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Prepared For
VI International Conference on
Atomic Collisions in Solids
Amsterdam
September 22-26, 1975

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ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

**operated under contract W-31-109-Eng-38 for the
U. S. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION**

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Some New Effects Seen in the Passage of Swift Ion Clusters Through Solids^{*}

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Distributions in angle and in energy have been measured for ions transmitted through thin solid targets bombarded by various molecular-ion beams. The results, which differ markedly from those expected on the basis only of a simple "Coulomb explosion", indicate the existence of an additional force tending to align the internuclear vectors of each cluster with the beam direction. An explanation is suggested in terms of a potential attributable to the plasma oscillations generated by the particles as they traverse the solid. Calculations are presented to substantiate this idea.

We report on some measurements and calculations of the distributions in energy and angle for the dissociation products created when tightly collimated beams of fast molecular ions bombard thin solid targets. At the energies employed (0.15–2.0 MeV per nucleon) it is to be expected [1] that the electrons binding an incident molecular ion will be torn off within the first one or two atomic layers encountered in the target and that the remaining molecular constituents will then continue as a cluster of naked nuclei exploding apart under the influence of their mutual Coulomb repulsion. Our measurements reveal large and hitherto unknown departures from the results expected for clusters undergoing simple Coulomb explosions [2].

As a pertinent example of the behavior expected from a simple Coulomb explosion, let us consider the protons created by the dissociation of a beam of 4-MeV (${}^4\text{HeH}^+$) incident on a 535-Å thick Au foil (similar considerations will, of course, apply to the α -particles simultaneously created). The incoming molecules will, in general, populate various vibrational and rotational states. One can make estimates of the relative populations of these states based on a consideration of conditions in the accelerator ion source and of decay probabilities during the flight to the target (see, for example, References 3 and 4 for the corresponding discussions pertaining to the case of H_2^+). But for our present purposes, it suffices to note that the potential well [5] for vibrational states in (${}^4\text{HeH}^+$) is not very asymmetric about the average internuclear separation ($\bar{r}_0 \approx 0.8 \text{ Å}$) in the ground state and that whatever the population distribution of the vibrational states might be, the shape of the

potential well is such that one expects a distribution in r_0 (the internuclear separation) having a width of about $0.5 \text{ \AA}$. More precise estimates can be made for molecular ions for which the formation mechanism is better understood. [3,4]. Figure 1 illustrates such a case—that of H_2^+ .

In our $(^4\text{HeH})^+$ example, let us consider an incident molecular ion for which, at the instant $t = 0$ of entering the target, the internuclear vector drawn from the α -particle to the proton has a length $r_0 = 0.8 \text{ \AA}$ and makes an angle θ with the incident beam direction. If we make the assumption that the two binding electrons are instantaneously removed at $t = 0$, then the velocity of the proton in the laboratory system after a time t is

$$\tilde{u} = \tilde{V} + \tilde{v}, \quad (1)$$

where $\tilde{V}$ is the incident beam velocity and

$$\tilde{v} \approx \tilde{v}_{\text{molec.}}(t = 0) + \tilde{v}_{\text{CE}}(r_0, t). \quad (2)$$

Here $\tilde{v}_{\text{molec.}}$ represents the instantaneous value (at foil entrance) of the proton's cm velocity due to vibrational and rotational excitation of the incident molecular ion and $\tilde{v}_{\text{CE}}$ is the velocity acquired by the proton as a result of the conversion of the initial Coulomb potential energy ($2e^2/r_0$) into kinetic energy. For an initial separation $r_0 = 0.8 \text{ \AA}$, the proton's share of this Coulomb energy (neglecting the relatively small amount of kinetic energy given to the screening electrons of the target) is about 29 eV. The time

required to reach 90% of the final value of $\tilde{v}_{CE}$ is 5×10^{-15} sec. (For comparison, the time to traverse the foil is 4.2×10^{-15} sec.) The vibrational energy that the proton has in the incoming molecule is typically on the order of 1 eV and the energy associated with molecular rotational excitations is typically an order of magnitude smaller still. Thus, "late" ($t \gg 10^{-15}$ sec.) in the Coulomb explosion, the second term in Eq. (2) far exceeds the first term, while the reverse is true "early" ($t \ll 10^{-15}$ sec.) in the explosion. Although the velocity $\tilde{v}_{molec.}$ has little influence on the value of $\tilde{v}$ finally achieved, the initial vibrational motion does have a large effect due to the associated spread in values of r_0 .

