nance line the usual Auger multiplet structure is generally shifted by the relaxation energy of about 5 eV for Xe and Kr to higher kinetic energies. We could also show that there is nearly the same probability for the two atoms to decay by the single Auger transition and by a Auger transition accompanied by a shake-up process leading to $5s^25p^47p$ and $4s^24p^46p$ excited states in Xe and Kr. The comparison between the measured spectra and the energies obtained from optical data[7] demonstrates the relevance of core rearrangement and other correlation effects in Auger transitions.

We want to thank H. W. Wolff for several helpful discussions.

[1]K. Siegbahn *et al.*, *ESCA Applied to Free Atoms* (North-Holland, Amsterdam, 1969).

[2]L. O. Werme, T. Bergmark, and K. Siegbahn, Phys. Scr. 6, 141 (1972).

[3]S. Othani, H. Nishimura, H. Suzuki, and K. Wakiya, Phys. Rev. Lett. 36, 863 (1976).

[4]V. Schmidt, N. Sandner, W. Mehlhorn, M. Y. Adam, and F. Wuilleumier, Phys. Rev. Lett. 38, 63 (1977).

[5]M. J. Van der Wiel, G. R. Wight, and R. R. Tol, J. Phys. B 9, L5 (1976).

[6]W. Eberhardt, G. Kalkoffen, and C. Kunz, to be published.

[7]C. E. Moore, *Atomic Energy Levels*, U. S. National Bureau of Standards Circular No. 467 (U. S. GPO, Washington, D. C., 1949, 1952, 1958).

[8]K. Codling and R. P. Madden, Phys. Rev. Lett. 12, 106 (1964).

[9]T. Åberg, in *Proceedings of the International Conference on Inner-Shell Ionization Phenomena and Future Applications, Atlanta, Georgia, 1972*, edited by R. W. Fink, J. T. Manson, I. M. Plams, and P. V. Rao, CONF-720404 (U. S. Atomic Energy Commission, Oak Ridge, Tenn., 1973), p. 1509.

[10]M. O. Krause and T. A. Carlson, Phys. Rev. 149, 52 (1966).

[11]M. Y. Adam, F. Wuilleumier, N. Sandner, V. Schmidt, and G. Wendin, to be published.

Observation of MeV Dissociative H_2^+ Ions Emerging from Very Thin Foils

M. J. Gaillard, J. C. Poizat, and J. Remillieux
Institut de Physique Nucléaire, and Institut National de Physique Nucléaire et de Physique des Particules, Université Lyon-1, 69621 Villeurbanne, France
(Received 17 March 1978)

We present angle and energy distributions of H atoms emerging from thin carbon foils bombarded with MeV H_2^+ and H_3^+ beams. If the target is thin enough, a projectile electron can participate in the formation of a neutral atom at emergence through dissociation of a vibrationally excited H_2^+ molecule. This process is not observable with thicker targets when the formation of a neutral requires the capture of a target electron. The results suggest that electrons can be bound to the projectile inside the solid.

Recent studies performed in our laboratory[1] have given new information about the neutralization of MeV protons at emergence from solids. It has been shown that hydrogen species containing electrons (H, H_2^+, H_3^+) produce more neutral atoms in the beam transmitted through very thin carbon foils (transit times $< 2\times10^{-15}$ s) than incident protons of the same velocity.

However, this evidence for what we shall call, perhaps abusively, a transmission of bound electrons through a solid target does not describe the electron-proton system when moving *inside* the solid. The existence of a bound state has been questioned by Brandt[2] and recently considered by Cross.[3] Moreover it was shown in Ref. 1 that the angular distributions of H atoms in the transmitted beam from incident H_2^+ projectiles are compatible with the repulsion of randomly oriented H^+H pairs along the dissociative $2p\sigma_u$ state of the H_2^+ molecule. This observation was, however, restricted to foil thicknesses for which the neutral atoms emerge after capture of a target electron.

In order to know more about the transmission effect in which the incident-projectile electrons participate in the formation of emergent H atoms, we have studied the angular distributions of H atoms transmitted through very thin carbon foils bombarded with H_2^+ and H_3^+ ions. This investigation was pursued further by measuring the ener-

gy distribution of the dissociation fragments transmitted at 0°.

As the neutral fraction in the transmitted beam is always much smaller than unity in this work, a neutral atom emerging from a foil may be considered as having one (two) bare proton(s) as partner(s). On the other hand, if the foil is thin enough, the separation between the cluster constituents increases only slightly before emergence and most of the repulsion, if any, takes place outside the target. Then the angle and energy distributions of the transmitted neutral atoms reveal the physical state of the cluster *outside* the solid. The differences observed in the neutral atoms "transmitted" or resulting from capture of a target electron suggest new ideas about the electronic state of the cluster *inside* the solid.

