Energy loss and dissociation of 10-MeV/amu H_3^+ ions in carbon foils

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Energy losses and fractions of H_3^+ and H_2^+ ions emerging from carbon foils of 1–8.5 $\mu g/cm^2$ thickness, after incidence of 9.6-MeV/amu H_3^+ ions, have been measured. From the decrease of the transmitted fraction of H_3^+ ions with increasing foil thickness, the dissociation cross section for 9.6-MeV/amu H_3^+ due to the H_3^+-C collision has been determined to be $(2.2\pm0.1)\times10^{-17}$ cm^2. Further, the potential energy of the $H_2^+ + H^+$ system has been derived from the energy spectra of the fragment H_2^+ ions, being 9.45±0.74 eV, with interproton separations being equal to those known for stable H_3^+ ions. For the stopping power of carbon for 9.6-MeV/amu H_3^+ ions a value of 83.3±5.5 eV/($\mu g/cm^2$) has been obtained from the energy-loss data. This result corresponds to an effective charge of 1.45±0.09 for H_3^+ in carbon. The observed stopping power and the derived effective charge agree well with the values calculated by using the first-order Born approximation.

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I. INTRODUCTION

The dissociation of energetic molecular ions in solids is of considerable importance in a number of applications, e.g, cluster fusion and cluster-ion deposition. For H_2^+ ions in the MeV region incident on a thin foil, a dissociation into atomic fragments takes place. In consequence, the fragments repel each other due to a mutual Coulomb interaction, which is a process well known as "Coulomb explosion" [1,2]. However, a small fraction of the incident MeV H_2^+ ions emerges from thin foils [3–5]. This fraction consists either of original H_2^+ ions, namely, those not having undergone a reconstitution process, or of those reconstituted from the fragmented H^+ ions at the exit surface. In the case of incident MeV H_2^+ ions, original H_2^+ ions can be observed only for foils thinner than about 2 $\mu g/cm^2$.

When triatomic molecular H_3^+ and D_3^+ ions of a few MeV are incident on thin foils, H_2^+ and D_2^+ ions will emerge from the foil, in addition to the transmitted H_3^+ and D_3^+ ones [6,7]. These diatomic ions can be seen as the ion's dissociation products, which originate in processes that can be described by the two dissociation schemes $H_3^+ \rightarrow H_2^+ + H^+ + e^-$ and $H_3^+ \rightarrow H_2^+ + H^0$, or by a recombination of fragment H^+ ions. The energy spectra of H_2^+ (or D_2^+) ions show a characteristic three-peak structure with its central peak being due to H_2^+ (or D_2^+) ions, formed by the dissociation $H_3^+ \rightarrow H_2^+ + H^0$, and the two H_2^+ (or D_2^+) outer peaks resulting from the Coulomb explosion of H_2^+-H^+ (or D_2^+-D^+) pairs.

However, as far as we know, no attempt has yet been made to study the energy loss of foil-transmitted original molecular ions in the MeV/amu region, maybe as a result of the small lifetimes of molecular ions penetrating solids. Energy losses of fragments and reconstituted molecular ions have been studied, mainly by using hydrogen cluster ions, and the experiments show that the losses are not just simply integral multiples of the energy losses of isolated atomic ions of the same velocity [8–11]. This fact has been attributed to a so-called "vicinage effect" that arises from the interference of the excitation of solid electrons by the fragments.

In order to study the state of molecular ions in solids, it is advantageous to use faster projectiles and thinner foils, enhancing the transmitted-incident-ion fraction. For the previous experiment, H_2^+ ions having energies of the order of 10-MeV/amu served as projectiles. They were provided by the AVF cyclotron at the Research Center for Nuclear Physics (RCNP) of Osaka University. By applying a high-resolution magnetic spectrograph (RAIDEN) for the ion energy measurements at the incidence of 9.6-MeV/amu H_2^+ ions, transmitted original H_2^+ ions could be observed even for carbon-foil thicknesses of about 10 $\mu g/cm^2$. The stopping power of carbon for H_2^+ and thus the effective projectile charge within carbon were derived. The results are mentioned in our previous paper [12].

