Stopping power for hydrogen molecular ions in solids: Influence of the inner-shell electrons of target atoms

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We present calculations of the electronic stopping power for hydrogen molecular ions H_2^+ in solids. These calculations include contributions from both valence electrons and inner-shell electrons of target atoms with the linear dielectric theory and the local-density approximation method. The screened Coulomb potential is used to describe the repulsion process of the two protons. The values of the stopping-power ratio Q for H_2^+ at the high velocities are decreased, obviously due to the contributions of the inner-shell electrons. The theoretical results are compared with some experimental data.

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I. INTRODUCTION

The study of the electronic stopping power for molecular ions moving in solids has been a topic of interest for many years. Motivation for the study comes from some applications, such as molecular-ion implantation, secondary-ion desorption [1] and surface-analysis technology. The recent application of large-cluster beams in cluster-induced fusion studies has also raised new interest in the calculation of the stopping power for molecular ions.

A molecular ion is an aggregate of two or more atomic ions held together by the binding valence electrons. When it impinges on a target, the binding valence electrons are stripped away in the first few atomic layers. The residual ionic fragments then immediately begin to recede from one another under the influence of interionic Coulomb forces. Thus, the distance between the fragments increase and the fragments finally separate in a few femtoseconds. This is called "Coulomb explosion" of the molecular ion. The energy loss of the molecular ion shows important differences—usually called "vicinage effects"—with respect to the separate ions. The origin of the effect is the interference in electronic excitation of the target due to the correlated motion of the ions in the cluster.

For hydrogen molecular ions H_2^+ the vicinage effect has been showed by Brandt and collaborators [2]. They reported measurements of the energy loss of hydrogen molecular ion beams through films of carbon and gold at energies of 75 and 150 keV/n in which the molecular stopping power per nucleus exceeded the atomic stopping power by as much as 20%. Following the work of Brandt and co-workers, the similar experimental measurements for the stopping power of molecular ions have been made by several authors [3,4]. Especially Ray *et al.* [5] recently reported the experimental results for the vicinage effects of large hydrogen clusters H_n^+ (up to $n=25$) in the energy range 10–120 keV per proton.

At the same time, with various theoretical models a number of authors have calculated the vicinage effect. One model is the linear dielectric theory [6–8] and another model is the quantum scattering theory [9,10]. The latter is mainly used to treat lower velocity molecular ion stopping. In both models the electron system of a target is regarded as a homogeneous valence-electron gas. In fact, when a molecular ion moves through a solid it can interact not only with the homogeneous valence-electron gas, but also with inner-shell electrons of target atoms, especially in the high projectile velocity.

In previous works [11,12], the contributions of the inner-shell electrons to the stopping power of the atomic ions H^+ have been considered with the local-density approximation method. In this work, we will extend the local-density approximation method for atomic ions to describe molecular ions H_2^+.

In the next section we present the general description of the stopping power for molecular ions H_2^+ according to the linear dielectric formalism and the local-density approximation method. In Sec. III, the Coulomb explosion of the molecular ions will be analyzed using a screened Coulomb potential. Calculations of the stopping-power ratio and the comparison of theoretical results with experimental data are presented in Sec. IV. Section V summarizes the conclusions of this work.

II. DIELECTRIC THEORY AND THE LOCAL-DENSITY APPROXIMATION

Let us consider a hydrogen molecular ion H_2^+ moving with velocity **v** through a solid. Separation between the

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two point charges is expressed by the vector **R**. In many experimental situations, the orientations of **R** are randomly distributed. Within the framework of the linear dielectric theory, the general expression for the stopping power of such hydrogen molecular ion is given by

$$(-dE/dx)_{H_2^+}=(4\pi e^4/m_e v^2)n_0[L(n_0,v)+I(n_0,v,R)] , \tag{1}$$

where n_0 is the valence-electron gas density, e is the element charge, and m_e is the electron mass. In Eq. (1), the proton stopping number $L(n_0,v)$ and the interference function $I(n_0,v,R)$ can be expressed by the dielectric function $\varepsilon(k,\omega)$ of the electron gas

$$L(n_0,v)=\left[\frac{2}{\pi\omega_p^2}\right]\int_0^\infty (dk/k)\int_0^{kv} d\omega\,\omega\,\mathrm{Im}[-1/\varepsilon(k,\omega)] \tag{2}$$

and

$$I(n_0,v,R)=(2/\pi\omega_p^2)\int_0^\infty dk[\sin(Rk)/(k^2R)] \times\int_0^{kv} d\omega\,\omega\,\mathrm{Im}[-1/\varepsilon(k,\omega)] , \tag{3}$$

where $\omega_p=(4\pi e^2 n_0/m_e)^{1/2}$. A local-field correction dielectric function [13] is used in present work for including the effect of the exchange-correlation interaction of the electron gas. The method of the numerical calculation for the above integrals has been discussed in detail in previous works [14].