The H_2^+ and H_3^+ incident beams had an angular dispersion less than ± 0.2 mrad at the targets, which were 60–600-Å carbon foils. The dissociation fragments emerging from the foil at ± 0.2 mrad with respect to the incident beam direction were energy analyzed by a 90° magnetic spectrometer. The relative energy resolution of the measurement was 5×10^{-4}. Energy alalysis of the neutral atoms was performed by inserting a permanent magnet which removes the charged components and a 3-μg/cm^2 carbon stripper.

Angular distributions of H atoms emerging from carbon targets of thicknesses 1.8, 3.5, and 7.5 μg/cm^2 bombarded with 2.3-MeV H_2^+ ions are shown in Fig. 1. The count number for each experimental point corresponds to the same number of incident projectiles. Then, because of the cylindrical symmetry, the volume generated by rotation of each distribution around the beam axis ($\theta = 0$) is a measure of the neutral fraction $\Phi_0^{H_2^+}$. The ratio $\Phi_0^{H_2^+}/\Phi_0^{H^+}$, where $\Phi_0^{H^+}$ is the neutral fraction measured with an incident proton beam of the same velocity, is also shown for each target on Fig. 1. We notice the change in the shape of the distribution when the foil thickness decreases. The presence of a peak at 0° seems to be correlated to the overproduction of neutral atoms which has been previously observed.[1] This peak corresponds to H^+H pairs which liberate nearly no transverse energy. These pairs originate either from H_2^+ molecules in a repulsive state and preferentially aligned along the beam direction or from randomly oriented systems which dissociate without repulsion. A similar observation was made with incident H_3^+ ions where a central plateau (instead of a peak) dominates the broad distribution of neutrals.

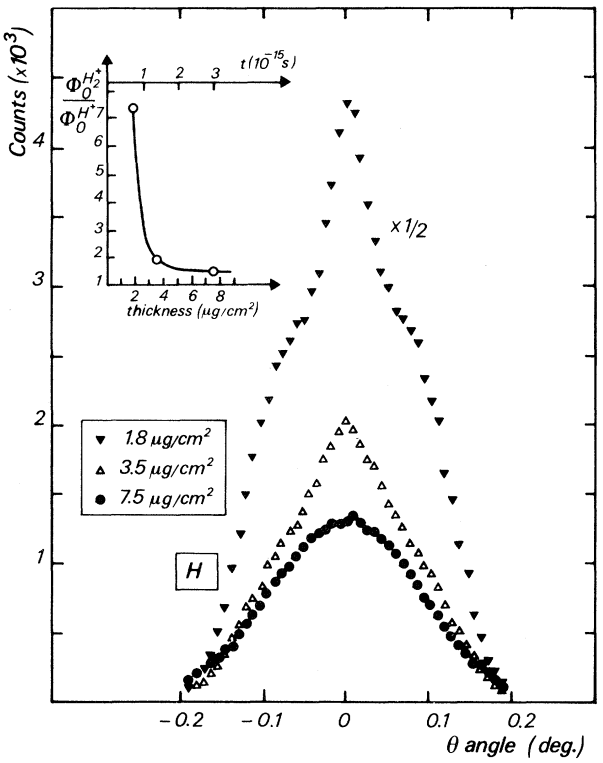

FIG. 1. The angular distribution of H atoms in the beam transmitted through carbon foils (1.8, 3.5, 7.5 μg/cm^2) for incident 2.3-MeV H_2^+ ions. Inset, the variation of the neutral fraction vs foil thickness and projectile transit time in the foil.

We measured the energy distribution of the neutral atoms emerging at 0°. We show in Figs. 2 and 3 the energy spectra of protons and H atoms emerging at ∓ 0.2 mrad from carbon foils of various thicknesses bombarded with 2.4-MeV H_2^+ and 2.2-MeV H_3^+ ions, respectively. The count numbers in the various spectra are not normalized and the zero energy has been set approximately at the center of symmetry of each spectrum and corresponds to the energy of fragments which would have received no repulsion energy. In part (a) of both figures the energy distribution of protons is shown in order to allow a comparison with the H^+H^+ Coulomb explosion.

