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Effect of the neutral charge fraction in the Coulomb explosion of H_2^+ ions through aluminum foils

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Abstract

The Coulomb explosion of the proton fragments dissociated from H_2^+ molecules moving through thin aluminum foils has been studied by means of their energy spectra, measured in the forward direction, and by computer simulations. The covered energy range goes from 25 to 100 keV/u. Estimations of the neutral charge fraction of the fragments inside the foil have been obtained by comparison of the experimental energy spectra with the computer simulations.

1. Introduction

The study of the interaction of molecular ions and clusters with matter has been an important subject of research in the last 25 years. Since the pioneering work of Brandt and coworkers [1] where the vicinage effects in the energy loss of the dissociated molecular ions through thin foils were first analyzed, a lot of work has been done in this and related topics, both theoretically and experimentally (see Ref. [2] for a complete list of references). In particular, the study of the interaction of H_2^+ molecules with matter has proven to be specially useful because the simplicity of the molecule allows to analyze the vicinage effects and test the theories [1–13]. In a further pioneering work Laubert and Chen [14] obtained information on the charge state fractions of hydrogen ions moving inside a solid from the analysis of the energy spectra produced by H_2^+ fragments. Most of the previous studies analyze the high velocity regime ($v > v_0$) where the theories can be applied successfully (here $v_0 = 1$ a.u. is the Bohr velocity). Nevertheless few papers were devoted to the study of the intermediate velocity regime ($v \sim v_0$), which is specially challenging, in particular because there is a significant change in the screening conditions, as well as a strong variation of the projectile charge states [15].

In this paper we present energy-distribution measurements of the proton fragments dissociated

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from the H_2^+ molecule in thin aluminum foils and emerging in the forward direction, in the intermediate velocity regime (24–102 keV/u). In addition, we perform a computer simulation study containing the basic interactions of the ions within the solid (Coulomb repulsion between the ionized fragments, electronic stopping forces, wake forces and elastic scattering with the atomic cores) and charge exchange process. By changing the value of the charge state fractions used in these simulations, and looking for the best fit with the experimental spectra, we have been able to obtain values of the charge fractions for various ion energies.

In Section 2 we describe the experimental procedure and in Section 3 we introduce the computational method used in the simulation. The main results are shown in Section 4 and the conclusions are presented in Section 5.

2. Experiment

The H_2^+ and H^+ projectiles were generated by electrostatic acceleration to 25–100 keV/u and magnetic mass selection of ions produced in a RF source. A target chamber with foils removable in vacuo from the beam path followed a collimation by two 1-mm diameter diaphragms separated by 109.5 cm. The energy spectra were determined by means of an electrostatic energy analyzer placed in the forward direction with an angular acceptance of 0.05°. The overall energy resolution, including the effect of the energy spread of the beam was 0.3%. The projectiles were detected by a discrete dynode electron multiplier followed by standard pulse counting electronics. The targets were self supporting 2 mm diameter, 19 nm thick aluminum foils, prepared under clean vacuum conditions [16]. The target thickness was determined by fitting the proton energy loss measured at 100 keV with the Andersen and Ziegler stopping power tables [17]. The thickness homogeneity was determined following a beam technique [18] resulting in a mean square deviation of $\leqslant 12\%$. Foil thickening by beam bombardment [19] was held with negligible limits using low beam doses and was checked

