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PHYSICS WITH FAST MOLECULAR-ION BEAMS

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Summary

Fast (MeV) molecular-ion beams provide a unique source of energetic projectile nuclei 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. The general features of several such experiments will be discussed and recent results will be presented.

Introduction

Although fast molecular-ion beams have been used for many years in the energy calibration of nuclear accelerators,^{1,2} it was not until quite recently that experimenters began to take advantage of the spatial and temporal correlation of the nuclei constituting the molecular projectiles. Among the earliest to recognize this unique property, Golovchenko and Laegsgaard³ used a 2-MeV H_2^+ beam in an unsuccessful attempt to study the effect of proton correlation on the "inelastic excitation of a solid carbon foil". Those authors measured the first "ring pattern" (a joint energy-angle distribution) for protons resulting from the dissociation of H_2^+ beam ions after passage through a $2 \mu g/cm^2$ ($\sim 100 \text{ \AA}$) carbon foil.

That experiment has been followed by further studies^{4,5} of this Coulomb explosion phenomenon, as it has now become called, and such measurements have begun to provide interesting new information about the charge states of fast ions inside and outside of solids, the interactions of such ions with the solid medium, and a variety of other atomic collision phenomena. Recently, it has been demonstrated that these techniques can be applied to the difficult problem of determining the geometric structures of molecular ions.^{6,7}

The apparatus shown in Fig. 1 has been used at Argonne for the past couple of years to perform high-resolution studies of the foil- and gas-induced dissociation of fast molecular-ion beams. Magnetically analyzed molecular-ion beams from Argonne's 4-MV Dynamitron accelerator are collimated to have a maximum angular divergence of $\pm 0.09 \text{ mrad}$ at the target position. A set of "pre-deflector" plates permits electrostatic deflection of the beam incident on the target. Similarly, a set of "post-deflectors" is used to deflect charged particles emerging from the target. Charged particles entering the electrostatic analyzer are energy-analyzed with a relative resolution of $\pm 3 \times 10^{-4}$. The angular acceptance of the analyzer is $\pm 0.11 \text{ mrad}$. Distributions in angle and energy are made for particles emerging from the target by varying the voltages on the pre-deflector and/or post-deflector in conjunction with that on the analyzer. Neutrals are energy-analyzed by first stripping them in a $\sim 100 \text{ \AA}$ thick carbon foil located just ahead of the analyzer. The overall angular resolution is $\pm 0.15 \text{ mrad}$ (0.008°). Selection of the required charge state for particles leaving the target is facilitated by a suitable combination of pre- and post-deflection. The optimal combination also rejects spurious incident beams (e.g. pre-dissociated fragments arising from interactions in residual gas upstream from the target). Elaborate precautions are taken to ensure that no carbon buildup occurs on the target foils. A gas target can also be used in lieu of foils. Recently, this arrangement has been modified to permit coincident detection of several dissociation fragments.^{6,7}

Figure 2a shows a typical distribution measured for protons resulting from the dissociation of HeH^+ ions incident on a thin carbon foil.⁸

Interpretation

The diameters of these measured ring patterns are primarily determined by the bond lengths of the incident projectiles and the charge states of the fragment ions inside and outside the target. For a foil target, it is

Fig. 1. Schematic diagram of the experimental arrangement used in experiments at Argonne's 4-MV Dynamitron accelerator.

Fig. 2. Ring patterns for protons resulting from the foil-induced dissociation of 2-MeV HeH^+ ions after passage through an 85 Å carbon foil. (a) shows the experimental distribution while (b) is the result of a calculation (from Ref. 8).

reasonable to assume that in the rest frame of the incident molecular projectile (hereinafter referred to as COM), the constituent nuclei are frozen in space, rotationless and vibrationless (see Ref. 9 for a discussion). The vibrational and rotational excitation of the beam is however manifest in the distribution of internuclear separations, $D(r)$ contained in the incident beam molecules.

For light projectiles (H_2^+ , HeH^+ , He^+ , etc.) incident at these energies upon a foil, the electrons that bind the projectile are almost always totally stripped off within the first few Å of penetration into the solid target. This is a consequence of the large cross sections ($\sim 10^{-16} \text{ cm}^2$) for electron loss¹⁰ and the fact that close collisions with target electrons cannot be avoided in a solid. There then follows a "Coulomb explosion" in which the bare (or nearly bare) nuclei of the projectile fly apart by virtue of their mutual Coulomb repulsion. The characteristic time for this explosion is typically $\sim 10^{-15} \text{ sec}$, which is on the same order as the dwell time of the projectile in the foil if the latter is about 100 Å thick. It is therefore to be expected that much of the Coulomb explosion takes place inside the foil and that it then runs to completion in the vacuum downstream after the fragments emerge from the target. Inside the target, the fragments each carry an effective (not necessarily integral) charge which determines the magnitude of the Coulomb explosion. The Coulomb repulsion inside the target can be assumed to be dynamically screened with a screening distance $a = V_0/\omega_p$, where ω_p is the volume plasma frequency of the target and V_0 is the projectile velocity. The length $2\pi a$ is simply the distance the fragments travel during one oscillation period of the target electrons. Once outside the target there is

then no further screening and the Coulomb repulsion will continue as the fragment ions adopt final integral charge states.

