation fragments can be used to determine the initial distribution of internuclear separations $D(r_o)$ for light diatomic beams such as H_2^+ and HeH^+.[4,5] We report here on an extension of those measurements with the aim of determining the effect of ion source conditions on Coulomb explosion experiments.

The excess kinetic energy released by the foil-induced Coulomb explosion of molecular ions produces sizable energy and angle shifts in the resulting fragment trajectories. We measure joint laboratory distributions in energy and angle of dissociation fragments observed downstream from the target. These distributions are simply related to velocity distributions in the rest frame of the incident molecule. For diatomic projectiles, the diameters of the measured "ring patterns" are approximately proportional to the geometric mean of the effective charges on the fragments and inversely proportional to the square root of the bond length.

The apparatus used in these experiments is extensively described in the literature.[4,6] In the present experiments, we accelerated beams of H_2^+, HeH^+, H_3^+, and OH_2^+ to energies in the range of 1.5--3.0 MeV. After passage of the molecular clusters through a thin carbon foil (~100 Å), the energy and angular spectra of the resulting proton fragments were recorded. Rather than measuring complete ring patterns, we concentrate on diametric "crosses" (i.e. an energy distribution for zero angular shift together with an angular distribution for zero energy shift).

The ring diameters are determined by the peak separations ΔE (for the energy spectrum at zero angle shift) and $\Delta\theta$ (for the angular spectrum at zero energy shift). These parameters thus are indicative of the mean internuclear separation contained in the incident molecular-ion beam. The widths of the peaks in the energy and angular spectra are a measure of the

range of internuclear separations. A complete discussion of the relationship between the spectral shapes and the distribution of internuclear separations can be found in ref. 4.

Variations in the ring diameters and peak widths were not observed when various ion source parameters (filament current, gas pressure, extraction voltage, etc.) were changed. Small changes, however, could be produced when H_2^+ beams were produced from differing source gas mixtures. Pratt and Chupka have suggested that an H_2^+ beam could be vibrationally cooled by ensuring sufficient collisions with He or Ne.[7] When fed with pure H_2 gas, both rf and duoplasmatron sources produced similar beams of H_2^+ at 3.0 MeV. For example, the ring diameters from the angular spectra observed with both sources were 3.73 ± .04 mrad with peak widths of 1.19 mrad (FWHM). When fed with a mixture of 10% H_2 and 90% Ne (or He), the diameters increased to 3.84 ± .04 mrad and the peak widths decreased to 1.00 mrad. These larger diameters, and sharper angular spectra, are indicative of vibrationally "cooler" beams (smaller internuclear separations). Results for the energy spectra were comparable.

The only other source-dependent effect that was observed was with beams of HeH^+. For all other beams tested, the vibrational state populations of the molecules reaching the target chamber were found to be remarkably constant, independent of whether a duoplasmatron or rf source was used to produce the molecular-ion beams. Beams of HeH^+, however, produced in our rf source were found to be vibrationally "hotter" than beams produced in the duoplasmatron source. Figure 1 shows the angular spectra of crosses measured with 2-MeV HeH^+ produced in each source. Using the techniques described in Ref. 4, we unfold from these spectra the corresponding distributions of internuclear separations $[D(r_o)]$ for each beam (see fig. 2).

While the effect of modifying $D(r_o)$ is clearly seen in the proton crosses of fig. 1, other experiments show this difference even more strikingly. Figure 3 shows complete ring distributions for neutral hydrogen fragments resulting from the dissociation of 2-MeV HeH^+ beams after passage through ~60-80 Å thick carbon foils.

Neutral fragments are particularly sensitive to the r_o-distribution at large distances. At the high energies used, the fragment ions are essentially bare while transiting the solid target and thus acquire velocities characteristic of a simple Coulomb explosion in the foil. Electron capture upon exit effectively truncates the explosion as the ions are screened and little further kinetic energy is acquired outside the foil. Those ions that enter the target with large internuclear separations transform a significantly smaller fraction of the available Coulomb energy into fragment kinetic energy than do those molecules with small separations and correspondingly higher accelerations. By truncating the explosion at exit from the foil, we limit the relative velocities acquired by fragments from the large-r_o ions. The ring in fig. 3(a) is measured for the duoplasmatron-prepared HeH^+ beam and though the explosion ring diameter is maller, the distribution is similar to that of the full explosion seen with protons.[8] By contrast, fig. 3(b) shows a similar measurement performed with the beam prepared in an rf ion source. Here we see a ring collapsed to smaller fragment separation velocities (large r_o). This is characteristic of the hotter population of vibrational states in the incident beam.