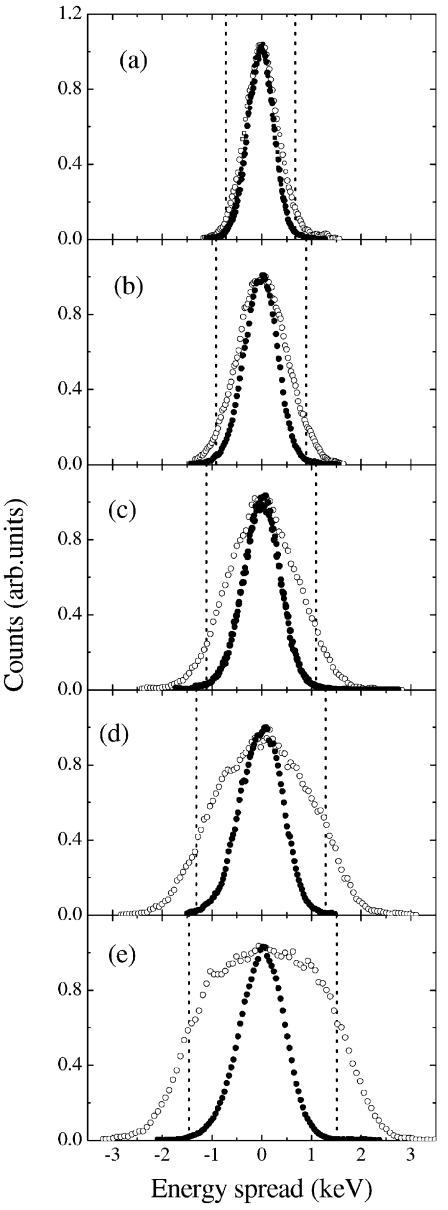

Fig. 1. Experimental energy distributions of protons dissociated from H_2^+ traversing a 19 nm thick aluminum foil, and exiting in the forward direction (empty circles). The incident projectile energies, E_0, are (a) 23.8 keV/u, (b) 39.2 keV/u, (c) 56.7 keV/u, (d) 78.8 keV/u and (e) 101.8 keV/u. The center of each spectrum corresponds to the following energies: (a) 21.8 keV, (b) 36.8 keV, (c) 54.1 keV, (d) 76.1 keV and (e) 99.2 keV. The experimental energy distributions for proton beams incident with the same energies per nucleon are also presented (full circles). The vertical dashed lines represent the energies after a pure Coulomb explosion in vacuum for a H_2^+ molecule.

by comparison of the energy loss before and after the measurements series.

Fig. 1 shows energy-distribution measurements for charged dissociation fragments emerging from aluminum foils in the forward direction arising from H_2^+ projectiles, for different incident energies, centered at the mean energy of the spectra. Additionally we also show the measured energy distributions corresponding to H^+ projectiles with the same incident energies. Comparison of the different energy spectra, empty circles for H_2^+ or full circles for H^+ projectiles, shows changes in the molecular effects with the beam energy.

For the sake of comparison, we indicate in Fig. 1 with vertical dashed lines the energy spread corresponding to a hypothetical pure Coulomb explosion in vacuum for a H_2^+ molecule with an initial internuclear separation of 1.29 Å [1]; in this way, we can compare the widths of the fragment energy spectra with those of the pure Coulomb case. The figure indicates a transition from an almost complete screening at the lowest projectile energy to a nearly unscreened Coulomb explosion at the highest energy value.

3. Computer simulation

When a H_2^+ ion penetrates into a solid it loses its binding electron in the first atomic layers [4] and then the fragments move as two correlated projectiles. These projectiles may capture or lose electrons along their path and so they can have different charge states while travelling inside the foil, i.e. H^+ and H^0, as it has been experimentally reported in this energy range [15,20,21].

The trajectories of the fragments dissociated from the H_2^+ molecular ion inside the foil were calculated by means of a computer code which has been described previously [12,13]. In this code we simulate the trajectories of each dissociated fragment by integrating the Newton equations of motion with a finite difference algorithm; in addition, we use a Monte Carlo code to incorporate the effects of the elastic scattering with the target nuclei [22]. The forces acting on each fragment are: the electronic self stopping force, the reciprocal wake forces due to the electronic excitations induced by the fragments, and the Coulomb repulsion (depending on the charge states). The statistical fluctuations of the projectile energy loss is taken into account through the energy loss straggling, calculated using the dielectric formalism, where the dielectric properties of the aluminum target were modelled by a linear combination of Mermin type energy-loss functions [23] to describe the outer electron excitations [24] together with a generalized oscillator strength for the K-inner-shell electron excitations [25]. The wake forces between the fragments of the H_2^+ molecule (both for protons and neutrals) were also calculated using the dielectric formalism [24] [1].

The processes of charge exchange, in which electrons are captured or lost by the fragments, were incorporated in the code choosing randomly at each time-step of the finite difference algorithm a state of charge (neutral or proton) for the fragment, according to the equilibrium neutral charge fraction Φ_0, which was a parameter of our simulation. At each time-step we have used the values of the wake forces, energy loss straggling and Coulomb repulsion corresponding to the chosen charge states.

