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Triatomic wake effect and the determination of the molecular structure of HD_2^+ from the Coulomb explosion*

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A new theoretical model of the triatomic molecular wake effect is proposed and applied to molecular ions D_3^+ and HD_2^+ while passing through a solid. The wake effects resulting from the reactions of the two similar ions with thin carbon foil are also investigated by using the Coulomb explosion technique. The experimental results are in good agreement with theoretical estimates and the molecular structure of HD_2^+ is determined by using the model.

Keywords: triatomic wake effect, Coulomb explosion, molecular structure, ion HD_2^+

PACC: 3400, 3500, 3600

1. Introduction

The polarization field induced in a foil target by the passage of a swift ion causes the immediately following particles to move towards the trace of the former ion;[1−5] this is called wake effect. Coulomb explosion of the micro-cluster ions yields a lot of temporal and spatial correlation fragments of this kind in target solids. The wake effect is one striking feature of the reaction between a micro-cluster ion with thin foil.[1−7]

The wake field induced by a single ion is well studied both in experimental detections and in theoretical estimates.[2−8] For the triatomic wake effect, only experimental detection of the H_3^+ ion is reported by several groups.[2−4] Theoretical calculation of the triatomic wake effect is absent because of the lack of a theoretical model. In the present paper, our experimental results briefly indicate the wake effect for triatomic molecular ions D_3^+ and HD_2^+ while passing thin carbon foils, with the isotopic effect taken into account, and a theoretical model is proposed to estimate the triatomic wake effect. Based on this method, the geometrical structure of HD_2^+ is determined. Detailed reports of the reactions between H_3^+, D_3^+ and HD_2^+ with solids and the wake effect of H_3^+ have been published elsewhere.[3,5,8−10]

2. Wake effect of Coulomb explosion of D_3^+ and HD_2^+

D_3^+ and HD_2^+ are isotopomers of H_3^+ and possess identical electronic but different rovibrational structure, with smaller vibrational level spacings for larger atomic mass values.[11−13]. We[3−5,8,9] studied their interactions with solids, and the experimental apparatus used is described elsewhere.[3] Briefly, when a fast (MeV) molecular ion passes through a carbon foil, its bonding electrons are all turned off within the first layers of atoms encountered in the target, and the remaining molecular constituents then continue to explode away as clusters of bare nuclei under the action of their mutual Coulomb repulsion. This is called a Coulomb explosion. A high-resolution atomic collision arrangement measures the energy spectra of fragments in the beam direction. Wake effects are indicated as asymmetries in yields and energy shift for 'backward-going' as compared to 'forward-going' protons. The carbon foil used in our measurement is 3 μg/cm^2, which contains only 100 layers of atoms in total. The measurements of the effect for D^+ spectra from the Coulomb explosion of D_3^+ is shown in Fig.1, and the H^+ spectrum from the dissociation of HD_2^+ is shown in Fig.2.[6]

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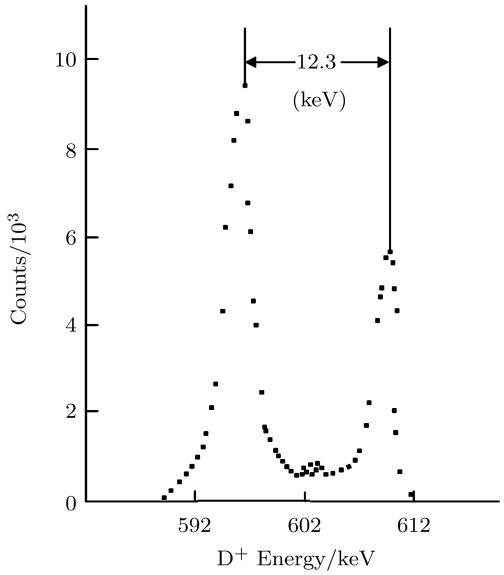


Fig.1. Coulomb explosion spectrum of D^+ from 1.8233 MeV D_3^+ while passing through thin foil.

