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Stopping power of fast, partially stripped molecules and clusters: Vicinage effects in charge states

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We present a consistent variational approach to the vicinage effect on ion charges in a cluster made of partially stripped ions, based on the Brandt–Kitagawa theory for isolated ions, supplemented by dynamically screened interionic interactions. Interferences in cluster stopping power are shown to be strongly influenced by the ion-charge dependence on internuclear separations, and may give rise to energy loss enhancement or deenhancement, when compared to the case when the ion charges are frozen at the value corresponding to isolated ions at the same speed. In addition, the ion charge reduction is shown to be responsible for softening the interionic interactions responsible for Coulomb explosion. Calculations of the vicinage effect on ion charges are performed for a fast N_2 molecule moving through a carbon target, and the roles of Coulomb explosion and multiple scattering in energy loss are discussed.

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I. INTRODUCTION

Following recent advances in accelerator techniques for production of energetic beams of atomic clusters, at speeds in excess of the Bohr velocity v_B, much interest has been raised over the past few years in energy loss of fast correlated ions in matter [1–6]. It is expected that a massive energy deposition introduced by a large cluster in a small volume of the target may give rise to qualitatively new effects in such applications as materials modification [7] and inertial confinement fusion [8]. On the theoretical side, soon after the first description of energy losses of fast di- and tri-proton clusters in solids [9], a theoretical model for stopping power of large clusters has been developed by Arista [10] using a linear-response formalism [11], in conjunction with statistical models of cluster structure [12]. The current status of research in cluster stopping in solids is critically examined in recent reviews [13–15], where open questions and future trends are outlined.

A simplified scenario of cluster penetration through solid targets can be summarized as follows. After entering a solid target, an atomic cluster at the speed $v>v_B$ will lose its binding valence electrons after traversing a few atomic layers, and the charge state of the emerging ions will be equilibrated within about 1 fs due to intense electron capture and loss processes in the target [16]. After this short initial break-up of the cluster, the constituent ions still retain the initial cluster structure, and their further penetration is accompanied by electronic excitations of the target, which exhibit strong interference due to close spatial correlation among the ions. This so-called vicinage effect [10] gives rise to enhanced energy loss per cluster ion, when compared to the energy loss of an individual ion moving at the same speed, as long as the interionic distances within the cluster are smaller than the characteristic length of the electronic excitation, v/ω_0, ω_0 being a resonance frequency of target electrons. At prolonged penetration times, a spatial dispersion of the cluster occurs due to Coulomb explosion [9,17], driven by the dynamically screened interionic interactions, which proceeds in an adiabatic manner in the cluster frame of reference, on a time scale of some 10 fs. Another mechanism of cluster dispersion, which competes with Coulomb explosion [13,18], is provided by multiple elastic scattering of cluster constituent ions on target atoms [19,20]. In either case, the vicinage effect on cluster stopping power will gradually diminish with increasing penetration time through the target, as a result of increasing interionic distances.

While the above scenario has been experimentally confirmed for small [9] and large [1,2] hydrogen clusters, where significant energy loss enhancements were demonstrated in thin foils and at high cluster speeds, the situation is quite different with clusters made of heavier atoms, such as C_n [3–5] and B_n [5,6]. In such cases, experimental data generally exhibit [3–6] a very weak vicinage effect on cluster stopping power, with quite contradicting tendencies of slightly enhanced or slightly deenhanced energy loss per ion in different measurements for the same type of clusters and the same target. Measurements using heavy-atom molecules N_2 [21] and O_2 [5] also show some deenhancement of energy losses per ion. One of the major reasons for this suppression, or even reversal, of the energy loss enhancement in heavy-atom clusters has been attributed in recent reviews [14,15] to the fact that the constituent ions are only partially stripped of their electrons, and that their charge state is influenced by the vicinity of other ions in the cluster. Experi-

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mental measurements on fast N_2 molecules [22] and C_n clusters [23] have shown that the average charge of cluster constituent ions, after traversing a thin carbon foil, emerges significantly reduced compared to the charge of a single ion at the same speed. The mechanism responsible for this charge reduction is taken to be the vicinage effect, where the neighboring ions create a perturbation of the potential, which results in a stronger binding of the remaining electrons at each ion within the cluster. Such a vicinage effect has been confirmed by measurements with different foil thicknesses [22,23], showing that the charge-state reduction diminishes with increasing penetration time on a time-scale characteristic of Coulomb explosion and/or multiple scattering.

