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THE TRANSMISSION OF FAST MOLECULAR IONS THROUGH THIN FOILS[§]

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We present new results on the transmission of fast molecular ions through thin foils and propose a mechanism for the transmission process. The main feature of the postulated mechanism is that a finite fraction of the incident molecular beam does not undergo a strong Coulomb explosion while traversing the foil. Because the emerging fragments are at large internuclear separations, there is an enhanced probability for the formation of bound, long-range, excited electronic states following electron capture at the rear surface of the target.

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There has been a great deal of interest aroused by several effects that are observed when beams of swift molecular ions bombard thin solid targets^{1,2)}. For light projectiles (H_2^+ , HeH^+ , He_2^+ , etc.) incident at MeV energies upon a foil, the binding electrons are, with a very high probability, stripped off within the first few Angstroms of penetration into the solid target. This is a consequence of the large cross section ($\sim 10^{-16} \text{ cm}^2$) for electron loss³⁾ and of 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) constituent nuclei of the projectile fly apart by virtue of their mutual Coulomb repulsion. The characteristic time for this explosion ($\sim 10^{-15}$ sec) is typically of the same order as the dwell time of the projectile in the foil, for foils about $100 \text{ \AA}$ thick. As a result, much of the Coulomb explosion takes place inside the foil and it then runs to completion in the vacuum downstream after the fragments emerge from the target. Inside the target, the individual fragment trajectories are also influenced by multiple scattering and by effects due to the electron polarization "wakes" induced behind each of the fragments traversing the foil. These wake effects have been shown⁴⁾ to give rise to easily observable modifications to the pattern of trajectories measured downstream from the target. These effects are now reasonably well understood in terms of a theoretical model describing the interaction of these explosion fragments with the wake⁵⁾.

Although the Coulomb explosion is the predominant mode of dissociation in foils (even for emerging neutral fragments), a small fraction ($\lesssim 10^{-4}$) of the incident beam has been found to exit the foil without undergoing a strong Coulomb repulsion. These particles

manifest themselves in two distinct phenomena. a) In several cases, an extremely small fraction of particles emerge from the foil as the incident molecular species, though having suffered an energy loss in the target^{1,6,7)}. This process, first studied by Poizat and Remillieux in 1971⁸⁾, is referred to as "transmission" of the incident beam.

b) At the same level of intensity as the transmitted ions, unexploded, though dissociated, molecular fragments^{1,2,9)} are seen as central peaks in the velocity spectra of the dissociation products¹⁰⁾.

No complete explanation of either of these processes has been published. In this Letter we report a series of experiments conducted to elucidate the nature of the transmission and central peak phenomena using a variety of combinations of targets and incident molecular ion beams. Preliminary results of this study were reported earlier¹⁾. Further, we propose a model which relates the transmission to the existence of non-exploded clusters (central peaks).

The basic apparatus used in these experiments has been described in detail in refs. 1,2 and 10. This setup allowed us to measure distributions in angle and energy of particles emerging from the target with an overall angular resolution of 0.15 milliradians and relative energy resolution of 6×10^{-4} . Transmission yields were determined by integrating the measured angular and energy distributions. By comparing these yields to those measured with protons of the same velocity (assumed to have 100% transmission probability) the transmission probabilities, T , were derived. Because of the high resolution of this system, the transmitted molecules were energetically clearly separated from that portion of the beam which passed through the target "pinholes" occurring normally in all but the thickest targets. In the measured energy spectra, the sharp pinhole peaks, which

can be more intense than the transmitted peak, served as an ideal calibration in measuring the energy loss and straggling of the transmitted molecules.

In the investigations of energy and angular distributions of the exploded fragments of molecular ion beams traversing thin foils it was found that a small fraction ($\sim 10^{-4}$) of molecules does not undergo a complete Coulomb explosion. This can be seen in the central peak of zero-angle energy distributions, as reported earlier^{1,2)} and as observed for lower energies and incident H_2^+ molecules by Laubert and Chen⁹⁾. Four examples are given in fig. 1. We have not yet made systematic studies of this central peak, e.g. the dependence of its intensity on particle energy, thickness or atomic number of the target. Such studies are difficult in our energy regime due to limitations of counting statistics as can be seen from fig. 1. However, the mere existence of a central peak in these energy distributions proves that there is a finite probability that a molecular cluster can traverse a foil without its constituents being violently separated due to strong Coulomb repulsion. We think that this result is an important clue to the mechanism of the transmission of molecules through foils. In fact, for all cases where we observe central peaks in the energy distribution of one of the exploded fragments, we find that the bound molecule can also be observed behind the foil with relatively high probability.