This modification of the H^o ring distribution for HeH^+ produced by changing ion sources is similar to what is observed by comparing H^o from H_2^+ and HeH^+ from the same (duoplasmatron) source.[9] Whereas the cooler HeH^+ shows a characteristic ring distribution, the vibrationally excited H_2^+ gives

a dramatically different profile indicative of a significantly smaller energy release. It is worth noting that unlike HeH^+, the already hot r_o-distribution for H_2^+ does not change observably when the ring is remeasured for a beam prepared in an rf source.

Another very sensitive probe of the large-r_o component of the incident beam is the measured yield of transmitted molecular ions. The transmission phenomenon has now been well described quantitatively by a recent model proposed by Cue et al.[10] According to this model, the transmission probability increases with increasing internuclear separation and hence transmission yields are most sensitive to the tail on the high side of the r_o-distribution of the incident ions. As in the case of the neutral fragments, this is a consequence of the very weak explosion experienced by large-r_o molecules. Comparing the radial distributions of fig. 2, one would expect to see an enhanced yield of transmitted ions when the beam is prepared in the rf source as opposed to the yield when the beam is produced in the duoplasmatron. Figure 4 shows a comparison of transmission yields as a function of target thickness for HeH^+ beams from each source. One sees, as expected, that the hotter rf beam is more than twice as likely to be transmitted. Calculations performed with the model of ref. 10 reproduce the data fairly well when the radial distributions of fig. 2 are used.

To summarize, we've seen that measurements of the angular distributions of explosion fragments from dissociations in solid targets can give quantitative information about the distribution of internuclear separations contained in the incident molecular-ion beams. These distributions, while indicating vibrational excitation, are relatively independent of ion-source tuning. The measured radial distributions are sufficiently accurate to predict other phenomena (such as transmission). With

the exception of H_2^+, ions prepared with a duoplasmatron are vibrationally cool. HeH^+ beams produced in an rf ion source are vibrationally hotter than HeH^+ beams formed in a duoplasmatron. Measurements with such ions as HeH^+, CH^+, OH^+, $^3He_2^+$, and other light diatomics show these beams to be predominantly in the ground vibrational state when prepared in a duoplasmatron.[8] The H_2^+ beam appears to have a distribution intermediate between a Franck-Condon distribution of vibrational states and the cooler distributions observed for other ions.

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Figure Captions

Fig. 1. Angular distributions, measured at zero energy shift, for outgoing H^+ from the dissociation of 3.0-MeV HeH^+ in a 108-Å carbon foil. The solid curve was observed when the HeH^+ beam was prepared in a duoplasmatron ion source while the chained curve resulted from a similar measurement using an rf source.

Fig. 2. Radial distribution functions for incident 3.0-MeV HeH^+ ions. These curves were derived from the data shown in Fig. 1 using the methods described in Ref. 4. The solid curve is the distribution observed from a duoplasmatron source while the chained curve is the result of measurements utilizing an rf source.

Fig. 3. The joint energy-angle distributions ("ring patterns") for neutral hydrogen fragments arising from the dissocition of 2-MeV HeH^+ in carbon foils. The distribution in (a) is measured when the beam is prepared in a duoplasmatron ion source fed with a gas mixture of 90% He and 10% H_2 while (b) shows the distribution obtained with an rf source using the same gas mixture.

Fig. 4. Comparison of transmitted HeH^+ yield for 3.0-MeV ions incident on carbon targets of varying thickness when beams are prepared in duoplasmatron and rf ion sources. The lines represent the results of calculations (see ref. 10) using the two distributions of fig. 2 for initial radial distributions.