The energy distribution of H atoms from H_2^+ projectiles exhibits a sharp central peak and two lateral peaks [Figs. 2(b) and 2(c)], a structure which is not observable with the thickest foil [Fig. 2(d)]. The separation between the two lateral peaks measured for the thinnest target is 7.3 keV, in very good agreement with the separation

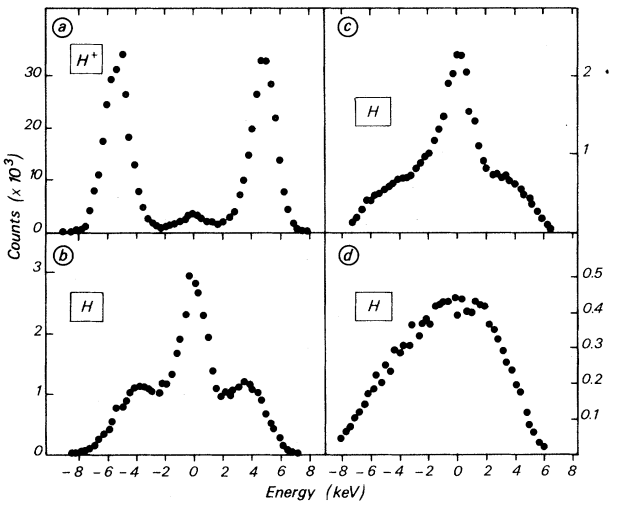

FIG. 2. The energy distribution of (a) protons and (b)–(d) neutrals emerging at 0° when 2.4-MeV H_2^+ are incident on (a), (b) 1-, (c) 3.3-, and (d) 9.5-μg/cm^2 carbon foils.

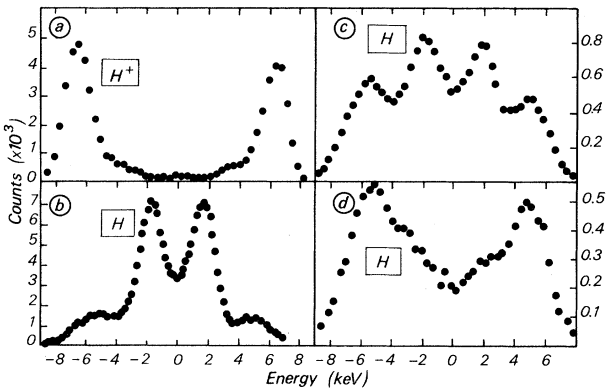

FIG. 3. As in Fig. 2, for 2.2-MeV H_3^+ incident on (a), (b) 1-, (c) 1.8-, and (d) 3.8-μg/cm^2 carbon foils.

expected from the dissociation of H_2^+ molecules along the $2p\sigma_u$ state, and confirms the role played by this repulsive state in the production of neutrals.

After ruling out the possibility for the central peak to be due to edge effects around pinholes in the foil, we note that the peak is wider than the experimental energy resolution and therefore corresponds to neutrals which have experienced a reduced repulsion; either their partner has been scattered by a target atom (process A), or they have separated gently from their proton partner downstream of the target (process B). Processes A and B are also responsible for the small central peak of Fig. 2(a). However, their contributions to the central peaks of Figs. 2(a) and 2(b) are different, since process B produces one neutral for one proton, whereas A produces only Φ_0 neutrals for $(1-\Phi_0)$ protons. From an estimated value of Φ_0 through the 1-μg/cm^2 target, and assuming that the probabilities of processes A and B are independent of the incident molecule orientation, one calculates from spectra [2(a)] and [2(b)] that ~1% of transmitted protons and ~15% of transmitted neutrals have experienced process A or B and that process A contributes ~75% to the central proton peak but only ~5% to the central peak of neutrals.

We interpret process B as resulting from the formation of a H_2^+ molecule in the electronic ground state $1s\sigma_g$, but in a state of vibration-rotation located in the continuum above the dissociation level. The corresponding molecular-ion–solid interaction can be described as a vibrational dissociation of H_2^+, a process which has been observed for a long time with gas targets.[4] We conclude that, in the electron-transmission regime, the neutrals are produced via two channels: autodissociation of the $2p\sigma_u$ state (~85%) and vibrational dissociation (~15%). Unfortunately, multiple-scattering effects, i.e., the increasing contribution of process A with increasing thicknesses to a widening central peak (because the repulsion between partners before scattering lasts longer in the average), and the smallness of the contribution of process B to the production of neutrals, even for a thin target, make it difficult to study the behavior for thicker targets; Fig. 1 suggests a correlation between the observation of vibrational dissociation and the electron-transmission regime.

During the course of this work, Laubert and Chen[5] have reported on the observation of a central peak in the energy distribution of protons emerging at 0° from thin foils bombarded with 100–300-keV H_2^+ ions. They interpret this peak as being due to protons which did not experience repulsion inside the target. We remark that these protons did not experience repulsion even outside the target. Their projectile dwell times are longer than ours and should not allow observation of the electron-transmission regime considered in our work. We think that process A could become dominant at lower energies, although Fig. 3 of Ref. 5 suggests that a small fraction of unrepelled protons could originate from process B.