The local-density approximation assumes that each volume element of the solid is an independent plasma with local electron density $n(r)$. Lindhard and Winther found that this approximation yields the same velocity dependence as the shell correction known from the atomic target system. With the substitution of $n(r)$ for n_0, an average electronic stopping power of the hydrogen molecular ion is obtained by integrating over the atomic volume in Eq. (1),

$$\overline{(-dE/dx)}_{H_2^+}=(4\pi e^4/m_e v^2)N[\bar{L}(v)+\bar{I}(v,R)] , \tag{4}$$

where

$$\bar{L}(v)=\int_0^{r_0} n(r)L(n(r),v)4\pi r^2 dr , \tag{5}$$

$$\bar{I}(v,R)=\int_0^{r_0} n(r)L(n(r),v,R)4\pi r^2 dr , \tag{6}$$

where N is the atomic density of the solid and $r_0=[3/(4\pi N)]^{1/3}$ is the atomic average radius.

It is necessary to know the solid-state electron density for calculating the stopping power. The electron density in the solid differs from that of a free atom due to the overlap of the electronic wave function. A simple model of the solid-state electron density $n(r)$ has been presented in previous works [11,12]. In the model $n(r)$ can be expressed by

$$n(r)=\begin{cases} n_0, & r\geq r_c \\ n_A(r), & r<r_c \end{cases} , \tag{7}$$

where $n_A(r)$ is the free-atom electron density and r_c is the radius of the inner shell of a target atom. r_c can be fixed by the continuity condition $n_A(r_c)=n$. Using the atomic independent model of Green, Sellin, and Zachor [15], a parametrized expression of $n_A(r)$ will be used in present work.

Under the local-density approximation, the stopping power of protons

$$\overline{(-dE/dx)}_{H^+}=(4\pi e_4/m_e v^2)NL(v) \tag{8}$$

has been calculated and a fitted expression of $\overline{(-dE/dx)}_{H^+}$ dependent on eight parameters based on the numerical results has been given [11,12]. Figure 1 shows the comparison of the theoretical results with the experimental data [16–18] for $\overline{(-dE/dx)}_{H^+}$ for protons in carbon films. It can be seen that the theoretical predictions agree well with the experimental data.

III. COULOMB EXPLOSION FOR HYDROGEN MOLECULAR IONS

In this section we consider the Coulomb repulsion for H_2^+. It is clear that the stopping power $\overline{(-dE/dx)}_{H_2^+}$ depends on the internuclear distance R, which is determined by the interionic Coulomb potential. At high projective velocities, the potential is an unscreened Coulomb potential $V(R)=e^2/R$ and the expression of the penetrating depth D as a function of R is given by

$$D=D_0[\sqrt{R/R_0}\sqrt{R/R_0-1} + \ln(\sqrt{R/R_0}+\sqrt{R/R_0-1})] , \tag{9}$$

where $D_0=v(M_p R_0^3/4e^2)^{1/2}$, M_p is the proton mass, and $R_0=1.07$ Å is the initial internuclear distance (the bond length) [19].

At low velocities, however, the screening by target electrons will be strong and so the two protons will ex-

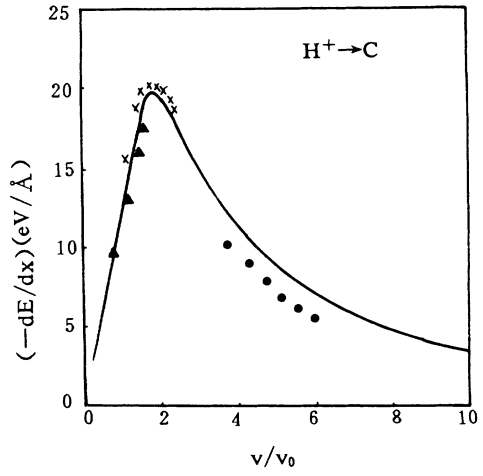

FIG. 1. The electronic stopping power for a proton in a carbon target as a function of proton velocity. The solid line is the theoretical result [Eq. (8)] and the sources of the experimental data are as follows: ▲, Ref. [16]; ×, Ref. [17]; ●, Ref. [18].