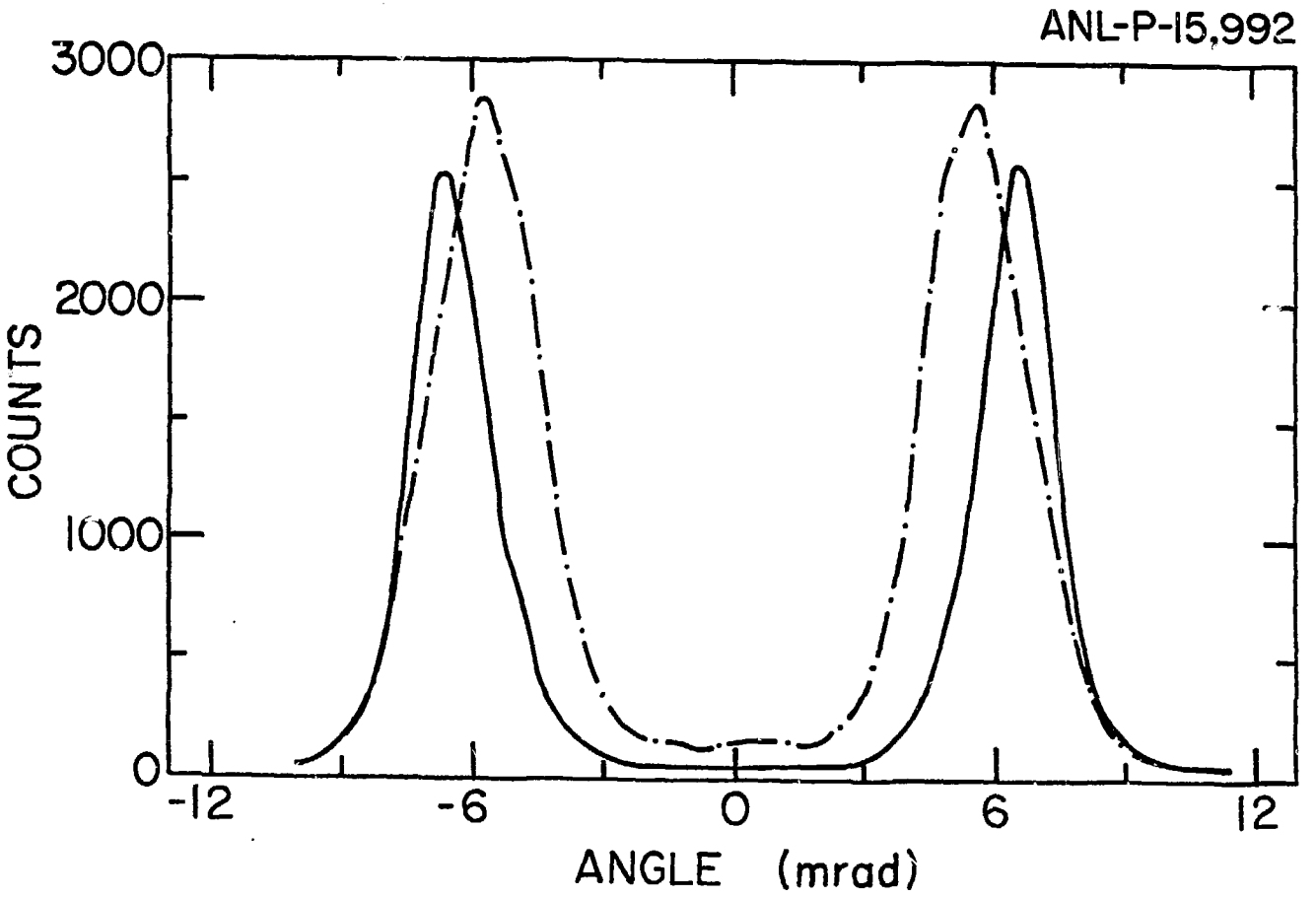