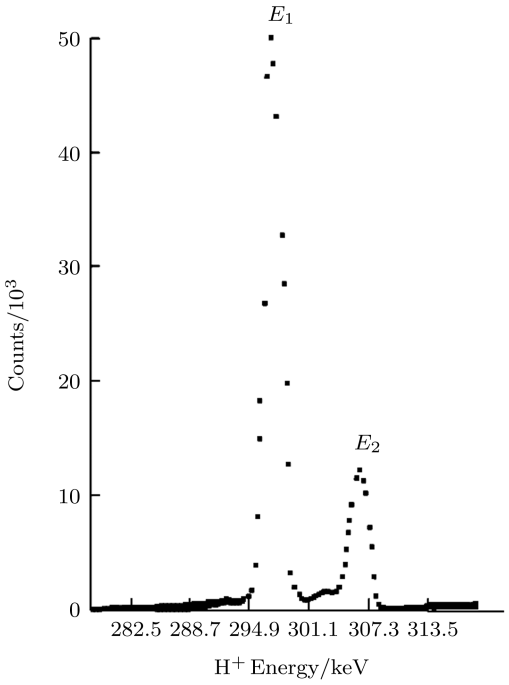


Fig.2. Coulomb explosion spectrum of H^+ from 1.4977 MeV HD_2^+ while passing through thin foil.

3. Theoretical study on the wake effect of triatomic ions while passing through thin foil

The plasma model introduced by Vager *et al*[7] has been proved to be a suitable approach to the wake field of a single particle. According to the electromagnetic theory, the Fourier components of polarization charge and the swift ion charge are related by

$$\rho_{\rm pol}(\boldsymbol{r},\omega)=\rho_{\rm proj}(\boldsymbol{r},\omega)\{[\varepsilon(\omega)]^{-1}-1\}, \qquad (1)$$

and the total polarization charge is

$$\rho_{\rm pol}(\boldsymbol{r},t)=\frac{Ze}{2\pi v}\delta(x)\delta(y)\int_{-\infty}^{\infty}{\rm e}^{-{\rm i}\omega(z/\omega-t)} \times\{[\varepsilon(\omega)]^{-1}\}{\rm d}\omega, \qquad (2)$$

where ε is the dielectric function. The charges are linearly distributed along the Z axis (beam direction).

The calculation of expression (2) needs to know the expression of the dielectric function ε. In the plasma model of Vager *et al*, it is simply given as (a more exact expression is a Legendre function):

$$\varepsilon(\omega)=1-\omega_{\rm p}^2/\omega^2, \qquad (3)$$

where $\omega_{\rm p}$ is the electron plasma frequency of solid media. It is expressed as $\omega_{\rm p}=(4\pi nl^2/m)^{1/2}$, where n is the mean electron number of sympathetic vibration, which usually is the number of outer electrons of the atom. The n value is 4.5[14] for carbon.

Substituting expression (3) into expression (2) gives the polarization charge expression

$$\rho_{\rm pol}(\boldsymbol{z},t)=Ze/a\sin[(z-vt)/a]\delta(x)\delta(y)\theta(vt-z), \qquad (4)$$

where a and $\theta(x)$ are given by

$$a=v/\omega_{\rm p}$$

and

$$\theta(x)=\begin{cases}0 & x\geq 0,\\ 1 & x<0.\end{cases}$$

The expression is a vibrational linear distribution with wavelength $\lambda=2\pi a$. The length is usually within several nm. The expression means that the polarization wave only occurs behind the swift particle.

Clearly, when an ion closely follows a moving charged particle in a solid, the later ion will inevitably be under the action of the polarization wave of the former particle, and the moving trace will be influenced. The influenced moving trace in turn reflects the information of the polarization wave.

Now we begin the discussion of the wake effect of triatomic ions while passing through a thin foil. Typically, we consider the Coulomb explosion of HD_2^+ and focus on the force acting on the proton. We consider a special case, i.e. 0^0 explosion of the proton. This is to say, the H^+ is moving along the beam direction.

In the strictly experimental condition of Coulomb explosion, the fragment H^+ of nonzero degree explosion could not be detected. But with the effect of the

polarization wave, a behindhand H^+ of nearly absolute zero degrees is pushed in the 0^0 direction. This leads to asymmetry in the yield for 'backward-going' as compared to 'forward-going' protons.