Vicinage effect on charge state of partially stripped ions in clusters of arbitrary size has been recently described by a theoretical model [24] which combines self-energy calculations for atomic clusters in solids [25,26] with the variational theory of Brandt and Kitagawa (BK) [27] for electron structure of isolated partially stripped ions in solids. Our model [24] has successfully described experimental data [23] for the dependence of average ion charge on the number n of constituent particles in carbon clusters C_n $(3\leqslant n\leqslant 10)$, as well as the dependence of this charge on the target thickness, based on cluster dispersion due to multiple scattering. Two further applications of this model [24] are elaborated in the present report, dealing with the roles of the vicinage effect on ion charges in cluster stopping power and in Coulomb explosion, the issues which are considered in recent reviews [13–15] to be of utmost importance for cluster penetration through solids. In this first approach to these problems, we present some general theoretical results, while the calculations are performed for diatomic molecules, such as N_2. We note, however, that the role of the vicinage effect on ion charges will be more pronounced in larger clusters, as evidenced in Refs. [23,24]. In Sec. II, we summarize the self-consistent variational procedure of Ref. [24] for obtaining the ion charges in a heavy-atom cluster. In Sec. III, a recent model of Arista's [15] for energy loss of clusters with partially stripped ions is combined with the variational ion charges to describe the dependence of stopping power for a fast N_2 molecule on the internuclear distance and on the molecule speed. Modifications of the interionic potentials due to the vicinage effect on ion charges, and their influence on Coulomb explosion of N_2, are discussed in Sec. IV. Concluding remarks are given in Sec. V. Atomic units (a.u.) are used throughout, unless otherwise indicated.

II. GENERALIZATION OF BRANDT–KITAGAWA THEORY TO CLUSTERS

In the BK theory [27], which provides one of the most efficient methods for dealing with the energy loss of partially stripped ions in solids [28,29], the distribution of N electrons, bound at an isolated ion with the nuclear charge Z, is modeled by the radially symmetric function $N(4\pi\Lambda^2 D)^{-1}\exp(-D/\Lambda)$. Here, D is the electron-nucleus distance, while the ion size is defined by the parameter Λ, with the value $\Lambda=(2a/Z^{1/3})(N/Z)^{2/3}/(1-(1/7)(N/Z))$, where $a=0.24$. Electronic energy of an isolated ion is obtained in the form

$$E_0(N)=-\frac{Z^{7/3}}{4a}\left(\frac{N}{Z}\right)^{1/3}\left(1-\frac{1}{7}\frac{N}{Z}\right)^2, \tag{1}$$

which agrees reasonably well with the result of the Thomas–Fermi model [29]. Dynamical equilibrium value of the charge $Q=Z-N$ on the ion, which moves through the solid at a speed v, is then obtained in the BK theory by minimizing the electron energy in the laboratory system, $Nv^2/2+E_0(N)$ (so-called energy stripping criterion [29]).

Next consider an n-component heteronuclear cluster, entering a solid target with velocity $\mathbf{v}$. The change in individual ion velocities due to energy loss, Coulomb explosion, or multiple scattering will be negligible compared to v, so that one may assume that all the ions move with constant velocity $\mathbf{v}$ in the course of penetration through the target. Let Z_j and N_j be the nuclear charge and the number of bound electrons at the jth ion $(1\leqslant j\leqslant n)$, which is placed at the position $\mathbf{r}_j$ in the cluster center of mass frame of reference. Dynamically screened interionic interaction energy of the cluster, or the vicinage energy, can be written as [24,26]

$$E_v=\frac{1}{2}\sum_{j=1}^{n}\sum_{j\neq l=1}^{n}\int\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{4\pi}{k^2}\frac{\rho_l^*(\mathbf{k})\rho_j(\mathbf{k})}{\epsilon(k,\mathbf{k}\cdot\mathbf{v})}e^{i\mathbf{k}\cdot(\mathbf{r}_l-\mathbf{r}_j)}, \tag{2}$$