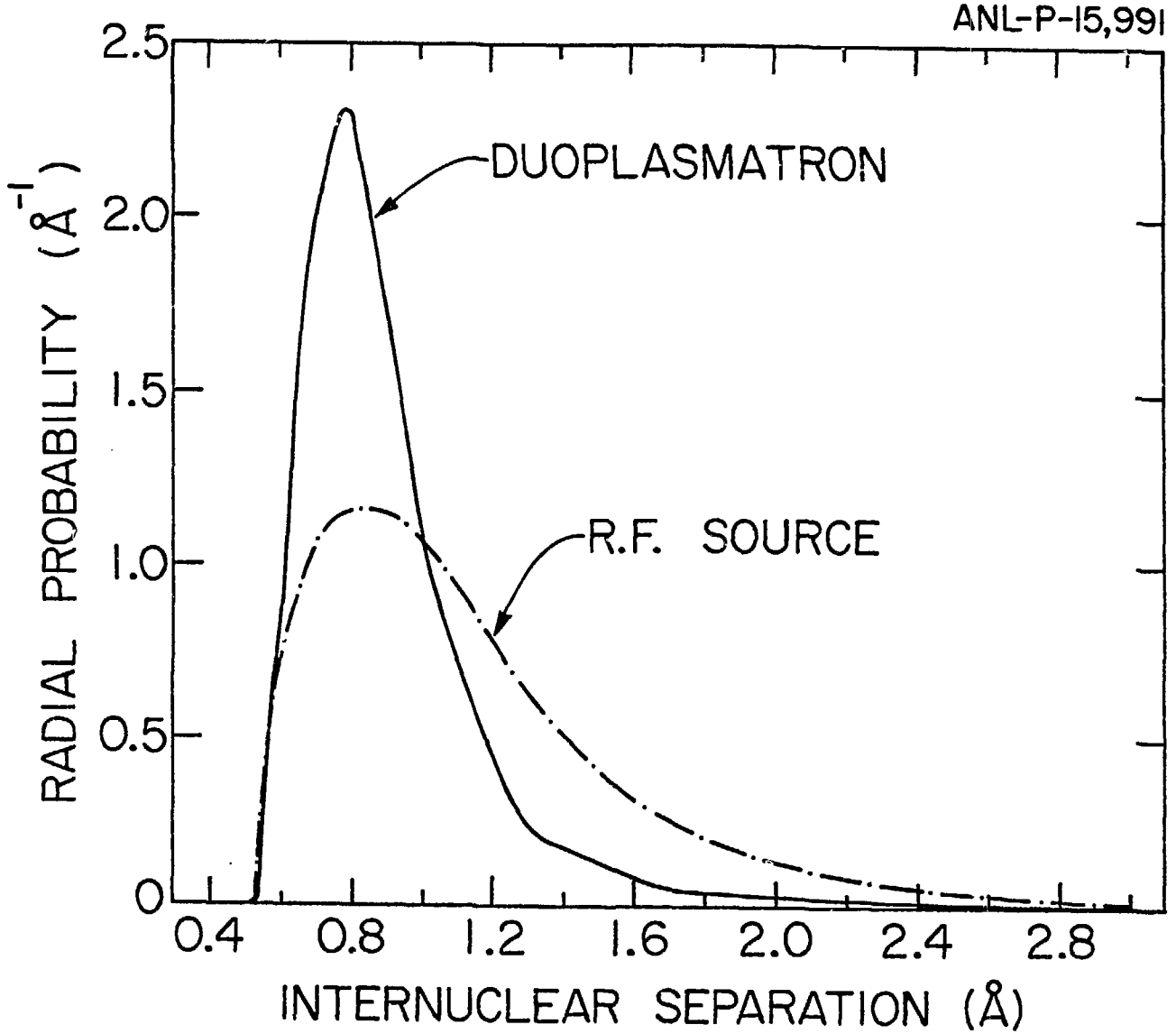