Inside the target the Coulomb repulsion can be described [6] with an exponentially screened Coulomb potential with a screening distance $a = V/\omega_p$, where ω_p is the volume plasma frequency of the target. After emergence from the target there is no screening. For our example with the Au target ($\hbar\omega_p = 25.8$ eV [7]), $a = 3.2$ Å. If $r_0 = 0.8$ Å, the separation at exit from the 535-Å foil is 3.3 Å (if we neglect the effects of multiple scattering in the target). Thus, according to the simple model considered here, electronic screening of the Coulomb explosion should play only a small role.

This simple picture predicts that the proton should be detectable downstream from the foil at an angle $\psi \approx (v/V) \sin \theta$ with respect to the incident beam direction and with a laboratory energy which exceeds its initial value (800 keV) by an amount $M_p V v \cos \theta$, where M_p is the proton

rest mass. Thus, for an incident beam of randomly oriented (${}^4\text{HeH}$)⁺ molecules, the model predicts that downstream from the target, the protons should be confined within a cone of half-angle $\psi_{\text{CE}} \approx \frac{v}{V} = 0.34^\circ$ centered on the beam direction and that in the beam direction itself protons should be observable in two equally populated groups (corresponding to forward and to backward CM motion) whose energies differ by $2\Delta E_{\text{CE}} \approx 2M_p Vv = 19.2 \text{ keV}$. The spread in initial values of r_0 will, of course, lead to a spreading of ψ_{CE} and $2\Delta E_{\text{CE}}$ about the above values. For comparison, it is interesting to note that the normal energy loss of 800-keV protons in a 535-Å thick Au target is 7.5 keV and the angular half-width due to nuclear multiple scattering is 0.5° .

These considerations indicate that if one were to attempt to study experimentally the details of the Coulomb explosion of 4-MeV (${}^4\text{HeH}$)⁺ in a 535-Å thick amorphous Au foil, the angular distribution would be totally dominated by nuclear multiple scattering, and this would also lead to a smearing out of the energy spectra for the transmitted fragments. To avoid the adverse effects of nuclear multiple scattering on the angular distributions, the experiments can be performed under planar channeling conditions using a monocrystalline Au target. This arrangement, while permitting angular distributions to be measured cleanly, has the disadvantage that the energy spectra of transmitted particles are complicated by channeling effects [8]. In this paper we report results obtained for angular distributions measured in a plane parallel to the (111)-channeling planes of monocrystalline Au targets and for energy distributions measured

at 0° (i. e. in the beam direction) for amorphous C foil targets. We used tightly collimated beams of H^+ , H_2^+ , H_3^+ , $(^4\text{HeH})^+$, D^+ , D_2^+ , D_3^+ , and $(^3\text{HeD})^+$.

Angular distributions were measured under channeling conditions at Argonne National Laboratory. Figure 2 shows the experimental arrangement. The overall angular resolution was 0.08° , full width at half-maximum (fwhm). The Au target crystals were prepared by epitaxial growth on silver-mica substrates. The (111)-planar channeling dips obtained in backscattering for a beam of 4-MeV $(^4\text{HeH})^+$ incident on a target crystal of thickness (measured in the beam direction) $535 \text{ \AA}$, are shown in Fig. 3. For the transmission measurements, a movable detector assembly was used. It contained an aperture behind which lay a scattering foil. A semiconductor counter detected particles that had passed through the aperture and that had been scattered by this foil. (This scattering foil and detector system served to reduce the count-rates to manageable proportions and also to avoid counting more than one breakup fragment at a time.) Figure 4 shows a typical set of angular distributions for transmitted protons created from various bombarding molecular ions of 800-keV/nucleon. The angular width measured for incident H^+ is mostly determined by the experimental angular resolution combined with electronic multiple scattering in the target. Most of the angular distributions were roughly Gaussian in shape. But for protons from $(^4\text{HeH})^+$ (Fig. 4) and deuterons from $(^3\text{HeD})^+$ characteristic "hips" were seen. The corresponding ^4He and ^3He distributions were too narrow (compared with the resolution) to permit analysis. By magnetic deflection

of the transmitted particles it was demonstrated that the protons and deuterons were essentially all singly charged.