As is known, when two closely separated (1–2 Å) particles pass through a solid, the total induced wake field is not a linear combination of wake fields produced by single particles. A mature theory or model is still lacking. Now we give our approach to the theoretical estimation of the wake effect caused by two ions while passing through a thin foil. The new model is based on the approach of the plasma model and in conjunction with the dissociation procedure of a swift molecular ion in a solid. Here are the considerations and approximations in the model:

a) the separating speed of the heavier fragment D^+ is smaller than that of the lighter fragment H^+. Approximately, in the mass-centre frame, the ratio of the varying separation distance between D–D and D–H equals the mass ratio between D and H, i.e. 1/2.

b) the initial separation distance of the two 'forward-going' particles remarkably affects the wake field; here we use the action factor $\mathrm{e}^{-r_{\mathrm{D-D}}}$ to express the effect. When $r_{\mathrm{D-D}} = 0$, the action factor is 1. In this case, the wake field produced by the two deuterons equals the wake field induced by a He^{++}. The action factor can be expressed as $\mathrm{e}^{-r_{\mathrm{D-D}}}\,\mathrm{e}^{-z/2}$ during the processing of separation. This expression has already taken consideration a) into account.

c) the polarization charge wave is a linear vibration distribution with wave length $\lambda = 2\pi a$; this wave length is usually in a range from one to several nm. When a molecular ion with an energy of a few MeV passes through a 100 Å–thin foil, the mean nucleus separation ranges from 2 to 5 Å. Clearly, the 'backward-going' proton is affected by the polarization field only within the first several Å.

d) in the mass-centre frame, the exploded fragments are evenly distributed over the stereo angle.

According to the theory of Coulomb explosion, in the simulation calculation we assume that the behindhand proton is only under the action of direct Coulomb field, which is expressed as $F = ke^2/r$, where r is the separation between the two nuclei, the wake field may be determined by the polarization charge density given in expression (4). Hence, when HD_2^+ is dissociated in the 0^0 direction, the 'backward-going' proton (with a lower energy) P_{L} is under the action of

$$\boldsymbol{F}(\boldsymbol{r}) = e(\boldsymbol{E}(\boldsymbol{r})_{\mathrm{2D-p}} + \boldsymbol{E}(\boldsymbol{r'})_{\mathrm{wake}}), \tag{5}$$

where $\boldsymbol{r}$ is the displacement between the deuteron and the proton, $e(\boldsymbol{E}(\boldsymbol{r})_{\mathrm{2D-p}}$ and $\boldsymbol{E}(\boldsymbol{r'})_{\mathrm{wake}})$ are given by

$$\boldsymbol{E}(\boldsymbol{r})_{\mathrm{2D-p}} = k\frac{e^2}{r^3}\boldsymbol{r} \tag{6}$$

and

$$\boldsymbol{E}(\boldsymbol{r'})_{\mathrm{wave}} = k\int_0^z \frac{e\rho_{\mathrm{pol}}(z)}{\boldsymbol{r'}^3}\boldsymbol{r'}\,\mathrm{d}z, \tag{7}$$

where $\rho_{\mathrm{pol}}(z)$ is written as Eq.(8).

$$\rho_{\mathrm{pol}}(z,t) = Ze/a\,\mathrm{e}^{-r_{\mathrm{D-D}}}\,\mathrm{e}^{-z/2}\sin[(z-vt)/a]\delta(x)\delta(y)\theta(vt-z), \tag{8}$$

then

$$\boldsymbol{E}(\boldsymbol{r'})_{\mathrm{wake}} = k\int_0^z \frac{Ze^2\,\mathrm{e}^{-r_{\mathrm{D-D}}}\,\mathrm{e}^{-z/2}\sin[(z-vt)/a]\delta(x)\delta(y)\theta(vt-z)}{ar'^3}\boldsymbol{r'}\,\mathrm{d}^3r', \tag{9}$$

with

$$\boldsymbol{r'} = \boldsymbol{r}_0 + x\boldsymbol{i} + z\boldsymbol{k},$$

where $\boldsymbol{r}_0$ is the initial displacement of the proton and taken as a constant in our calculation, $\boldsymbol{i}$ and $\boldsymbol{k}$ represent the unit vectors in the vertical and horizontal directions.