In the present paper the transmission of 9.6-MeV/amu H_3^+ ions through carbon foils of various thicknesses is treated. Considerable fractions of incident H_3^+ ions and their H_2^+ dissociation fragments, which survived during the passage through the foil, have been observed. Energy losses and dissociation for H_3^+ ions penetrating carbon foils are reported.

II. EXPERIMENT

A momentum-selected H_3^+ beam from the AVF cyclotron at the RCNP was collimated to a maximum diame-

ter of 1 mm at the target position of a high-resolution magnetic spectrograph (RAIDEN). The 9.6-MeV/amu H_3^+ ions were provided by the cyclotron's low-voltage arc-type ion source. The fraction of H_2^+ ions in the incident H_3^+ beam was less than 5×10^{-3}. Self-supporting carbon foils of thicknesses ranging between 1 and 8.5 $\mu g/cm^2$ served as targets. They were mounted on a movable ladder bearing 12 foils. Only foils that did not show visible pinholes and wrinkles under an optical microscope were chosen. The momentum resolution $\Delta p/p$ for RAIDEN was 4×10^{-5} [full width at half maximum (FWHM)] [13,14]. A position-sensitive proportional counter in conjunction with a plastic scintillation counter was used to detect the energy-selected ions. Subsequent to the energy-loss measurements at the RCNP, we measured the target-foil thicknesses by applying Rutherford backscattering of 2-MeV He^+ ions, provided by the 4-MV Van de Graaff accelerator at Kyoto University.

Due to the fact that a slight change in the beam position at the target resulted in a position shift of the energy-analyzed beam, the ion source of the AVF cyclotron and all the driving currents of the magnets in the beam transport were kept untouched during a set of measurements. In addition, to avoid a possible position shift in the position-sensitive proportional counter, the counting rate of the analyzed ions was reduced to less than 2×10^2 counts/s.

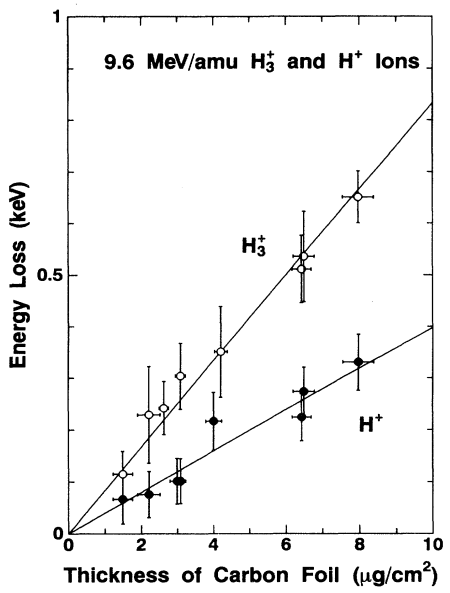

FIG. 2. Dependence of the observed energy loss of H_3^+ ions on the carbon-foil thickness for incident 9.6-MeV/amu H_3^+ ions.