A somewhat similar arrangement was used at Lyon to measure energy spectra at 0° [see Fig. 5(a)]. The acceptance angle was 0.12° (fwhm) and the energy resolution was 5 keV (fwhm). The scattering foil in the detection system consisted of $10 \mu\text{g}/\text{cm}^2$ of C with 30 Å of Au evaporated on the front surface. The spectra exhibited two peaks as expected, but the low-energy peak was always more intense than the high-energy one and the separation was always somewhat less than $2\Delta E_{CE}$. These departures from the results expected from a simple screened-Coulomb explosion increased with the carbon-foil thickness [Figs. 5(b) and 5(c)].

Almost identical results were obtained in Rehovot with a different technique (Fig. 6). Here protons emerging from the target inside a cone of half-angle 0.09° centered on the beam direction were incident on thin secondary targets of either Al or CaF_2 . The γ -ray yield from the reaction $^{27}\text{Al}(p, \gamma)$ or $^{19}\text{F}(p, \alpha\gamma)$ was measured as the bombarding energy was varied over a small range so as to excite a narrow nuclear resonance. Both measurement techniques indicated that the detailed shapes of the energy spectra depended not only on the thickness but also on the composition of the target foil.

In Munich the channeling experiments were computer simulated by calculating, for several thousand randomly oriented incident molecules, the trajectories of the individual bare nuclei assumed created on entering the target. The initial internuclear separations r_0 were assumed

to have a Gaussian distribution. At first the particle motions were calculated with use only of a screened-Coulomb repulsion and a continuum channeling potential [9]. The influence of the target's stopping power on the trajectories is small and was neglected. Similarly, the small effects due to multiple scattering (almost entirely electronic) were not calculated microscopically but were taken into account (together with the angular resolution of the apparatus) by smearing the calculated angular distributions with Gaussians whose widths were those measured with monatomic beams. The widths of the angular distributions thus calculated were always too large and the shapes often differed markedly from the measured ones (see Fig. 7, dashed lines). We were unable to fit the experimental data by assuming larger initial internuclear separations or lower ionic charge states inside the solid.

The calculated angular distributions exhibit a central peak and two peaks located at $\pm\psi_{CE}$. Contributions to the central peak come mainly from incident molecules whose internuclear vectors lie close to the normal plane (i. e. , a plane that is perpendicular to the channeling planes of the crystal and that contains the beam direction). The relative height of this central peak is increased when one introduces into the calculations an additional force corresponding to a wake potential assumed to trail behind each particle as it traverses the solid. Good fits (examples are shown by the solid curves in Fig. 7) to a wide range of data were obtained with a wake potential of the form

$$\phi(s, \rho) = -(2Ze/a)\sin(s/a)K_0(\rho/a)S(s), \quad (3)$$

where Ze is the particle's charge, s and ρ are distances from the particle measured antiparallel to and perpendicular to the particle's velocity vector, respectively, K_0 denotes the zero-order modified Bessel function of the second kind, and $S(s)$ is the unit step function. The wakes of leading particles in a cluster influence the motions of trailing particles so that the internuclear vectors tend towards alignment with the normal plane (defined above). This causes a narrowing of the observed angular distributions.

The overall quality of the fits obtained for the angular distribution data may also be judged from Fig. 8 in which measured and calculated widths of the distributions are shown plotted as a function of the bombarding energy (MeV/nucleon). The fits are obtained with values of $\bar{r}_0$ and $2\Delta r_0$ (the fwhm of the assumed Gaussian distributions in r_0) that are entirely reasonable in terms of the discussion given above.