After exiting the foil, the fragments experience a pure Coulomb repulsion in the vacuum if both projectiles are positively charged, or otherwise they move freely. Consideration of the Coulomb repulsion outside the foil is important because the Coulomb explosion occurring inside the foil is not complete, especially at the highest projectile energies. To take into account the angular acceptance of this experiment, we registered only those protons emerging within an angular cone 0.05° around the forward direction (which corresponds to the employed detector). Therefore, in order to have a good statistics in our results we needed to simulate up to $\sim 10^7$ histories, depending on the projectile velocity and the fraction of neutrals we were considering.

[1] In the present work, we use the following parameters to describe the outer electron excitations of the aluminum target: $A_1 = 1$, $\omega_1 = 0.551$ a.u. and $\gamma_1 = 0.035$ a.u. For the L-shell: $A_2 = 0.067$, $\omega_2 = 3.9$ a.u. and $\gamma_2 = 3$ a.u.

Using this computer code we have calculated the energy distribution of the proton fragments exiting from 19 nm thick aluminum foils in the forward direction for the energies used in the experiment. For each incident projectile energy we have done several simulations varying the neutral charge fraction parameter Φ_0 to analyze the dependence of the energy distributions with Φ_0 and to compare them with the experimental spectra.

4. Analysis of results

In order to compare the simulated energy distributions of the proton fragments dissociated from H_2^+ molecules with the corresponding experimental spectra we convoluted the simulated distributions with Gaussians whose widths depend on the foil roughness coefficient and on the experimental energy resolution of the spectrometer [26]. Under the conditions of this experiment, the main broadening arises from the effect of foil roughness.

In Fig. 2 we show the convoluted energy distributions of the proton fragments dissociated from H_2^+ molecules and exiting in the forward direction after traversing a 19 nm thick aluminum foil, and compare them with the experimental results, for different incident energies. The foil roughness coefficient used for the convolutions was 12%. We have included the calculations for several neutral charge fractions to determine the parameter Φ_0 which fits best the experimental distribution widths. All distributions are normalized to unit area. The open circles correspond to the experimental data shown in Fig. 1 while the set of lines correspond to simulations with different values of the neutral charge fraction Φ_0, as indicated.

We can see from the simulations at high incident energies that if the neutral charge fraction is zero (i.e. all the fragments are protons), we obtain energy distributions with two peaks. These peaks correspond to the fragments that are retarded or accelerated due to the Coulomb repulsion. However we observe that, even at the highest energy (101.8 keV/u) the experimental distribution do not show the double-peak structure obtained from the