Figure 2 shows results of our transmission measurements for various types of molecules, energies and targets. The transmission is strongly dependent on all of these parameters. Not shown are negative results i.e. examples where we did not observe transmission even with the thinnest target foils. Such cases

include 4-MeV O_2^+ ions and 2-MeV CO^+ ions where we found upper limits of $T \lesssim 10^{-11}$

for the transmission through $\sim 100 \text{ \AA}$ carbon foils. Again it is noteworthy that for these ions we also did not observe a central peak in the energy distributions of the exploded fragments.

In another series of measurements we used "sandwich" targets consisting of a $2 \text{ \mu g/cm}^2$ Au layer on a $2 \text{ \mu g/cm}^2$ carbon foil. Here we observed the interesting effect that the existence of the central peak in the energy distribution of protons from HeH^+ molecules depends on the orientation of the bi-layer targets with respect to the incoming molecules. While in the case where the molecules enter the target through the Au layer a pronounced central peak is obtained (about the same magnitude as the one in fig. 1a, H_2^+), no central peak is resolved within the same counting statistics when the molecules first traverse the C foil. The fact that, for the latter orientation, the transmission is smaller by $\sim 30\%$ clearly connects these two phenomena, at least qualitatively.

Further, we want to point out that the data for D_3^+ are the first reported observation of the transmission of a triatomic molecule (we chose D_3^+ rather than H_3^+ to avoid possible contamination by the diatomic HD^+ molecule). We were also able to measure the transmission for HeH^+ and H_2^+ through $\sim 200 \text{ \AA}$ Al foils and $\sim 400 \text{ \AA}$ Al_2O_3 foils, finding that the transmission through these foils is about 1 order of magnitude larger than for carbon foils of the same thickness (see connecting vertical lines in fig. 2). It is, however, not a general trend that the transmission increases with increasing atomic number of the target since we found an upper limit of $\lesssim 10^{-8}$ for the transmission of 2-MeV H_2^+ through a composite $100 \text{ \mu g/cm}^2$ Au— $20 \text{ \mu g/cm}^2$ Si foil

($\sim 1400 \text{ \AA}$ thick) whereas the transmission through a carbon foil of the same thickness is $\sim 10^{-7}$.

From our experimental observation of the connection between unexploded clusters and emerging molecules, which is most clearly demonstrated with the double-layer targets, we are led to the following dual process of transmission of molecules through foils: Upon entering the foil the molecule rapidly loses its binding electrons. An estimate for the mean free path for this process can be obtained from our data using the fact that the transmission should go to unity for target thicknesses approaching zero. From the transmission measured with the thinnest carbon targets and, e.g., incident 2-MeV HeH^+ , $T \sim 10^{-6}$ at $\sim 160 \text{ \AA}$, we obtain an upper limit for the mean free path of $\lambda_D \lesssim \frac{-160}{\ln T} \lesssim 10 \text{ \AA}$. This is smaller than the mean free path for atomic electron loss λ_L , which is measured to be $\lambda_L \sim 20-40 \text{ \AA}$ (e.g. for MeV hydrogen atoms in carbon)¹¹⁾. For very thin targets ($< 100 \text{ \AA}$) there can therefore be sizeable contributions of undissociated molecules and neutral (but dissociated) fragments exiting the foil. These contributions ($\sim 10^{-5}$ at $100 \text{ \AA}$) fall off much too rapidly with path length however to account for the data with thicker targets. The observed falloff is clearly non-exponential and much slower than either λ_D or λ_L imply.

In order to account for the large transmission yields with the thick targets, one must consider the motions of the exploding fragments. In particular, we concentrate on those incident molecules having large incident internuclear separations r_0 ¹⁰⁾. These projectiles, after losing their binding electrons, move apart only slightly and very slowly during passage through the foil. This is a consequence of inertia and electronic screening of an already weakened Coulomb repulsion.

