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COLLISIONAL EFFECTS IN THE PASSAGE OF FAST MOLECULAR IONS THROUGH THIN FOILS*

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Abstract: Some recent developments in studies on the interactions of fast molecular-ion beams with thin foil targets are discussed.

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1. INTRODUCTION

Experiments with fast (MeV) molecular-ion beams offer many attractive possibilities for studying atomic collisions in solids. Of particular value in such experiments is the possibility of determining the force fields (primarily the induced electric field) that surround ionic fragments traversing a solid. One has the opportunity to evaluate these fields not just at the fragments themselves (as one would, for example, in stopping-power measurements with monatomic projectiles) but in the spatial regions extending out to several Angstroms from the fragment positions.

In this paper we give a brief introduction to the subject and present some recent results. For more detailed information, the reader is reminded of the existence of the published proceedings^{1,2)} of two recent workshops on these topics. There also exist several recent review articles³⁻⁷⁾ including one⁸⁾ on the use of the experimental techniques to determine the stereochemical structures of molecular-ion projectiles.

In a typical experiment with fast molecular ions, a tightly collimated magnetically analyzed beam of molecular projectiles is directed onto a thin target foil (most commonly carbon, in order to reduce multiple-scattering effects). In the front surface layers of the foil, some or all of the electrons on each projectile are stripped off, so that, after a time typically about 10^{-16} sec (see, eg. ref.³⁾, there exists a cluster of fragment ions each with a well-defined equilibrium effective charge. These fragments separate in a "Coulomb explosion". The characteristic time for the Coulomb explosion to develop (a few femtoseconds) is usually comparable with the dwell time in the target. (For dwell times much less than 1 fsec, non-equilibrium effects are observed in which the emergent molecular fragments retain a memory of the initial electronic configuration of the projectile--see the paper by

Cue at this conference). Emerging from the foil, the fragments may capture sufficient electrons to reform a bound state of the initial projectile species (this is usually termed "transmission"), or some other species based on a combination of projectile fragments. Most commonly, a dissociative molecular state is formed which leads to individual monatomic fragment ions in various charge states. The emergent fragments may be in excited states that decay by photon^{9,10)} or electron^{11,12)} emission.

2. EXPERIMENTS WITH FAST LIGHT MOLECULAR PROJECTILES

a Dissociation

Figures 1 and 2 show results obtained for the joint energy-angle distributions of protons arising from the dissociation of 2-MeV HeH^+ ions in a thin carbon foil.^{13,14)} The observed "ring pattern" [e.g. fig. 1(c)] is readily understood in terms of the vector diagram shown in fig. 1(b), where the beam velocity $\underline{v}$ and the Coulomb explosion velocity $\underline{u}$ of the proton have magnitudes of about 10^9 cm/sec and 10^7 cm/sec, respectively. Figure 1(d) shows the result of a computer simulation¹³⁾. The diameter of the measured ring [fig. 1(c)] and the shape of the "rim" are well reproduced. The non-uniform distribution of proton intensity around the ring is a consequence of wake effects (see, eg. refs. 7 and 13) and is reproduced only qualitatively in the simulation. An improved simulation can be achieved by taking into account the rotational motion of the incident projectiles [see the following paper by Plessner¹⁶⁾]. From an analysis of the shape of the rim of the ring, one can derive the distribution, $D(r_0)$, of initial internuclear separations¹⁴⁾. This is shown in fig. 2 together with the population of vibrational states that is implied if one takes the HeH^+ electronic ground-state potential as being that calculated by Kolos and Peek¹⁵⁾. From fig. 2(c)

one sees that the "inner ring" feature seen in the calculation of fig. 1(d) is observable in a careful measurement---it is due to protons that continue to Coulomb-explode away from He^+ ion partners (~6% of all emerging ions) after leaving the foil.

Generally, computer calculations based on either a simple plasma wake model⁷⁾ or on a wake calculated^{7,17)} using Lindhard's dielectric function¹⁸⁾, agree well with experiment, especially when rotational motion of the projectiles is taken into account and when multiple-scattering and straggling effects are small.

b Transmission

Figure 3 shows a set of data on the transmission of fast molecular ions through thin foils. The phenomenon of molecular-ion transmission which was discovered by Poizat and Remillieux²¹⁾, has recently been quantitatively accounted for²²⁾ using a model in which the molecular projectiles dissociate upon entering the target and then are reconstituted by capturing one or more electrons as they exit the target.

The following factors act to increase the transmission of molecular ions through foils:

- i) Short dwell times. The internuclear separations and relative momenta of the fragments upon exit are then more likely to favor reconstitution of the projectile.
- ii) A weak Coulomb explosion inside the foil. This can result from large values of r_0 (the initial internuclear separation), low values of the effective charges inside the foil, and large values of the reduced mass of the projectile fragments.
- iii) Multiple scattering. This is essential in order to provide a fraction

of the beam that, after a long dwell time, has exit fragment separations and momenta suitable for molecular reconstitution^{22,23}). The calculations of Cue et al.²²) indicate that without multiple scattering, no transmission of H_2^+ could occur for dwell times longer than about 4 fsec.