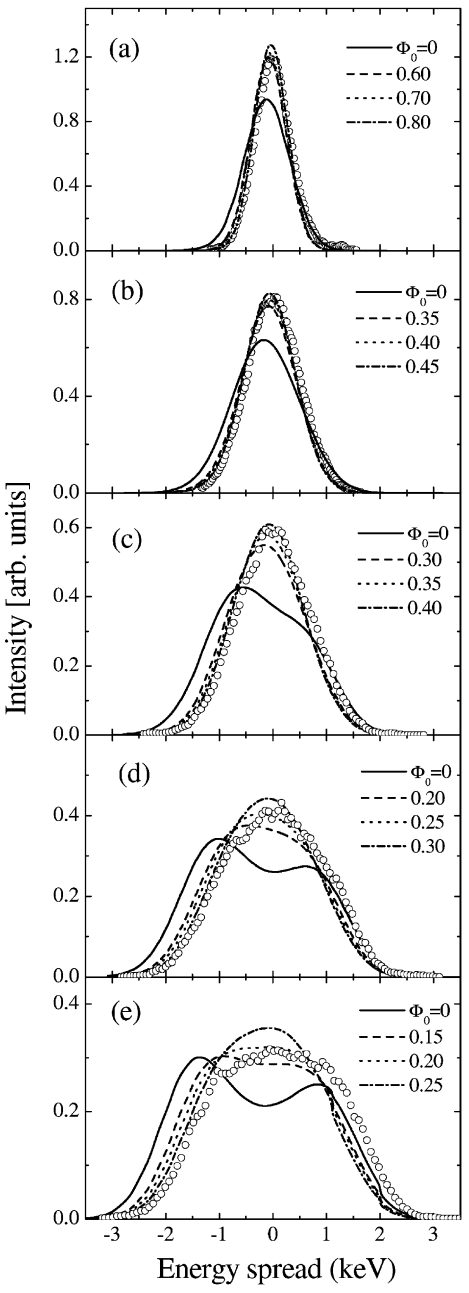

Fig. 2. Convoluted simulation energy distributions of the proton fragments of H_2^+ molecules after traversing a 19 nm thick aluminum foil, together with the experimental data (open circles). The incident molecule energies are the same as in Fig. 1. The simulations corresponding to different values of the neutral charge fraction Φ_0 are indicated by the various lines.

simulation for the $\Phi_0 = 0$ case. This is a strong indication that a significant neutral fraction is present at this energy. We can observe that the simulation with a neutral fraction of $\simeq$20% yields a good representation of the data.

With decreasing energies the neutral charge fraction increases, therefore the path fraction in which both fragments travel as H^+ diminishes, and so the action of the Coulomb repulsion is reduced, leading to a narrower energy distribution as observed in the figures. Simultaneously, there is an increase of those particles which did not experience repulsion at all, therefore increasing the central region of the energy distributions [14,27–29]. As Fig. 2 shows, this behavior is well represented by the simulations considering higher Φ_0 values as the energy decreases. By looking for the best fit with the experimental data we derive the values of Φ_0 representing the charge state fractions of the particles moving at different energies inside the solid.

In Fig. 3 we show the results of Φ_0 obtained by this method, as a function of the incident projectile energy. Additionally we have included in this figure a theoretical estimation [30] of the neutral state fraction, and experimental determinations [15,20, 21] of the neutral fraction of hydrogen projectiles emerging from aluminum foils. We notice the good agreement, within the scatter of the data, among the Φ_0 values obtained by different experiments and the present simulation.

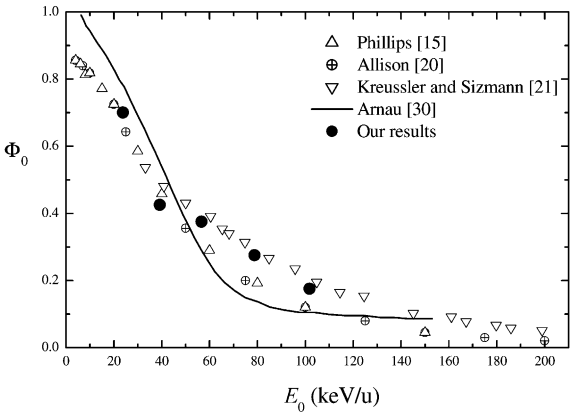

Fig. 3. Neutral charge fraction Φ_0 for H_2^+ fragments in aluminum as a function of the projectile energy. Our results from the simulations are shown by solid circles. Other values for the case of protons in aluminum are also shown: experimental determinations (△: Phillips [15], ⊕: Allison [20], ▽: Kreussler and Sizmann [21]) and a theoretical calculation (solid line: Arnau [30].

5. Concluding remarks

We have studied both experimentally and through a computer simulation code the energy distributions of the fragments resulting from the dissociation of H_2^+ molecules after traversing aluminum foils. It was possible to reproduce the experimental spectra including in the computer simulations two charge states of the hydrogen projectiles inside the foil, a frictional energy loss and its straggling, Coulomb explosion and wake forces, and elastic scattering with the atomic cores. We have focused on the intermediate range of projectile energies ($E_0 \simeq$ 25–100 keV/u) where the variation in neutral charge fraction is most important. This variation in the neutral charge fraction has a direct effect on the Coulomb repulsion undergone by the fragments, which greatly determines the width of the energy distributions. Comparing both experimental and simulated energy distributions we were able to obtain estimations of the charge state fractions of the molecular fragments inside the foil, which are in reasonably good agreement with data obtained by other methods. The shape of the energy distribution is also well described by our simulations.

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