Now the vertical velocity of the proton can be obtained through

$$\dot{x} = v_x = \int_0^{z/v_0} (v_{o_x} + a_x(t))\,\mathrm{d}t, \tag{10}$$

where $a_x(t) = e(E_{\mathrm{12D-P}} + E'_{\mathrm{1wake}})/m$. When the behindhand proton moves along the beam direction (z direction), the resultant Coulomb force acting on the two former deuterons is zero in the vertical direction,

expression (10) now becomes

$$a_x(t) = eE'_{1\text{wake}}/m. \tag{11}$$

The horizontal velocity of the proton is given by

$$\dot{z} = v_z = \int_0^{z/v_0} (v_{o_z} + a_z(t))\,\mathrm{d}t, \tag{12}$$

where

$$a_z(t) = e(E_{\text{HP}} + E'_{\|\text{wake}})/m. \tag{13}$$

Since the resistance to the proton from the wake field is infinitesimal and usually ignored,[1−3] then we have

$$a_z(t) = eE_{\text{HP}}/m. \tag{14}$$

We use z_0 as the thickness of the foil, then in the mass-centre frame, the tangent of the emergence angle of the proton in the beam direction is given as $\dot{x}|_{z=z_0}/\dot{z}|_{z=z_0} = \tan\theta$. Through the conversion of coordinates, in the laboratory frame we have

$$\tan\varphi = \frac{\left(\dfrac{ke^2}{r_0 m}\right)^{1/2} \sin\theta}{v_0 + \left(\dfrac{ke^2}{(1+\tan^2\theta) r_0 m}\right)^{1/2}}. \tag{15}$$

As stated above, in the mass-centre frame, the exploded fragments are evenly distributed over the stereo angle. To estimate the relative counts of fragments, only a specific smaller angle which all the exploded protons are pulled into under the action of the wake effect needs calculating. Within the specific angle, the entire fragments are detected by the experimental apparatus. In our experimental arrangement, the angle is 4.5×10^{-4} rad.[3,7]

In our calculation, in order to estimate the influence of the wake effect, the height ratio between the spectra of 'backward-going' and 'forward-going' protons at the same spectrum curve positions is needed. The estimated relative spectrum altitudes represent the relative counts of the explosive fragment.

To obtain the full explosive spectrum, the energy spectrum of the incident beam is needed. For convenience, we use the real explosive spectrum of the proton to fit a Gaussian curve as the spectrum of the incident beam

$$C_{\text{E}} = C_0 \exp(-K(E - E_0)^2), \tag{16}$$

where E_0 is the incident energy of the beam, 1.4977 MeV for our experiment. Derived from the real FWHM 2.1 keV of the proton spectrum, the fitted K in expression (15) is 0.66 keV^{-1}.

As discussed before, the wake force acting on the 'backward-going' proton exists only in the first several Å, in the calculation the integral of $\boldsymbol{E}(\boldsymbol{r}')_{\text{wake}}$ accumulates the value in the first half wavelength of the polarization (the length usually ranges from ten to tens Å). Energy loss and straggling in the thin solid foil are neglected. Furthermore, the action of the wake field is considered to be the same for all 'backward-going' protons. Thus, the calculation result of the wake effect at one point on the 'backward-going' spectrum, i.e. the central peak, will give the whole affected 'backward-going' spectrum.

To examine whether our method has a reasonable accuracy, we first calculate the wake-effect spectra of deuteron after the Coulomb explosion of D_3^+ in the beam direction taking into account all the real parameters in our experiment. A comparison between the calculated results and experimental spectra shows a reasonable agreement. Figure 3 is the estimated spectrum of deuteron after Coulomb explosion from D_3^+ by using the present model. In the experimental