- iv) Low projectile velocity. This increases the electron capture probability upon exit.
- v) The need to capture only a few electrons (e.g. one).
- vi) Favorable spatial orientation of the exiting fragments. The data of Eckardt et al.²⁴) and Levi-Setti et al.²⁵) indicate that the transmission of low-velocity (~ 1 a.u.) H_2^+ is favored when one proton directly trails the other ("longitudinal" orientation).

The greatly increased transmission of H_2^+ as compared with HeH^+ is primarily due to factors ii) and v). The distribution $D(r_0)$ extends to much larger values of r_0 for H_2^+ than for HeH^+ . Similarly, the inability of experiments to detect transmission for O_2^+ , CO^+ , OH^+ , etc. is a result of factor v). In an experiment at Argonne²⁶), we have shown that transmitted 1.0-MeV H_2^+ molecules have internuclear separations larger than those in the primary beam (fig. 4).

c. Reconstitution of other species

Figure 5 shows an example of the reconstitution of a diatomic species, D_2^+ , from a triangular triatomic projectile, D_3^+ . The process is evidently highly orientation-dependent--if it were not, the contour plot in fig. 5(d) would be circular with equal diameters in parallel and transverse velocities.²⁷) The data of fig. 5 indicate that leading or trailing deuteron pairs are more likely to pick up a binding electron than "sideways-going"

pairs in which one deuteron trails another. In this latter orientation, capture of a binding electron only occurs when the third deuteron is far away. The energy spectrum in fig. 5(b) contrasts with that found by Cue et al.²⁸⁾ for H_2^+ arising from 2.2-MeV H_3^+ . There, a reverse asymmetry appeared and was explained by the authors as resulting from disruption of the binding of trailing H_2^+ ions by the wake of the leading H^+ .

d. Rare charge states

The ring patterns obtained for rare charge states frequently display strong orientation dependences. The "peculiar" distributions obtained for H^0 produced from the bombardment of thin carbon foils with H_2^+ and HeH^+ are examples^{14,19)}.

For H^- produced from the foil dissociation of HeH^+ , the ring pattern is found to be indistinguishable in shape from that of the H^0 fragments¹⁹⁾. This strongly suggests that the H^- fragments arise from a molecular state formed upon foil exit in which the dissociation proceeds as in the H^0 case (most probably to form He^{++} and H^0) but now in the presence of either a loosely bound ("Rydberg") or continuum ("convoy") electron. This electron, although not much affecting the dissociation energy, has some probability of attaching to the H^0 fragment to yield H^- .

Similar evidence for processes of this type is to be found in the spectra of H^0 fragments from the foil-induced breakup of OH^+ projectiles^{29,30)}.

e. Effects due to modification of $D(r_0)$

It is of interest to see to what extent the distribution $D(r_0)$ is affected by ion-source parameters such as source type, gas pressure,

extraction voltage, gas mixture, etc. At Argonne, after much effort, we have succeeded in finding only one case where $D(r_0)$ can be easily and significantly modified. This is for HeH^+ produced from an r.f. source as contrasted with our more usual duoplasmatron source. Figure 6 shows how the higher vibrational excitations in HeH^+ ions from the r.f. source manifest themselves in various effects (see the contribution by Kanter et al.³¹) at this conference).

f. Cluster stopping powers

Following the discovery by Brandt, Ratkowski and Ritchie³²) of the influence of cluster effects upon the slowing down of ions in solids, there have been several experimental and theoretical investigations of this phenomenon (see, for example, the article by Arista³³) and references contained therein). Figure 7 shows some recent data on the slowing down of "longitudinally aligned" protons from 800-keV H_2^+ . In general the cluster stopping powers for light projectiles like H_2^+ are fairly well described on the basis of wake models derived from Lindhard's dielectric function (see also, refs. 5, 24, 25, 32, 33 and 34).

g. Extended measurements on wake potentials

To further test the applicability of various wake models, it is desirable to study fragment interactions over internuclear separations comparable with wake wavelengths (typically several Angstroms). Going to thicker targets would permit this, but unfortunately multiple-scattering effects effectively blur out the experimental resolution and sensitivity. This difficulty can be largely overcome by employing channeling in a monocrystalline target (where multiple scattering is greatly reduced). Such

experiments have been performed previously (eg. ref. 35) but not with adequate energy resolution. Figure 8 shows results from a recent molecular-ion channeling measurement³⁶⁾ indicating the feasibility of such experiments in relatively thick targets. (If one were to ignore the influence of the crystal upon the motion of the fragments, the proton and helium fragments in the experiment depicted in fig. 8 would be separated at exit by about 7 Angstroms, i.e. about 0.4 plasma oscillation wavelengths.)