In a typical measurement, we measure in the LAB frame the yield of a dissociation fragment as a function both of its angular shift (θ_x or equivalently, θ_y) from the direction of the incident beam (z-direction) and of its energy shift (ΔE) from the value $E = \frac{1}{2} m_f V_0^2$ corresponding to the fragment's share of the projectile's kinetic energy (m_f is the fragment's mass, and V_0 is the beam velocity). These LAB variables are related in a simple way to the asymptotic COM velocity v acquired by a fragment following the dissociation of a projectile. Since the momentum transfer is very small in most of the collisions leading to dissociation at these incident energies, we can assume that the projectile COM is undeflected in the collisions. If $V = V_0 + v$ is the asymptotic LAB velocity of the fragment, then, for $v \ll V_0$

$$\theta_x = v_x/V_0; \quad \theta_y = v_y/V_0; \quad 1a)$$

$$\Delta E = \frac{1}{2} m_f (V^2 - V_0^2) \approx m_f v_z V_0. \quad 1b)$$

and

$$\Delta E/E \approx 2v_z/V_0. \quad 1c)$$

For a diatomic projectile, the Coulomb potential energy immediately after the loss of the binding electrons is $Z_1 Z_2 e^2/r$, where $Z_1 Z_2$ are the effective charges on the ions as they begin to separate from an initial distance r . (For fast light ions and solid

targets, Z_1 and Z_2 are very nearly equal to the atomic numbers of the fragments.) This Coulomb energy is typically several tens or even hundreds of eV. In comparison, we may normally neglect the small energies of vibrational and rotational motion that the projectile may possess at the beginning of the Coulomb explosion. The COM velocity subsequently achieved by a fragment of mass m_f is then

$$v = (1/m_f)(2\mu Z_1 Z_2 e^2/r)^{1/2}, \quad (2)$$

where μ is the reduced mass. [In some cases, e.g. when treating dissociation in a gas target, it is obviously more appropriate to write Eq. (2) in terms of the proper dissociative potentials for the electronic states involved—see Ref. 9.]

From the foregoing, one sees that the diameter of an observed ring pattern is approximately proportional to the geometric mean of the effective charges on the fragments and inversely proportional to the square root of the bond length. Experiments with several species of diatomic molecular-ion projectiles have established that the bond lengths obtained from such data agree well (typically within 0.01 Å) with calculated values when the effective charges Z_1 and Z_2 are chosen to have values that give the correct stopping power.

Wake Effects

Assuming that the incident molecular-ion projectiles enter the target foil with a uniform population of all possible orientations of the internuclear axes, then for a simple Coulomb explosion, the dissociated fragments would uniformly populate a thin spherical shell in COM velocity space. Our measured angle-energy ring distributions are simply equatorial cuts of these spherical distributions so that one would expect the intensity for a given fragment species to be uniformly distributed around the ring. However, for foil targets (see Fig. 2a) the measured patterns display a pronounced peaking of intensity at positions along the ring corresponding to alignment of the projectiles' internuclear axes with the beam direction.¹¹

This phenomenon was accounted for in terms of the interaction between the fragment ion and the electron polarization "wake" induced by its partner fragment.⁸ Figure 2b shows a simulation of the measured ring calculated on the basis of the model proposed in Ref. 8. It should be noted that although the idea of an electron "wake" trailing fast-moving ions in solids was first proposed by Bohr¹⁰ in 1948, it was not until these experiments with molecular ions that this wake was experimentally probed.

Radial Distributions

Calculations⁸ on the influence of wake forces have shown that they modify the Coulomb explosion least when the projectile's internuclear vector is perpendicular to the beam direction. Angular distributions for fragments emitted in this transverse orientation can therefore be used to extract the distribution of internuclear separations contained in the beam.¹² After deconvoluting the effects of multiple scattering from an angular distribution (at zero energy shift) the resulting velocity spectrum $G(v)$ can be simply related to $D(R)$.