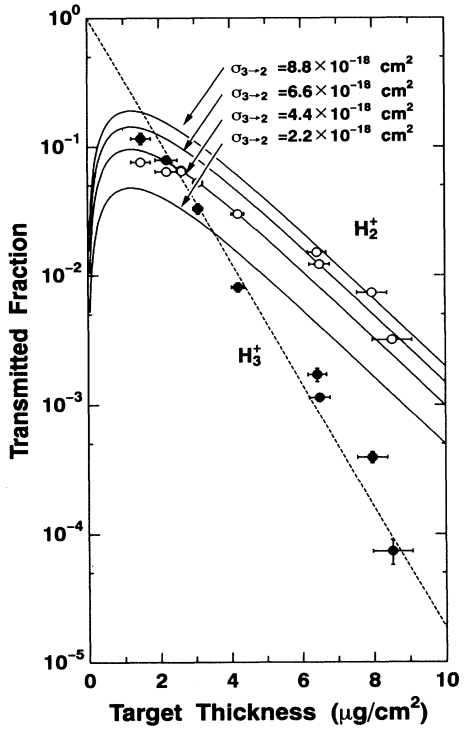

FIG. 1. Dependence of the observed yields of the transmitted H_3^+ (solid circles) and H_2^+ (open circles) ions on the carbon-foil thickness for an incident H_3^+-ion energy of 9.6 MeV/amu. The broken line shows the best fitted $F_3(z)$, and the solid lines show the calculated $F_2(z)$'s with the use of Eq. (4).

Energy spectra of the incident and foil-transmitted H_3^+ ions were measured alternatively throughout the whole experiment to avoid errors due to any possible energy shift of the incident ions. The energy loss of the H_3^+ ions was derived by calculating the difference between the mean energies of the incident and the foil-transmitted beams. Full details of the data processing are given elsewhere [14]. The fractions of the emerging H_3^+ and H_2^+ ions, which were all energy analyzed, were then obtained from the total count ratio deduced from the energy spectra taken with and without the foil.

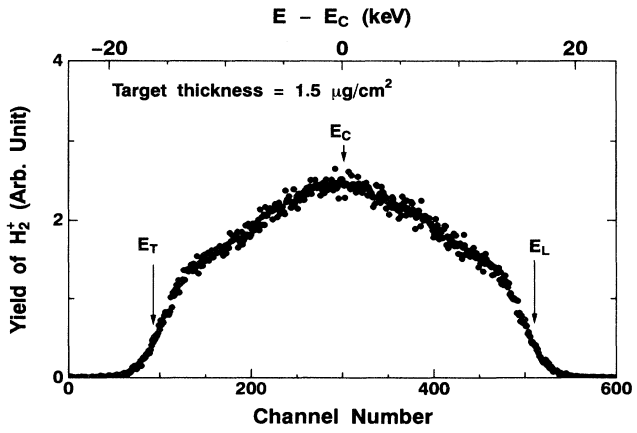

FIG. 3. Energy spectrum of H_2^+ ions emerging from a 1.5-$\mu g/cm^2$ carbon foil for incident 9.6-MeV /amu H_3^+ ions.

III. EXPERIMENTAL RESULTS

Figure 1 shows the dependence of the total yields of the transmitted H_3^+ and the emerging H_2^+ ions on the carbon-foil thickness for incident 9.6-MeV/amu H_3^+ ions. From the exponential decrease of the yield with foil thickness, it is suggested that the observed H_3^+ ions are transmitted original projectiles that have survived against dissociation in the foils. The H_2^+ yield also decreases with increasing foil thickness. However, the fact that the decay curve does not exhibit a straight line in the semilogarithmic plot suggests that the observed H_2^+ ions are dissociation fragments.

Figure 2 shows the measured thickness dependence of the energy loss of H_3^+ ions. For comparison the thickness dependence of the energy loss for 9.6-MeV H^+ ions is displayed as well. Assuming the data can be represented by a straight line passing through the origin, the stopping power S_H of carbon for 9.6-MeV H^+ ions is obtained as 39.6 ± 4.2 eV/(μg/cm^2). This result agrees well with the tabulated stopping power 42.1 eV/(μg/cm^2) [15]. Applying the same procedure to the determination of the stopping power S_{H_3} of solid carbon for 9.6-MeV/amu H_3^+, a value of 83.3 ± 5.5 eV/(μg/cm^2) is found.

Figure 3 displays an example of the energy spectra of the transmitted H_2^+ ions, taken for a 1.5 μg/cm^2 carbon foil. The spectrum shows a broad peak with a shape almost independent of the foil thickness. It can be assumed that the spectrum is composed of a broad peak at the center and a uniform distribution between E_T and E_L. From Fig. 4, which shows the dependence of the energy difference $(E_L - E_T)$ on the foil thickness, it can be seen that this energy difference hardly depends on the latter.