An alternative method for the study of long-distance wake interactions is provided by the two-foil technique being pursued by Kumbartzki et al.³⁷⁾ at the Bonn cyclotron.

3. EXPERIMENTS WITH SLOW HEAVY MOLECULAR PROJECTILES

It is of interest to see to what extent current wake theories are applicable to the case of slowly-moving heavy projectiles where the effective charges are significantly less than the nuclear charges. To this end, we have recently initiated at Argonne a series of measurements on the dissociation³⁸⁾ and stopping power³⁴⁾ of N_2^+ projectiles in the energy range 1.0 to 3.6 MeV.

a. Dissociation

Figure 9 shows ring patterns measured for N^+ and N^{4+} fragments emerging from a 73-Å carbon foil bombarded by 3-MeV N_2^+ . The ring diameters can be fairly well understood in terms of a model in which the nitrogen fragments repel each other inside the foil with the same (not necessarily integral) effective charges that determine their individual stopping powers. Outside the foil, the detected fragments (either N^+ or N^{4+} in this case) continue to Coulomb-explode away from their partners which can assume one of several possible integral charge states. (The probability distribution for

these charge states can be measured and is close to that obtained with monatomic projectiles.) Thus, although inside the foil the effective charges of the detected N^+ and N^{4+} are the same, the ring diameter measured for N^{4+} is larger than for N^+ .

In figs 10 and 11 are shown comparisons of these measured ring patterns with rings calculated using three different wake models. The calculations are performed with the "Fokker-Planck" technique described in the following paper by Plesser.¹⁶⁾ The distributions $D(r_0)$ were derived using Franck-Condon factors to estimate the population of vibrational levels in the X and A states of N_2^+ . This gave a most probable value of $r_0 = 1.12 \text{ \AA}$ in good agreement with the value tabulated by Huber and Herzberg³⁹⁾.

The three wake models tried were:

- 1) "Lindhard". Obtained by numerical integration¹⁷⁾ of Lindhard's dielectric function for a Fermi gas of free electrons.
- 2) "Vager-Gemmell". A simple classical plasma wake¹³⁾ based on the high-frequency approximation to the dielectric function

$$\epsilon(\omega) = 1 - \frac{\omega_p^2}{(\omega + i\gamma)^2},$$

where γ is a damping parameter.

- 3) "Coulomb". Based on a polarization charge distribution derived from Coulomb wave functions

$$\rho_{pol}(\underline{r}) = ne \langle |\psi_c(\eta, \underline{r})|^2 \rangle_u - 1),$$

as described by Faibis et al.⁴⁰⁾.

It is clear from figs. 10 and 11 that none of the computer

simulations fits the experimental data very well. In particular, the "Lindhard" wake gives a very poor fit. This is no great surprise since a linear-response theory, in which the projectile charge is treated as a perturbation in the electron gas, is not expected to be valid for these heavy ions. One might expect the best fit to be given by a non-linear wake model of the "Coulomb" type. Figure 12 shows the quality of fit obtained using the "Coulomb" wake (c.f. fig. 9).

b. Cluster stopping power

Figure 13 shows the stopping power ratio, R , (defined as the average rate of slowing down for the two detected N^+ fragments from N_2^+ divided by the rate of slowing down for a monatomic N^+ beam of the same velocity) as a function of dwell time in a carbon target, for N_2^+ energies between 1 MeV ($v = 1.2$ a.u.) and 3.6 MeV ($v = 2.28$ a.u.). Only N^+ fragments from fragment pairs that were "longitudinally" aligned upon foil exit, were detected in these measurements. In contrast with most of the data for light molecular projectiles, the stopping power ratios thus determined for N_2^+ beams are all less than unity. (Crudely speaking, this is because much of the induced negative polarization charge now lies between the two ion fragments.)

As indicated in figs. 14 and 15, none of the calculations that have so far been performed, based on the three wake models described above, is able to reproduce the experimental R -values. In fact, the least bad fit is obtained with the "Lindhard" wake and the worst with the "Coulomb"—just the reverse from the situation in fitting the fragmentation patterns.

c. Validity of wake models

It is clear that none of the wake models tested thus far is able to account simultaneously for both the fragmentation and the stopping power

measurements. This failure is not unexpected. The main factors limiting the applicability of these wake models to the N_2^+ problem are:

- i) A non-linear theory is clearly required. (Of the three models tested, only the "Coulomb" wake is not of a linear-response type.)
- ii) Although the "Coulomb" wake was used successfully by Faibis et al.⁴⁰⁾ to describe the dissociation of OH^+ , it is expected to be of limited validity for N_2^+ . This is because two heavy ions are involved, whereas in the OH^+ case the proton can be treated as a perturbation.
- iii) The fragments are not point charges. Although the point-charge approximation may be useful in describing the slowing down of monatomic heavy-ion beams, it has obvious limitations in describing the force fields away from the ion centers.
- iv) The target foils are not uniform Fermi gases. This feature is expected to become more pronounced for heavy ions.
- v) There are indications that strong re-orientation and wake-trapping effects play a significant role in the motions of the fragments within the foil. These effects have not yet been taken into account in calculations.