In our analysis we assume that in a dissociative collision the molecular projectile makes a sudden electronic rearrangement while leaving its nuclear

constituents unperturbed. That is, we consider the projectile as undergoing a sudden vertical transition up to some excited electronic state which then dissociates liberating a total COM kinetic energy of $U(r) - U(\infty)$, where $U(r)$ is the potential energy at an internuclear separation r for the particular final electronic state involved. For the case of solid targets, $U(r)$ is assumed to be a simple Coulomb potential.

If we now further assume that the cross section for the electronic excitation is independent of the spatial orientation of the projectile and also independent of its internuclear spacing, then we can readily derive the relationship between the distribution functions for v and r .

$$G(v) = (4\pi v^2)^{-1} D(r) dr/dv, \quad (3)$$

where v and r are related via Eq. (2) and the distribution functions are normalized by

$$\int_0^\infty D(r) dr = 1, \quad (4)$$

and

$$4\pi \int_0^\infty G(v) v^2 dv = 1. \quad (5)$$

The distribution $D(r)$ so derived for 3.63-MeV HeH^+ ions produced by a duoplasmatron source is shown in Fig. 3.

Fig. 3. Radial distribution function for incident 3.63-MeV HeH^+ ions. The ions are prepared in a duoplasmatron ion source fed with a gas mixture of 90% He and 10% H_2 . The solid curve is the best fit to the data, obtained by adjusting the fractional population of each of the eleven vibrational levels of the electronic ground state of HeH^+ . The chained curve is the distribution for the ground-vibrational state only. [See Refs. 9 and 12 for details].

Fig. 4. Joint energy-angle distributions for protons from 1.2-MeV H_2^+ and 3.0-MeV HeH^+ dissociating in gaseous Ar (7.8 mtorr) and He (9.5 mtorr), respectively (Ref. 9).

Dissociation in Gas

As one might expect, ring patterns for fragments arising from dissociations in gas targets are strikingly different (Fig. 4) from those observed with solid targets (Fig. 2). Most noticeably, there is an absence of any wake alignment. Also, because of the predominance of large impact parameter collisions in the gas target, one expects to see dissociations through repulsive excited electronic states of the molecule thereby producing smaller laboratory velocity shifts.⁹ This is evidenced in Fig. 4 by the intense central peaks.

Charge-State Effects

Because of the proximity of the ions emerging from the foil, Coulomb explosion experiments provide a way to study the formation of final charge states of heavy ions. Already, several experimenters have reported charge-state effects.¹³⁻¹⁷ By studying thickness dependence¹⁴ and final charge-state dependence¹³ of the energy-angle distributions for heavy-ion explosion fragments, it is possible to separate the parts of the Coulomb explosion which occur inside and outside the target foil. The data have been found to be consistent with a model in which the heavy-ion charge state is constant in the foil and then rapidly assumes one of a range of integer values upon emergence into the vacuum.

For short dwell times in the target (so that the fragments are close upon exit), fragment charge-state distributions are found to be shifted to lower values.^{16,17} Also for short dwell times, this enhanced electron capture appears to be orientation dependent.¹⁵

This orientation dependence can be seen in the ring distributions in Fig. 5 measured for N^+ and N^{4+} fragments from the foil-induced dissociation of 3-MeV

N_2^+ ions.¹⁸ Although the intensity variation along the ring varies strongly according to final charge state, there do not appear to be any correlations in the charge states of fragments from the same parent molecule.¹⁸

Wake Models

Finally, we note that these recent measurements of the Coulomb explosions of heavy molecular ions provide a sensitive test of wake models.¹⁹ In experiments at the Weizmann Institute, the ring distributions of protons, resulting from the foil-induced dissociation of 11.2-MeV OH^+ ions, have been measured in coincidence with various oxygen fragment charge states.^{14,20} By studying the motion of protons in the wake of a highly charged ion, these experiments provide a test of the linear response model of the wake used in Ref. 8. These experiments indicate (see Fig. 6) that a more realistic model of the wake is required.^{19,21}

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Fig. 5. Joint energy-angle distributions for (a) N^+ and (b) N^{4+} fragments from the foil-induced dissociation of 3-MeV N_2^+ ions (Ref. 18).

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Fig. 6. Ring patterns for protons from the dissociation of 11.2-MeV OH^+ ions in a $740 \text{ \AA}$ thick carbon foil (Ref. 20). The experimental data are at the top right while the remaining distributions are computer simulations using different wake models (Ref. 19) based on Coulomb wave functions (top left), Lindhard's dielectric function (bottom left) and a plasmon wake (bottom right).