where $\epsilon(k,\omega)$ is the dielectric function of the medium [11], and $\rho_j(\mathbf{k})$ is the space Fourier transform of the total (nuclear plus electronic) charge distribution localized at the jth ion. Using the BK model for bound electrons, one obtains [27]

$$\rho_j(\mathbf{k})\equiv\rho_j(k)=Z_j\frac{q_j+k^2\Lambda_j^2}{1+k^2\Lambda_j^2}, \tag{3}$$

with $q_j=1-N_j/Z_j$ being the ionization degree of the jth ion. We further use the fact that the typical ion sizes Λ_j are much smaller than the interionic distances $|\mathbf{r}_l-\mathbf{r}_j|$ to obtain a point-charge approximation for the vicinage energy by taking $\rho_j(\mathbf{k})\simeq\rho_j(k=0)=Z_j-N_j$ in Eq. (2). Thus, the total cluster energy in the laboratory frame of reference consists of the kinetic energy of all bound electrons due to cluster motion at speed v, the internal energy of isolated ions, and the vicinage energy,

$$E_L=\sum_{j=1}^{n}N_j\frac{v^2}{2}+\sum_{j=1}^{n}E_0(N_j)+\sum_{j=1}^{n-1}\sum_{l>j}^{n}(Z_j-N_j)(Z_l-N_l)U(\mathbf{r}_{jl}), \tag{4}$$

where

$$U(\mathbf{r}_{jl})=\int\frac{d^3\mathbf{k}}{(2\pi)^3}\frac{4\pi}{k^2}\cos(\mathbf{k}\cdot\mathbf{r}_{jl})\mathrm{Re}\left[\frac{1}{\epsilon(k,\mathbf{k}\cdot\mathbf{v})}\right] \tag{5}$$

is the dynamically screened interaction potential between two unit point charges at the relative position $\mathbf{r}_{jl}=\mathbf{r}_l-\mathbf{r}_j$.

Following the variational approach of the BK theory [27], we apply the energy stripping criterion [29], where we seek the minimum of the total energy E_L relative to the electron populations N_j on all the ions. This minimum is obtained by solving n coupled equations $\partial E_L/\partial N_j=0$, viz.,

$$\frac{v^2}{2}+E_0'(N_j)-\sum_{j\neq l=1}^{n}(Z_l-N_l)U(\mathbf{r}_{jl})=0,\quad \text{for}\quad 1\leqslant j\leqslant n, \tag{6}$$

with $E_0'(N)$ being the derivative of the function in Eq. (1). Systematic solution of these equations provides, in principle, all the equilibrium ion charges throughout the cluster traveling at the velocity $\mathbf{v}$, while taking full account of the spatial distribution of ions and any anisotropy of the dynamically screened interactions. It is clear that each ion charge, obtained in this way, is a function of the coordinates of all ions, so that the vicinage term (double summation) in Eq. (4) is no longer a pairwise superposition of interionic interaction potentials. In the same time, the vicinage term of Eq. (4) determines the interactions responsible for the (adiabatic) dynamics of Coulomb explosion of the cluster in its own frame of reference, thus making self-consistent solutions of equations of motion for all ions a serious challenge.

Even in a static case, solving Eqs. (6) for ion charges may be quite tedious in a general case of a large heteronuclear cluster with a given frozen structure relative to the direction of motion $\mathbf{v}$. We have used in [24] a simplifying assumption that all ion charges N_j are equal to an average value N in a randomly oriented homonuclear cluster, $Z_j=Z$, and employed a two-particle distribution function [12,20,26] to describe the structure of such a cluster. However, such simplifying assumptions are not required to solve the system of Eqs. (6) for the case of a homonuclear di-cluster ($n=2$) with the nuclear charges $Z_1=Z_2=Z$, where the minimum of the energy E_L is obtained by a symmetric solution, $N_1=N_2=N$. Here, the electron population N on each ion in the di-cluster is obtained by solving the equation

$$\frac{v^2}{2}+E_0'(N)-(Z-N)\,U(\mathbf{r})=0, \tag{7}$$

so that the equilibrium value of N becomes an implicit function, $N=N(\mathbf{r})$, of the inter-nuclear relative position $\mathbf{r}\equiv\mathbf{r}_{12}$. A second-derivative test shows that such a symmetric solution provides a global minimum of E_L for a di-cluster, as long as the internuclear distance r exceeds several ion sizes Λ, in accord with the above mentioned point-charge approximation for E_L. We note that the result of standard BK theory [27,29] for the equilibrium electron population N_0 of a single isolated ion at speed v can be obtained from Eq. (7) by letting $U(\mathbf{r})\to 0$, that is, $r\to\infty$.