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Figure Captions

- Fig. 1. Results for protons arising from 2-MeV HeH^+ projectiles bombarding an 85-Å thick carbon target^{13,14}).
- Fig. 2. (a) Distribution $D(r_0)$ determined from experiment¹⁴) on the dissociation of 2-MeV HeH^+ (solid curve) together with the calculated $D(r_0)$ assuming population of only the ground state of HeH^+ (dashed curve).
- (b) Vibrational levels of HeH^+ (ref. 15) together with their populations determined from a HeH^+ breakup experiment¹⁴).
- (c) Energy spectrum for protons emerging in the beam direction from 2-MeV HeH^+ bombarding a 59-Å carbon foil.
- Fig. 3. Transmitted fractions, T , of fast beams of H_2^+ , $^4\text{HeH}^+$, $^3\text{HeH}^+$, $^3\text{He}_2^+$ and D_3^+ through thin foils¹⁹). All of the data points except the one labelled " Al_2O_3 " were obtained with carbon targets. The dashed curve for 2-MeV H_2^+ represents data taken by the Lyon group²⁰).
- Fig. 4. Energy distributions for protons emerging at 0° from 1-MeV H_2^+ incident upon a 75-Å carbon foil. The data for the dashed curve were obtained with the direct beam from the accelerator, while the solid curve is for H_2^+ that had previously been transmitted through a 330-Å carbon foil and then magnetically analyzed²⁶).
- Fig. 5. (a), (b), and (c). Energy and angle distributions for D_2^+ arising from 3.6-MeV D_3^+ incident upon a 216-Å carbon foil. (c) A contour plot of the D_2^+ intensity as a function of transverse and parallel velocities (arb. units) in the c.m. The two velocity scales in (c) are equal²⁷).
- Fig. 6. Comparison of effects seen for protons from the foil-induced dissociation of 2-MeV HeH^+ ions produced from an r.f. source and a

duoplasmatron source³¹⁾. a) angle spectrum, b) distribution $D(r_0)$, c) HeH^+ transmission and d) ring patterns for H^0 .

Fig. 7. Stopping power ratio, R , (i.e. the average rate of slowing down for the two protons from H_2^+ compared with that for protons of the same velocity) for 800-keV H_2^+ in aluminum [open circles (experiment) and long-dashed line (calculation)] and in carbon [crosses (experiment) and short-dashed line (calculation)]. Only protons emerging in the beam direction from the target are counted³⁴⁾.

Fig. 8. Ring pattern for protons emerging in the (111) planar direction from a gold crystal 950 Å thick bombarded by 3-MeV HeH^+ incident along the same (111) planar direction. In Fig. 8(d), the velocity scales are arbitrary, but equal. The angular coordinate in Figs. 8(a) and (c) is measured with respect to the beam direction in the (111) plane³⁶⁾.

Fig. 9. Ring patterns for N^+ (a, b and c) and N^{4+} (d, e and f) fragments arising from the bombardment of a 73-Å carbon foil by 3-MeV N_2^+ projectiles³⁸⁾.

Fig. 10. Comparison of the experimental ring pattern (Fig. 9) for N^+ with patterns calculated on the basis of three different wake models as described in the text.

Fig. 11. Same as Fig. 10 except that the fragments are now N^{4+} .

Fig. 12. Calculated ring patterns for N^+ (a, b and c) and N^{4+} (d, e and f) fragments from the dissociation of 3-MeV N_2^+ in a 73-Å carbon foil³⁸⁾. The calculations used a "Coulomb" wake (see text).

Fig. 13. Stopping power ratio (see text) for N_2^+ projectiles of various energies penetrating carbon foils³⁴⁾. Only N^+ fragments from "longitudinally" oriented fragment pairs are detected.

Fig. 14. Comparison of the measured R -values for 3-MeV N_2^+ projectiles with

calculated values based on the various wake models described in the text. The data pertain to the case of "longitudinally" aligned projectiles³⁴).

Fig. 15. As in Fig. 14, but with incident N_2^+ energies of (a) 1 MeV and (b) 3.6 MeV.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6

Fig. 7

Fig. 8

ANL-P-16,225

$N_2^+ \rightarrow N_2^+$

Fig. 9

Fig. 10

Fig. 11

Fig. 12

Fig. 13

Fig. 14

Fig. 15