Fig. 1


ANL-P-15,991
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2.0
1.5
1.0
0.5
0
RADIAL PROBABILITY (Å^{-1})
DUOPLASMATRON
R.F. SOURCE
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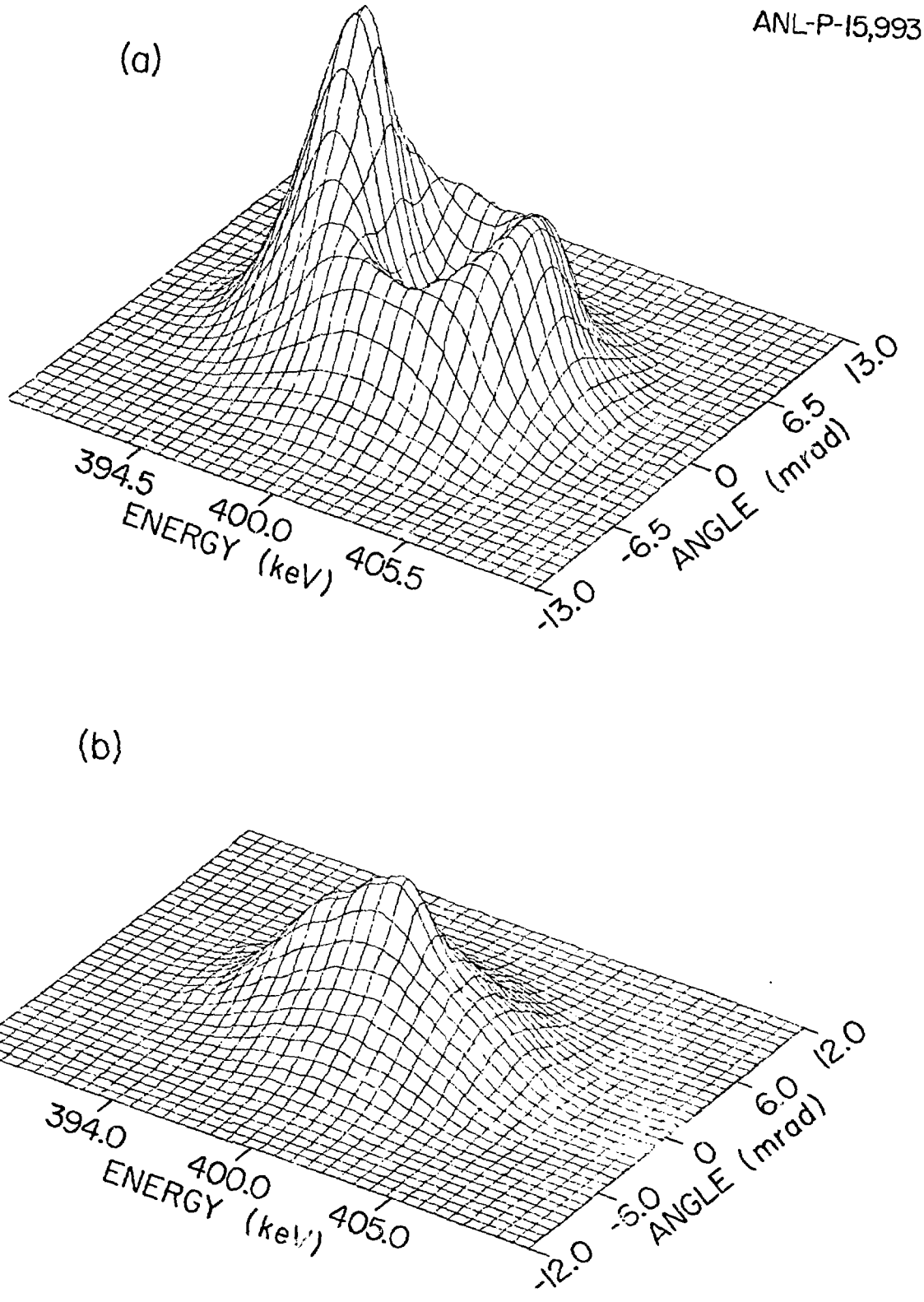
0.8
1.2
1.6
2.0
2.4
2.8
INTERNUCLEAR SEPARATION (Å)

Fig. 2


(a)
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400.0
405.5
ENERGY (keV)
13.0
6.5
0
-6.5
-13.0
ANGLE (mrad)
(b)
394.0
400.0
405.0
ENERGY (keV)
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6.0
0
-6.0
-12.0
ANGLE (mrad)