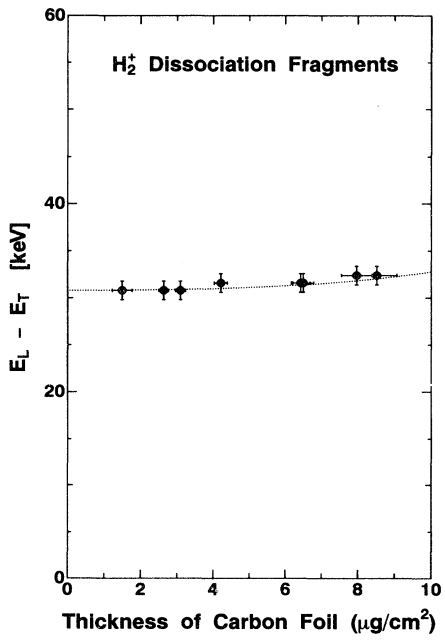

FIG. 4. Dependence of the energy width $(E_L - E_T)$ of the observed H_2^+ ions on the carbon-foil thickness for incident 9.6-MeV/amu H_3^+ ions.

IV. DISCUSSION

A. Thickness dependence of transmitted H_2^+ and H_3^+ fractions

When H_3^+ ions are incident on carbon foils, some fraction of the projectile H_3^+ ions will dissociate, and in consequence fragment ions are formed. To describe the dissociation processes of H_3^+ ions during the collision with carbon atoms, several cross sections are needed. σ_{3d} and σ_{2d} are the dissociation cross sections for H_3^+ and H_2^+, respectively. $\sigma_{3\to2}$ is the cross section for the process leading to the formation of a H_2^+ ion from H_3^+. Thus $\sigma_{3\to2} \leq \sigma_{3d}$.

The fractions $F_3(z)$ for H_3^+ and $F_2(z)$ for H_2^+ ions, after having penetrated a depth z within the foil, satisfy the following differential equations:

$$\frac{dF_3(z)}{dz} = -N\sigma_{3d}F_3(z) , \tag{1}$$

$$\frac{dF_2(z)}{dz} = N\sigma_{3\to2}F_3(z) - N\sigma_{2d}F_2(z) , \tag{2}$$

where N is the density of atoms in the carbon foil. In the above equations, a reconstitution of the molecular ions from their fragments as well as charge-exchange processes are neglected. Introducing the initial condition $F_3(0)=1$, these equations can be solved:

$$F_3(z) = \exp[-N\sigma_{3d}z] , \tag{3}$$

$$F_2(z) = \frac{\sigma_{3\to2}}{\sigma_{3d}-\sigma_{2d}}(\exp[-N\sigma_{2d}z] - \exp[-N\sigma_{3d}z]) . \tag{4}$$

Since the fraction $F_3(z)$ is the yield of H_3^+ ions emerging from a foil of thickness z, the dissociation cross section σ_{3d} for the H_3^+ ions is obtained from the exponential decay of the H_3^+-ion yield displayed in Fig. 1. We find $\sigma_{3d} = (2.2\pm0.1)\times10^{-17}$ cm^2. Figure 1 provides a comparison of the fraction $F_3(z)$ calculated from this cross section with the observed yields.

From the measured yields of the foil-transmitted 9.6-MeV/amu H_2^+ ions the dissociation cross section σ_{2d} for the H_2^+-C collision process has been obtained as $(1.2\pm0.1)\times10^{-17}$ cm^2 [12]. Neglecting the effect of the neighboring-partner H^+ ions on the dissociation of those H_2^+ ions which are dissociation products of H_3^+, this observed cross section may be used as σ_{2d} in Eq. (4). Thus the remaining unknown cross section for $F_2(z)$ in Eq. (4) is $\sigma_{3\to2}$. This can be derived by fitting the calculated $F_2(z)$ with the experimental data. The calculated $F_2(z)$'s are shown by the solid lines in Fig. 1. The best fit to the data gives $(4.4\pm1.0)\times10^{-18}$ cm^2 for the cross section $\sigma_{3\to2}$.