perience little repulsion. It is difficult to give the correct form of the potential $V(R)$ in this case. As a first approximation we will estimate R using the screened Coulomb potential

$$V(R)=e^2\exp(-R/a)R\ , \tag{10}$$

where

$$a=\begin{cases} v_F/\omega_p, & v\le v_F \\ v/\omega_p, & v>v_F \end{cases} \tag{11}$$

is the screening length, $v_F=(3\pi^2 n_0)^{1/3}a_0 v_0$ is the Fermi velocity of the electron gas, $a_0=0.529\times10^{-8}$ cm is the Bohr radius, and $v_0=2.18\times10^8$ cm/s is the Bohr velocity. The anisotropy of the polarization around each moving ion has been neglected in this form of the potential, which can modify the initial orientation of the molecular axis. With the potential, the equation of the relative motion for H_2^+ is given by

$$\frac{d^2R}{dD^2}=\left[\frac{2m_e}{m_p}\right]\left[\frac{v_0}{v}\right]\left[\frac{1}{R^2}+\frac{1}{Ra}\right]e^{-R/a}\ . \tag{12}$$

For H_2^+ moving in carbon film ($D=400a_0$), the internuclear distance R as a function of the velocity v is shown in Fig. 2. Here the continuous line corresponds to the screened potential and the dashed line to the unscreened Coulomb potential. By comparing the curves, one can see that in low-velocity regions there is obviously a discrepancy between both results. In Fig. 3 the internuclear distance R as a function of the penetrating depth D is shown for H_2^+ in carbon at a velocity $v=4v_0$.

IV. RESULTS FOR THE STOPPING-POWER RATIO

For comparison of numerical results with experimental data, it is useful to introduce the stopping-power ratio

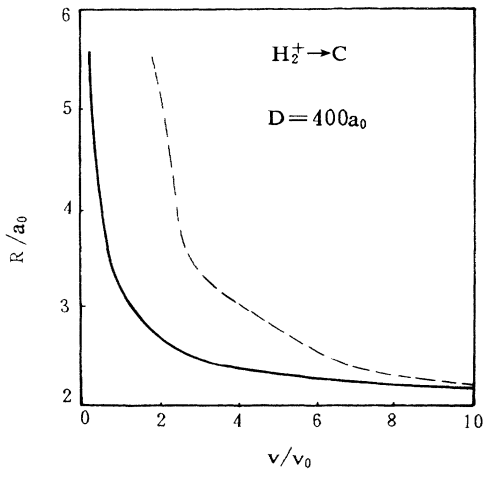

FIG. 2. The internuclear distance for H_2^+ in a carbon target as a function of the projectile velocity. The continuous line and the dashed line correspond to the screened potential and unscreened potential, respectively.

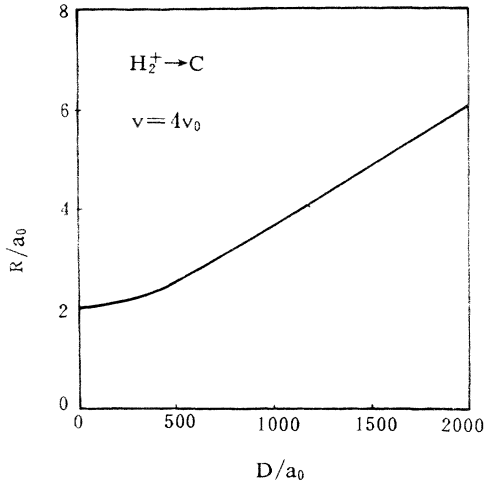

FIG. 3. The internuclear distance for H_2^+ in carbon target as a function of the penetrating depth.

$$Q=\overline{(-dE/dx)_{H_2^+}}/[2\overline{(-dE/dx)_{H^+}}]$$
$$=1+\bar{I}(v,R)/\bar{L}(v)\ . \tag{13}$$

In Fig. 4 the influence of the inner-shell electrons of target atoms to the stopping-power ratio Q is shown for H_2^+ in carbon and for $R=3a_0$. Here the continuous line is given by the results of the local-density approximation based on Eqs. (13) and (4), and the dashed line by the results of the homogeneous valence-electron gas based on Eq. (1). It can be seen from the figure that at high velocities the results of the stopping-power ratio are obviously decreased due to the contributions of the inner-shell electrons.