Equation (3) is identical with the (undamped) oscillatory part of the potential derived by Neufeld and Ritchie [10] in considering the passage of charged particles through a plasma. The wake potential results from electron-density oscillations generated by the projectiles. (It has recently been suggested [11] that charged particles could become trapped into bound states in such a wake potential.) The data on plasmon energies [7] and also the results presented here indicate that about eight electrons per Au atom contribute to the plasma oscillations. Channeling offers a unique opportunity for studying interactions with such a dense electron gas over usefully long interaction times without the disturbing influence of nuclear

multiple scattering. Since in our experiments, the clusters remain compact during traversal of the target, our measurements do not test the detailed form of the oscillatory part of Eq. (3). However, the fits are quite sensitive to the value chosen for the plasma frequency and also to the functional form chosen for the radial part of Eq. (3). Because of the compactness of the clusters, plasmon damping (even at the fast rate known to exist for Au) does not play a significant role here.

The unequal populations of the peaks observed in the energy spectra may be qualitatively understood by noting that trailing particles in a cluster experience a force tending to align their velocities with the beam direction, whereas leading particles experience no such force. Consider, for example, those protons arising from the breakup of $(^4\text{HeH})^+$ molecules whose initial internuclear vectors lie close to the beam direction. Protons emitted forwards at some small angle θ in the c.m. frame may miss a detector placed on the beam axis and having a restricted acceptance angle ($\sim \pm 0.06^\circ$ in our experiments). Protons emitted backwards (initially at an angle $\pi - \theta$ in the c.m. frame) would also miss the detector were it not for the force exerted by the wake of the α particle. This force reduces the angle between the proton velocity and the beam direction so that the proton may now strike the detector. A crude order-of-magnitude estimate of the extent to which the wake force can alter the laboratory angle for trailing particles emitted in the breakup of a diatomic cluster with nuclear charges Z_1e and Z_2e is given by

$$\Delta\psi = (Z_1 Z_2 e^2 d/a^2 E) \sin(\bar{s}/a) K_1(\bar{\rho}/a), \quad (4)$$

where d is the target thickness, E is the laboratory kinetic energy of the trailing particle, K_1 denotes the first-order modified Bessel function of the second kind, and $\bar{s}$ and $\bar{\rho}$ are average separations between the two nuclei while inside the solid, measured parallel to the perpendicular to the beam direction, respectively. For a case like the one depicted in Fig. 5(c) [protons from 2-MeV (^4HeH) $^+$ incident on 380-Å-thick carbon, $\hbar\omega_p = 22$ eV, dwell time in the target = 4.2×10^{-15} sec], Eq. (4) yields $\Delta\psi \approx 0.3^\circ$ if we take $\bar{s} = 1.7$ Å and $\bar{\rho} = 1.0$ Å (corresponding to a c.m. angle $\theta \approx 30^\circ$). By way of comparison, the laboratory angle expected for these protons in the absence of a wake force is $\psi \approx 0.25^\circ$. The longer the dwell time in the target, the longer the wake force can operate and the greater will be the disparity in the peak intensities in the 0° energy spectra. This is in accord with our experimental results. Thus, for 2-MeV H_2^+ incident on C foils of 2, 10, and $14 \mu\text{g}/\text{cm}^2$, we observe the high-energy proton peak to be less intense than the low-energy one by 7%, 15%, and 20%, respectively [12]. Similarly, as the bombarding energy is raised, the peaks become more nearly equally populated.

If the explanation offered here for the observed narrowing of the angular distributions and the asymmetry in population of the peaks in the 0° energy spectra is correct, then we have seen the first evidence for the

dynamic interaction between plasma oscillations in a solid and the motions of fast nuclear projectiles traversing the solid. To further elucidate the matter, we are continuing our experiments with a wide range of targets and projectiles and are extending our calculations to include triatomic molecular ions and also to cases in which nuclear multiple scattering is expected to play a significant role.

We gratefully acknowledge helpful discussions with Professor R. Sizmann and also the assistance of J. N. Worthington, R. Kirsch, and Y. Wolfson in performing the experiments.