B. Stopping power of carbon for H_3^+ ions and the effective charge of H_3^+

The observed stopping power for 9.6-MeV/amu H_3^+ is 2.10±0.26 times as large as that for protons of the same velocity. Defining the effective charge Z_{eff} of H_3^+ ions in the foil by $Z_{\text{eff}}^2 = S_{H_3}/S_H$, the experimental effective charge Z_{eff} of 9.6-MeV/amu H_3^+ ions in carbon is 1.45±0.09.

The stopping power of carbon foil for 9.6-MeV/amu H_3^+ ions is calculated by applying the first-order Born approximation. This procedure has already been performed for the case of H_2^+ [12]. A general expression for the electronic stopping power is given by [16]

$$\frac{dE}{dx} = \frac{8\pi N e^4}{v^2\hbar^2} \sum_n (E_n - E_0) \times \int_{q_{\min}}^{q_{\max}} \frac{dq}{q^3} |F_{00}^p(-q)|^2 |F_{n0}^t(q)|^2 , \quad (5)$$

where v is the projectile velocity and E_n and E_0 are the energies of the nth excited and the ground state of the target atom. The limits for the integration in Eq. (5) are given by $q_{\min} = (E_n - E_0)/\hbar v$ and $q_{\max} = 2mv/\hbar$, respectively, with m being the electron mass. $F_{00}^p(-q)$ is the elastic form factor of the projectile and $F_{n0}^t(q)$ is the transition form factor of the target atom. The zeroth-order approximation for the wave function of H_3^+ may be expressed as

$$\varphi_{\text{mol}}(\mathbf{r}) = \sqrt{\tfrac{2}{3}}\{\phi_{1s}(\mathbf{r}) + \phi_{1s}(\mathbf{r}-\mathbf{R}_1) + \phi_{1s}(\mathbf{r}-\mathbf{R}_2)\} , \quad (6)$$

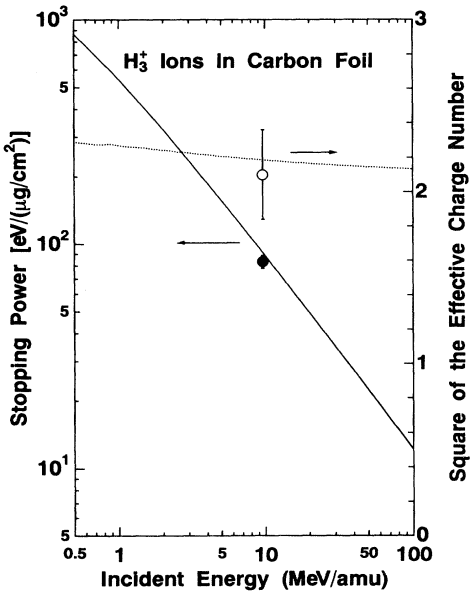

FIG. 5. Calculated stopping power (solid line) of carbon foil for H_3^+ and square of the effective charge (dotted line) of H_3^+ in solid carbon in the energy range 0.5–100 MeV/amu. Experimental data are displayed by circles.

where $\phi_{1s}(\mathbf{r})$ is the ground-state wave function of the hydrogen atom and $\mathbf{R}_j$ $(j=1,2)$ the interproton vector of the ground-state H_3^+ ion. The H_3^+ molecule has equilateral-triangular shape [1,17], and thus $|\mathbf{R}_1| = |\mathbf{R}_2| = R$, where the effect of molecular vibrations is not included.