Recently, Ray *et al.* [5] have presented a body of ex-

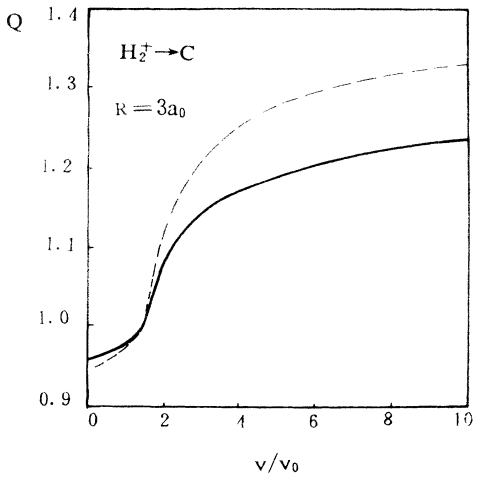

FIG. 4. The stopping-power ratio for H_2^+ in carbon target as a function of the projectile velocity. The continuous line corresponds to the results of the local-density approximation and the dashed line to the results of the homogeneous valence-electron gas.

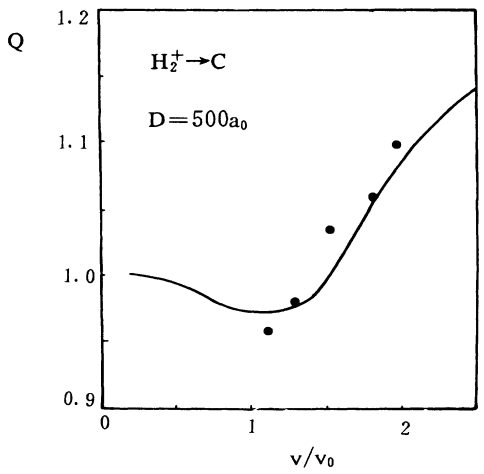

FIG. 5. The stopping-power ratio for H_2^+ in a carbon target as a function of the projectile velocity. The solid line is the theoretical result and the dots are the experimental data [5].

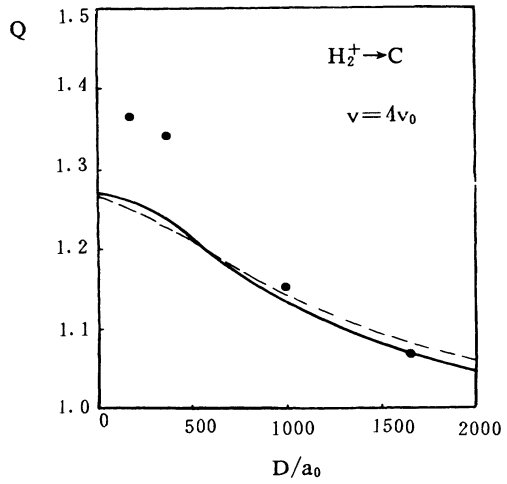

FIG. 6. The stopping-power ratio for H_2^+ in carbon target as a function of the penetrating depth. The solid line is our theoretical result, the dashed line is the result of Steinbeck *et al.* [21], and the dots are the experimental data [20].

perimental data for the energy loss of large hydrogen clusters H_n^+ (up to $n=25$) in the energy range 10–120 keV per proton and has also given a model for describing the energy loss, which was derived on the basis of an oscillator model of the target response. In the low-energy region, the agreement between theory and experimental data is quite well; however, at high velocities the theoretical predictions are systematically above the experimental data. In Fig. 5 we show the comparison of our theoretical results with the experimental data for H_2^+ in a carbon film ($D=500a$). One can see that our results agree very well with the experimental data.

Steuer *et al.* [20] have measured the stopping-power ratio Q as a function of the target thickness D for H_2^+ incident on carbon, projectile velocity $v=4v_0$. Figure 6 shows the comparison of the theoretical results with experimental data. Here the dots are the experimental data, the solid line corresponds to our calculations, and the dashed line to the calculations of Steinbeck *et al.* [21]. In the work of Steinbeck *et al.* the contributions of the inner-shell electrons have been taken into account with the theory of the atomic collision. We can see from this figure that for larger target thickness both theoretical results are close to the experimental data; however, for small target thickness both theoretical results are below the experimental data.

V. SUMMARY

We have investigated the stopping-power ratio for hydrogen molecular ions H_2^+ with the linear-response dielectric theory and the method of the local-density approximation. The Coulomb explosion of H_2^+ is considered using a screened Coulomb potential and the internuclear distance R is calculated for various values of projectile velocity and target thickness. It has been found that the inner-shell electrons of target atoms act by dropping the values of the stopping-power ratio at high velocities. In comparison with experiment it has been shown that our investigation is reasonable. The calculations of energy loss for large ion clusters will be reported in future work.

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