Consider, for example, the case of 2-MeV H_2^+ incident on a $200 \text{ \AA}$ carbon foil. Assuming a simple Coulomb explosion (no screening, no multiple scattering, etc.) while in the foil, the amount of potential energy converted

to kinetic energy during this transit time is 2.97 eV and 0.05 eV for r_0 values of 1 Å and 3 Å, respectively. During the 1.4 femtosecond dwell time in the target, the 1 Å separation would increase by 26% to 1.26 Å while the 3 Å initial separation only increases by 1.1% to 3.03 Å. These considerations lead to the inevitable conclusion that those molecules with initially large internuclear separations produce exiting clusters of nuclei whose spatial and velocity relations are not very dissimilar from those of the incident molecule.

At, or very near to, the exit of the foil, the cluster may capture one or more binding electrons. Thus, as the cluster emerges into the vacuum, a molecular ion may be formed (possibly in a vibrationally-excited and/or electronically-excited state).

This three-step process will produce both transmitted molecular ions and central peak dissociation fragments. Both can arise with foil targets when there is a large internuclear separation in the incident projectile. In both cases, there is a very weak Coulomb explosion (or more appropriately whimper), followed by electron capture at the foil exit. If the capture leads to a bound state of the molecule, transmission results. In the case of capture to an unbound state, the molecular ion so formed dissociates into a neutral and a charged fragment with very small relative kinetic energy (because of the already large internuclear separation). These dissociation fragments therefore produce a central peak in the measured energy spectrum at 0°, similar to that seen in dissociation in gas^{1,10}).

This mechanism for molecular-ion transmission also explains our previous observation¹⁾, first suggested by Gaillard et al. for H_2^+

transmission, that the velocity and target-thickness dependence of the transmission of HeH^+ can be factored in the form $T = P(\tau) \cdot \phi(v)$ where $P(\tau)$ depends only on the projectile's dwell time inside the target and $\phi(v)$ depends only on the projectile velocity. In a plot of T/ϕ versus τ we then obtain a single curve for all energies and target thicknesses (see ref. 1). The function $\phi(v)$, which turns out to vary as v^{-9} , gives then the purely velocity-dependent probability for electron capture, while $P(\tau)$ describes the probability for molecular recombination of exploded fragments. A similar scaling property was found to hold for H_2^+ 7).

It must be stressed that it is essential to have a weak Coulomb explosion as a main feature of the transmission mechanism. For example, if we assume a Coulomb explosion starting from the equilibrium separation of $0.8 \text{ \AA}$, the internuclear separation for the case of 2-MeV HeH^+ molecule after having traversed $540 \text{ \AA}$ carbon is $\sim 5 \text{ \AA}$, the c.m. energy is $\sim 95\%$ of the total released Coulomb energy, i.e. $\sim 34 \text{ eV}$. The potential well of the HeH^+ bound electronic ground state has a depth of $\sim 2 \text{ eV}$ and extends significantly to $\sim 2 \text{ \AA}$ internuclear distance. Only if the fragments are close in energy and distance to those characteristic of some bound electronic state of the molecule is there a significant recombination probability. We have performed a quantum-mechanical calculation, based on the sudden approximation, for 2 MeV HeH^+ . Assuming all ions are incident in the ground vibrational state, and electron capture at exit takes place only to the ground electronic state, the calculated transmission falls too rapidly with dwell time to account for the data beyond $\sim 10^{-15}$ second dwell time. For the above example, the probability of capturing electrons and making a transition to the electronic ground state is estimated to be

$<10^{-12}$ in contrast to the measured transmission of $T \sim 10^{-8}$. Clearly one must take account of large internuclear separations in both entering and exiting particles.

The data to support the above model of transmission and central peaks come from a variety of sources. Our experimental method allows not only the determination of transmission yields but also energy loss, energy straggling, and multiple scattering of the transmitted ions. There have also been stopping power determinations published in the literature ^{1,12}). The results of these experiments indicate that the stopping power of transmitted molecules is a) independent of thickness and b) is, at low energies, representative of independent particles. Studies of the stopping powers of explosion fragments however reveal a marked dependence on internuclear separation ¹³) increasing above the independent particle value as the separation decreases. One concludes from the above that transmitted molecules are incident on the target with large internuclear separations [because of (b)] and the separation remains fairly constant while in the foil [because of (a)]. As the velocity of the incident beam is increased, and the dwell time therefore decreases, molecules with smaller internuclear separations can contribute to the transmitted fraction. Correspondingly, the observed stopping power approaches that of united nuclei ¹).