Neglecting the cross terms of the ϕ_{1s}'s in the calculation of the projectile's form factor $F_{00}^p(q)$, Eq. (5) reduces to

$$dE/dx = \frac{12\pi N Z e^4}{mv^2} \int_{I/\hbar v}^{2mv/\hbar} \frac{dq}{q} \left[1 + \frac{2\sin qR}{qR}\right] \times \left[1 - \frac{2}{3(1+a_B^2 q^2/4)^2}\right]^2 , \quad (7)$$

with a_B being the Bohr radius, and Z and I the atomic number and the mean excitation energy of the target atoms. Taking 0.1 nm for the most probable value of R [1,17] and setting $I = 79$ eV [15] in Eq. (7), the stopping powers of carbon foil for H_3^+ and H^+ of the same velocity were calculated. In Fig. 5 the stopping power as well as the square of the effective charge Z_{eff} for the H_3^+ ions in the energy range from 0.5 to 100 MeV/amu are displayed. For 9.6-MeV/amu H_3^+ ions the calculated stopping power and Z_{eff} are 91.5 eV/(μg/cm²) and 1.48, respectively. These results agree well with the experimental values derived from Fig. 2.

C. Coulomb explosion of H_3^+

When a H_3^+ ion dissociates into a H_2^+ and a hydrogen atom or ion (either H^0 or H^+), both repel each other through the mutual interaction force. Supposing the effect of the foil on the interaction to be of subordinate importance, one-third of the initial potential energy U_{dis} of the H_2^+-H system will be converted into the kinetic energy of H_2^+ in the center-of-mass (c.m.) frame as the explosion develops. Thus the asymptotic c.m. speed of a H_2^+ ion is $[U_{\text{dis}}/(3M)]^{1/2}$, with M being the proton mass. In the laboratory (lab) frame the maximal energy shift $\pm E_{\max}$ of the H_2^+ ions emerging along the beam direction is given by

$$E_{\max} = \tfrac{2}{3}\sqrt{6U_{\text{dis}}E} , \quad (8)$$

where E is the energy per amu of the projectile, and the sign of the shift corresponds to whether the H_2^+ ions are lading (+) or trailing (−) their partner hydrogen ions.

In the present experiment no well-defined peaks for leading and trailing H_2^+ ions could be observed due to the chosen experimental geometry. All H_2^+ ions emerging from the target foils were detected, irrespective of the direction of motion of the fragments in the c.m. frame. In the lab frame, the isotropic explosion of the fragments in the c.m. frame becomes a uniform distribution from $(E_c - E_{\max})$ to $(E_c + E_{\max})$, where E_c corresponds to the energy of the center of mass of the fragments in the lab frame. However, there are two dissociation schemes re-

sulting in the formation of H_2^+ ions: $H_3^+ \rightarrow H_2^+ + H^0$ and $H_3^+ \rightarrow H_2^+ + H^+ + e^-$. As a consequence, the observed energy spectrum of the H_2^+ ions should be a superposition of two trapeziums; one due to H_2^+ ions resulting from the dissociation $H_3^+ \rightarrow H_2^+ + H^0$, and the other due to H_2^+ ions resulting from the scheme $H_3^+ \rightarrow H_2^+ + H^+ + e^-$. In the observed energy spectrum of the H_2^+ ions, displayed in Fig. 3, we assume that the central broad peak is due to H_2^+ ions resulting from the dissociation $H_3^+ \rightarrow H_2^+ + H^0$, because U_{dis} and thus $2E_{max}$ are small in this situation. The underlying trapezium from E_T to E_L corresponds to the H_2^+ ions resulting from the dissociation scheme $H_3^+ \rightarrow H_2^+ + H^+ + e^-$. From the observed $(E_L - E_T)$ extrapolated at zero thickness in Fig. 4, $2E_{max}$ is obtained as 30.8 ± 1.2 eV, and thus for the potential energy U_{dis} released during the explosion $H_3^+ \rightarrow H_2^+ + H^+ + e^-$ the value 9.45 ± 0.74 eV is found. This initial potential energy U_{dis} is that of a H_2^+-H^+ system in which the relative positions of the three protons are those in an equilateral-triangular H_3^+ ion.

Due to experimental uncertainties the potential energy U_{dis} for the $H_2^+ + H^0$ dissociation could not be obtained. However, it should be less than one-third of that measured for the H_2^+-H^+ system.

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