The existence of two dissociation channels, evidenced with H_2^+ beams, has been confirmed with H_3^+ beams. In Fig. 3 we show the energy spectra of H atoms emerging at 0° from targets of thicknesses (b) 1, (c) 1.8, and (d) 3.8 $\mu g/cm^2$ for incident 2.2-MeV H_3^+ ions. For the thickest target (d), the H atoms are due to target-electron capture and one observes mainly two peaks, separated by 10 keV, due to the repulsive dissociation of the unstable H_3^{++} molecule into H-2H$^+$ fragments.

With thinner targets one observes two new peaks: They appear in Fig. 3(c) and dominate in Fig. 3(b). Their energy separation reveals a reduced repulsion of the H atom by its two H$^+$ partners. We believe that these new peaks have the same origin as the central peak observed with incident H_2^+ ions, and we interpret them as resulting from a two-step dissociation of the triangular[6] triproton cluster, as follows: (i) The emergent cluster behaves like a dicluster consisting of a bare proton plus a H_2^+ molecule in the continuum of the $1s\sigma_g$ state. A few 10^{-15} s are needed by this repulsive $H^+H_2^+$ system to liberate its kinetic energy. (ii) Simultaneously the H_2^+ molecule undergoes a vibrational dissociation which takes much more time and can be then considered as taking place after completion of the $H^+H_2^+$ repulsion. This last process transfers no additional energy to the resulting H$^+$H pair.

The $H^+H_2^+$ repulsion was directly measured from the energy distribution of the *bound* H_2^+ molecules[7] emerging at 0° from the 1-$\mu g/cm^2$ target with a 2.2-MeV H_3^+ beam: The spectrum exhibits two peaks separated by 8.7 keV. The energy is to be compared to twice the energy separation between the peaks of Fig. 3(b), 2×3.4 keV. This value is 20% too low, because at emergence the mean proton separation is probably larger in a molecule vibrationally excited to the continuum than in a bound molecule, and furthermore increases slightly during the $H^+H_2^+$ repulsion. We consider the agreement good enough to support the idea of a neutral atom separating from its two partners through the described double process.

Here the fraction of neutral atoms produced through vibrational dissociation reaches 45% of the total number of neutral atoms emerging from the thinnest foil [Fig. 3(b)]. This value, contrary to the H_2^+ case, is high enough to enable us to study the evolution of the phenomenon with increasing target thicknesses. We observe a decrease of the fraction of neutrals due to vibrationally excited H_2^+, which appears to be correlated with the decay of the electron-transmission regime (the maximum thickness for electron transmission, given in Ref. 1, is ~3 $\mu g/cm^2$ for 2.2-MeV H_3^+ ions). We believe that this correlation holds in the case of H_2^+ projectiles, where we saw it was not easily observable.

The study of angle and energy distributions of neutral atoms emerging from very thin carbon foils bombarded with H_2^+ and H_3^+ projectiles[8] can be summarized as follows: The projectile electons still accompanying the cluster at emergence may interact with it and form a H atom in a specific way which is the formation and the dissociation of an intermediate H_2^+ molecule vibrationally excited to the continuum. This observation raises questions about the state of projectile electrons *inside* the solid, since we showed that transmitted electrons may occupy a molecular ground state at emergence as they did in the incident projectile. This work does not prove, but strongly suggests, that these electrons are in a bound state inside the foil.

[1]M. J. Gaillard, J. C. Poizat, A. Ratkowski, J. Remillieus, and M. Auzas, Phys. Rev. A 16, 2323 (1977).

[2]W. Brandt, *Atomic Collisions in Solids* (Plenum, New York, 1975), Vol. 1, p. 261.

[3]M. C. Cross, Phys. Rev. B 15, 602 (1977).

[4]See, for example, R. Caudano and J. M. Delfosse, J. Phys. B 1, 813 (1968).

[5]R. Laubert and F. Chen, Phys. Rev. Lett. 40, 174 (1978).

[6]M. J. Gaillard, D. S. Gemmell, G. Goldring, I. Levine, W. J. Pietsch, J. C. Poizat, A. Ratkowski, J. Remillieux, Z. Vager, and B. J. Zabranski, to be published.

[7]M. J. Gaillard, J. C. Poizat, A. Ratkowski, and J. Remillieux, Nucl. Instrum. Methods 132, 69 (1976).

[8]A similar observation has been made by D. S. Gemmell in the case of HeH$^+$ projectiles (private communication).