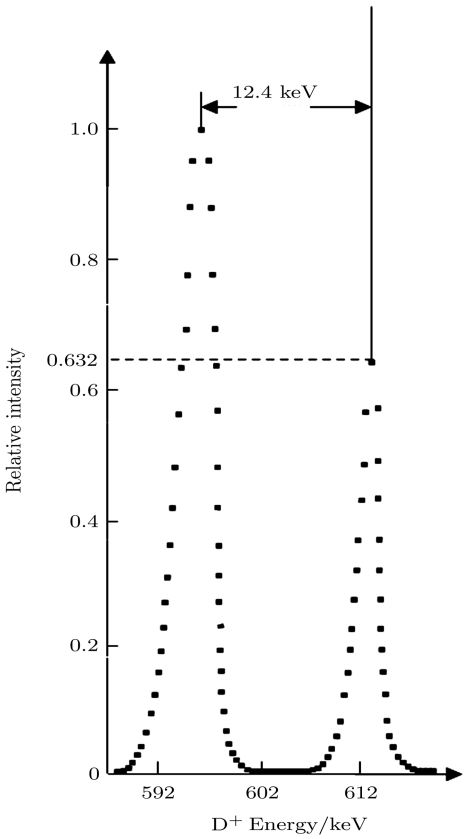


Fig.3. Estimated spectrum of H^+ from Coulomb explosion of 1.8233 MeV D_3^+ in 100 Å-thick carbon foil at 0^0 direction.

spectrum as shown in Fig.1, the ratio between

the 'backward-going' peak and the 'forward-going' peak , $C_{\rm B}/C_{\rm F}$, is 9552/5878, i.e. $C_{\rm B}/C_{\rm F} = 9552/5878 = 1.63$. The ratio of the stereo angles, $C_{\rm B-Relative}/C_{\rm F-Relative}$, which represents the relative count, is estimated to be 1/0.632, i.e. $C_{\rm B-Relative}/C_{\rm F-Relative} = 1/0.632 = 1.58$ as given in Fig.3.

4. Determination of molecular structure of HD_2^+

Naturally, the structure of HD_2^+ is assumed to be that of an isosceles triangle, i.e. a C_{2v} symmetry structure. During Coulomb explosion, the added kinetic energies of the centres of peak fragments D^+ and H^+ are 8.65/2 keV and 10.54/2 keV in the laboratory frame respectively.[7,10] Then the structure of HD_2^+ at an energy of 1.4977 MeV can be determined. But while analysing the data and the process of the dissociation of HD_2^+, we find that two kinds of structures yield the same energy distributions of the fragments D^+ and H^+ as shown in Fig.4.[8] The structure of HD_2^+ cannot be determined only by using the energy spectra of the fragments. In Fig.4(a), $r_{\rm D-D} < r_{\rm D-H}$, and in Fig.4(b), $r_{\rm D-D} > r_{\rm D-H}$. Because the experimental results are the mean values for all the HD_2^+ in the incident beam, the two kinds of structures imply different physical meanings, i.e. to different extents D–D and D–H are separately combined with each other tightly in HD_2^+.

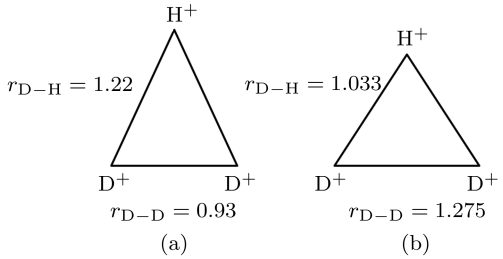


Fig.4. Two structures of $HD2^+$ derived from the energy distribution of the 1.4977 MeV HD_2^+ Coulomb-explosion fragment. The measured spacing is in units of Å.

Now, if our model is used to estimate the energy spectrum of the fragment H^+ in the beam direction, with the two sets of nuclei separations in Fig.4 accepted as parameters separately, the two calculated spectra indicate a remarkable difference in asymmetry between 'backward-going' and 'forward-going' protons as shown in Fig.5.[8] Figure 5(a), which is for the theoretical estimations of the spectrum by using the the structure in Fig.4(a), indicates that the ratio between