References and Footnotes

*Work supported in part by the Energy Research and Development Administration.

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- [1] From cross sections derived, for example, by N. Bohr, Kgl. Dan. Vidensk. Selsk., Mat.-Fys. Medd. 18, No. 8 (1948), one can estimate that the mean free path for electron loss in the target is always less than $10 \text{ \AA}$ for the ions considered here.
- [2] A preliminary account of our measurements has recently been published: D. S. Gemmell, J. Remillieux, J.-C. Poizat, M. J. Gaillard, R. E. Holland, and Z. Vager, Phys. Rev. Letters 34, 1420 (1975).
- [3] W. L. Walters, D. G. Costello, J. G. Skofronick, D. W. Palmer, W. E. Kane, and R. G. Herb, Phys. Rev. 125, 2012 (1962).
- [4] J. Remillieux, Proceedings of the Fifth International Radiation Congress, Seattle, Washington, 1974 (in press).
- [5] See, for example, "Selected Constants Spectroscopic Data Relative to Diatomic Molecules" B. Rosen (Pergamon, New York 1970).
- [6] For a recent discussion, see W. Brandt, in Atomic Collisions in Solids, edited by S. Datz, B. R. Appleton, and C. D. Moak (Plenum, New York, 1975), p. 261.

- [7] M. Glicksman, in Solid State Physics, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1971), vol. 26, p. 338.
- [8] For a review, see D. S. Gemmell, Rev. Mod. Phys. 46, 129 (1974).
- [9] W. M. Gibson and J. Golovchenko, Phys. Rev. Lett. 28, 1301 (1972).
- [10] J. Neufeld and R. H. Ritchie, Phys. Rev. 98, 1632 (1955).
- [11] V. N. Neelavathi, R. H. Ritchie, and W. Brandt, Phys. Rev. Lett. 33, 302 (1974).
- [12] Similar measurements using $2\text{-}\mu\text{g}/\text{cm}^2$ C foils have been reported by J. Golovchenko and E. Laegsgaard, Phys. Rev. A 9, 1215 (1974).
Although these authors fit their energy spectra with a symmetric calculated curve, the data themselves display an asymmetry in agreement with that which we observe.

Figure Captions

- Fig. 1. Estimated distributions of vibrational excitations and internuclear distances for H_2^+ ions [4].
- Fig. 2. Schematic diagram of the experimental arrangement used in measuring angular distributions under channeling conditions.
- Fig. 3. Dips measured in the yields of backscattered α particles and protons for a 535-Å thick Au crystal bombarded by 4-MeV $(^4\text{HeH})^+$ ions under (111)-planar channeling conditions.
- Fig. 4. Angular distributions measured under (111)-channeling conditions for protons transmitted through a 535-Å thick Au crystal bombarded by a variety of molecular-ion beams at 0.8 MeV/nucleon.
- Fig. 5. (a) Experimental arrangement used at Lyon to measure 0° energy spectra
(b) and (c) Energy spectra for protons transmitted at 0° through 105- and 380-Å carbon foils bombarded by 2-MeV H_3^+ and $(^4\text{HeH})^+$.
- Fig. 6. Experimental arrangement used in Rehovot shown together with γ -ray yield curves measured for carbon foils of 100 Å and 500 Å bombarded by ~ 2 MeV H_2^+ beams.
- Fig. 7. Angular distributions for (a) protons from 4-MeV $(^4\text{HeH})^+$ and (b) deuterons from 1.6-MeV D_2^+ , measured under (111)-planar-channeling conditions with a 535-Å-thick Au crystal. The points are the experimental data. The curves are calculated with (solid lines) and without (dashed lines) the wake potential given in Eq. (3).

Fig. 8. Measured (solid dots) and calculated (crossed circles) values of the widths (fwhm) of the angular distributions measured for protons and deuterons transmitted through 535-Å-thick Au crystals bombarded under (111)-channeling conditions by a variety of incident molecular-ion beams. The indicated values of $\bar{r}_0$ and $2\Delta r_0$ refer to the mean and fwhm values for the assumed Gaussian distribution in internuclear separations for the incident molecular ions.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

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