The angular width of the multiple scattering of the transmitted HeH^+ ions increases proportional to the square root of the target thickness ¹), indicative of a fixed internuclear separation while the molecule is in the target.

To test our model, we have conducted an experiment in which 1 MeV H_2^+ ions transmitted through a 330 Å carbon foil are separated from pinhole and deuterium contaminants with an analyzing magnet and then used in a conventional Coulomb explosion measurement. By breaking these transmitted molecules in a second 75 Å carbon target and measuring the resulting angular and energy distributions of the exiting protons, we deduce the Coulomb energies and hence internuclear separations contained in the transmitted beam.

Figure 3 shows such energy spectra for proton fragments from ions that are transmitted through the first target and for fragments from those ions which pass through pinholes of the same, first target. The energy separation seen for pinhole ions is found to be 6.7 keV while transmitted ions result in proton energy shifts of 5.3 keV. A simple estimate based on these figures shows that whereas the mean internuclear separation in the pinhole ions is ~ 1.1 Å, the transmitted molecules are 60% larger (1.8 Å). A detailed unfolding of the distributions of internuclear separations has been performed using the techniques described in ref. 10. Those results (see fig. 4) clearly demonstrate the importance of large internuclear spacings in the transmitted beam.

Whether or not this large mean radius is a result of capture to electronically excited states of H_2^+ ($3\sigma_g, 2p\pi_u$ for example), or simply vibrationally excited levels of the ground electronic state, is difficult to resolve from the present data. Both probably contribute, though the more long-range electronic states would be expected to become more important at longer dwell times, where lower counting rates prohibit similar experiments. Furthermore, during the 1 μsec flight time between foils, most of the $2p\pi_u$ population would decay to the ground $1\sigma_g$ (populating primarily high vibrational levels). The $3\sigma_g$ population would decay to the dissociative $2p\sigma_u$ state and thus contribute to the observed central peaks. The net result would be that the molecules

observed after this long flight time would in any case probably be in high vibrational states of the ground electronic state. This interpretation is in agreement with the measured distribution of internuclear spacings shown in fig. 4.

One further piece of evidence from this experiment is the lack of any wake alignment effects in the explosion of transmitted ions whereas the explosion fragments from pinhole molecules demonstrate considerable alignment. This would suggest that "wakeriding" is not occurring to any appreciable extent (i.e., there is no preferred orientation for transmitted molecules).

The main conclusion reached in this work is that it is not necessary to invoke wake-riding electrons or other exotic processes^{14,9,12)} to account for the observed transmission and central peak phenomena. In fact, such experiments are relatively insensitive to the potential experienced by moving particles in the solid. Rather, such experiments are a sensitive probe of the initial state distribution of the incident molecular-ions and of the character of the final states of the same ion. In particular, it is possible that among the final electronic states formed at the foil exit, there will be a preference for long-range states of the molecule. Such states, though well known theoretically for many years, have never been directly observed experimentally¹⁵⁾. The process of Coulomb explosion and subsequent molecular reformation is an excellent method to study such molecules.

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Figure Captions

- Fig. 1. Some representative energy spectra of Coulomb explosion fragments showing central peaks. a) 3-MeV $\text{H}_2^+ \rightarrow \text{H}^+$; b) 3.5-MeV ${}^4\text{HeH}^+ \rightarrow \text{H}^+$; c) 3-MeV ${}^3\text{He}_2^+ \rightarrow {}^3\text{He}^{++}$, and d) 3-MeV ${}^3\text{He}_2^+ \rightarrow {}^3\text{He}^+$. Note the significant enhancement at the central peak in c) over that in d).
- Fig. 2. Transmitted molecular ion yields as a function of target thickness.
- Fig. 3. Energy spectra of protons from 1 MeV $\text{H}_2^+ \rightarrow \text{H}^+$. The incident H_2^+ were first prepared by passage through a 330 Å carbon foil.
- Fig. 4. Distribution of internuclear separations for H_2^+ transmitted through thin carbon foil. The curves are both normalized to unity. The curves are fits of the form $D = \exp[\sum_{n=0}^6 a_n r^n]$, used to normalize the distributions.

Fig. 1

Fig. 2