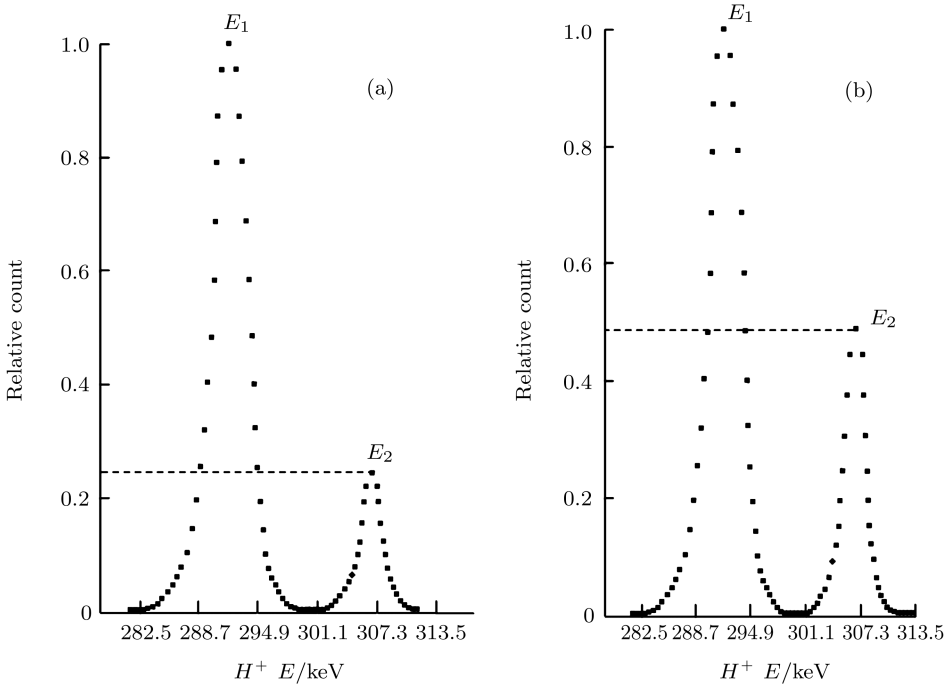


Fig.5. H^+ spectra calculated from the estimation of Coulomb explosion from 1.4977 MeV HD_2^+ in 100 Å carbon foil, where panel 5 (a) shows the spectrum estimated from structure in Fig.4(a), and panel 5(b) indicates the spectrum estimated from structure in Fig.4(b).

the relative counts at the peak centres, $C_{\mathrm{B-Relative}}/C_{\mathrm{F-Relative}}$, is 1/0.21, i.e. $C_{\mathrm{B-Relative}}/C_{\mathrm{F-Relative}} = 1/0.21 = 4.55$. However, the ratio in Fig.5(b), which is a theoretical estimate of the spectrum by using the structure in Fig.4(b), $C_{\mathrm{B-Relative}}/C_{\mathrm{F-Relative}}$, is only 1/0.47, i.e. $C_{\mathrm{B-Relative}}/C_{\mathrm{F-Relative}} = 1/0.47 = 2.13$. When we look back at the experimental spectrum of Fig.2, we find the ratio of the corresponding detected counts, $C_{\mathrm{B}}/C_{\mathrm{F}}$, is 50812/11656, i.e. $C_{\mathrm{B}}/C_{\mathrm{F}} = 50812/11656 = 4.36$. Clearly, the structure in Fig.4(a) is the correct one. Here, we obtain the structural values of HD_2^+ by using the Coulomb explosion technique as

$$r_{\mathrm{D-H}} = 1.22 \text{ Å},$$

and

$$r_{\mathrm{D-D}} = 0.93 \text{ Å}.$$

This states that D–D are combined with each other tightly in HD_2^+ compared with D–H. Actually, this conclusion has also been reached in other studies: in the field of spectroscopy, Carrington and Kennedy[15] noticed that for identical frequency ranges more H^+ fragment ions were contained than D^+ fragment ions. In collision physics, the cross sections for the production of D_2^+ and HD^+ diatomic ionic fragments by dissociation of HD_2^+ in He, in an energy range from 1 to 5 keV, were measured by Alvarez *et al*,[16] showing the cross section for the production of D_2^+ was roughly a factor of 1.5 higher than that of HD^+ on average. Because of this, in the interactions of H_3^+, D_3^+ and HD_2^+, the H^+ fragment from HD_2^+ has the strongest wake effect.

5. Summary

A new theoretical model is proposed to estimate the triatomic wake effect in the field of micro-clusters while passing through a thin foil. Compared with the experimental results for the same particles, the results obtained from the model prove that this model has been successfully used to investigate the wake effect of isotope triatomic molecular ions. Furthermore, the molecular structure of HD_2^+ is also determined by using the model in our Coulomb explosion experiment. The method has potential application in the determination of micro-cluster structures and the study of target materials.

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