Fig. 3

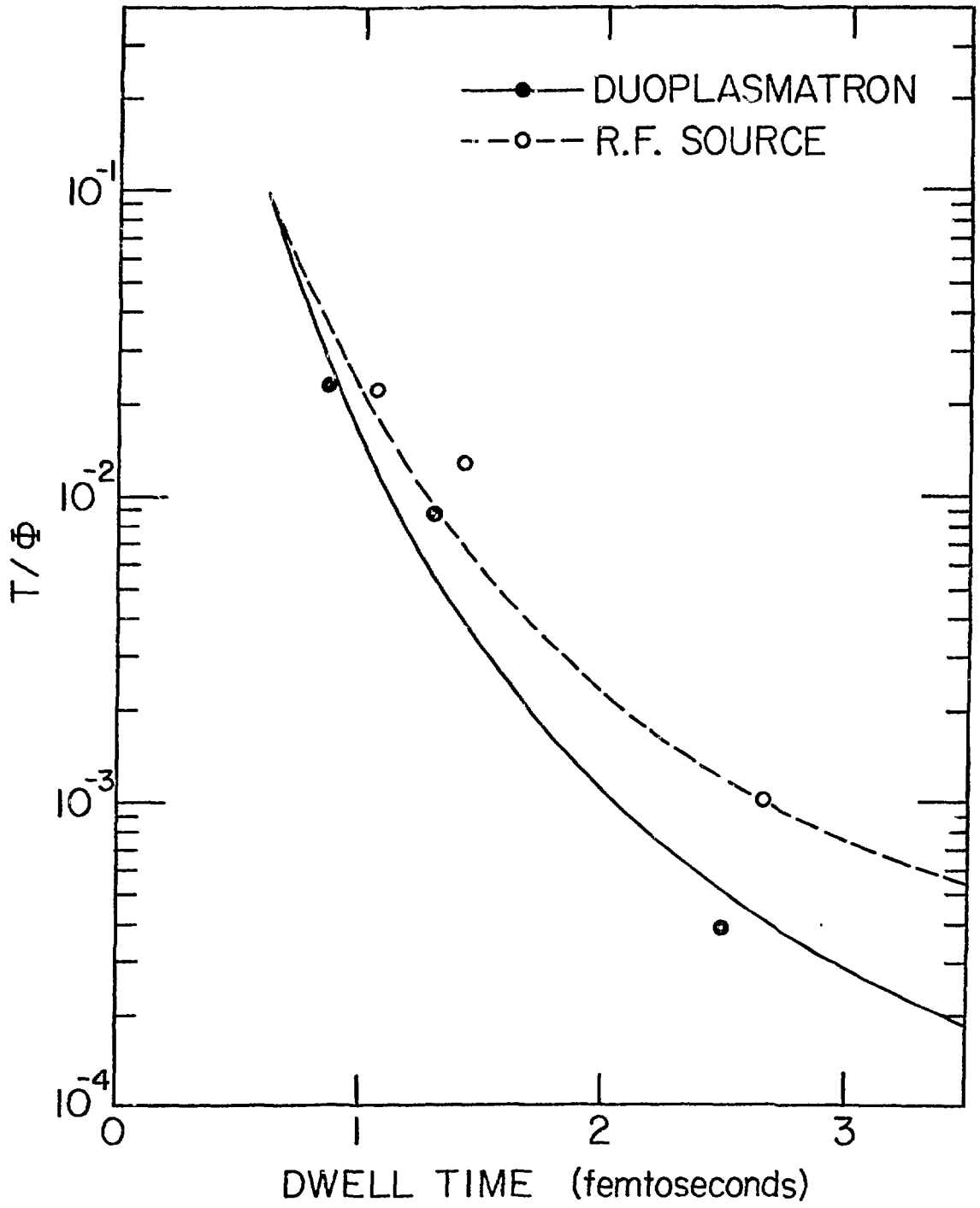
DUOPLASMATRON
R.F. SOURCE
10^{-1}
10^{-2}
10^{-3}
10^{-4}
T/Φ
0
1
2
3
DWELL TIME (femtoseconds)

Fig. 4