An important problem in calculations of the vicinage effect on ion charge states is the modeling of electronic response of the medium by an appropriate dielectric function $\epsilon(k,\omega)$ in Eqs. (3) and (5). We note at once that this problem is intimately connected to the modeling of Coulomb explosion, because the interaction potential (5) plays central roles in both problems. We shall focus here only on models describing the collective response of a quasi-free electron gas in the solid, but a few words will be mentioned in Sec. IV regarding a possible role of the core electrons [30,31] in the interionic potential [32,33]. In traditional frozen-charge approaches to Coulomb explosion of both small [5,34–36] and large [1,37] clusters, the most popular approach employs an empirical Yukawa-type interaction potential. In the present context, we obtain the interaction potential (5) in a Yukawa-like form $U_Y(r)=r^{-1}\exp(-k_s r)$, by using $\mathrm{Re}[1/\epsilon(k,\omega)]\simeq k^2/(k^2+k_s^2)$, where k_s^{-1} is a velocity-dependent screening length [38], with the high-velocity limit $k_s^{-1}\simeq v/\omega_p$, ω_p being the electron plasma frequency of the target. A more realistic approach to Coulomb explosion uses a dynamically screened interaction potential, exhibiting an oscillatory wake pattern, which is responsible for a tendency of diatomic molecules to align in the direction of motion [17,39,40], and gives rise to asymmetric Coulomb explosion patterns in large clusters [41]. In the present study of the vicinage effect on ion charges in a di-atomic molecule, we reduce the parameter space of the theory by ignoring the orientation effects, and only consider the wake effects on a *randomly oriented* molecule, whence the factor $\cos(\mathbf{k}\cdot\mathbf{r})$ in Eq. (5) is to be replaced by $(kr)^{-1}\sin(kr)$. By using the frequency-dependent dielectric function in Eq. (5), $\epsilon(k,\omega)=1-\omega_p^2/\omega(\omega+i\gamma)$ with $\gamma\to 0^+$, we then obtain an analytical result [26], exhibiting a wake pattern in the interaction potential, as follows

$$U_w(r)=\frac{1}{r}\cos\left(r\frac{\omega_p}{v}\right)+\frac{\omega_p}{v}\,\mathrm{si}\left(r\frac{\omega_p}{v}\right), \tag{8}$$

where si is the sine integral. We note that, when radially symmetric model potentials $U_Y(r)$ and $U_w(r)$ are used in Eq. (7), the solutions for the electron populations in a homonuclear di-cluster will be also radially symmetric functions, $N=N_Y(r)$ and $N=N_w(r)$.

As a quantitative example, we consider a randomly oriented N_2 molecule ($Z=7$) with an equilibrium internuclear distance $r_0=2.12$, moving in a carbon target with $\omega_p=25$ eV. We obtain the ionization degrees $q=1-N/Z$ from Eq. (7) with the Yukawa- and the wake-type model potentials and plot in Fig. 1 the ratios q/q_0 (q_0 being the ionization degree of an isolated N ion at the same speed) against the internuclear distance r, for two speeds: $v=1.7$ and $v=2.3$. It is seen that the positive (repulsive) interactions in the model potentials $U_Y(r)$ and $U_w(r)$ generally reduce the ion charges of the constituent ions in the molecule, when compared to the charge of an isolated ion, while the attractive (negative) parts of the wake-type interaction $U_w(r)$ enhance those charges. It is also obvious that the wake-type interaction makes the vicinage effect on ion charges much more sensitive to the speed than the Yukawa-type interaction. Similar conclusions can be drawn from Fig. 2, where we plot the ratios q/q_0 versus the speed v for two internuclear distances, $r=r_0=2.12$ and $r=2r_0=4.24$.

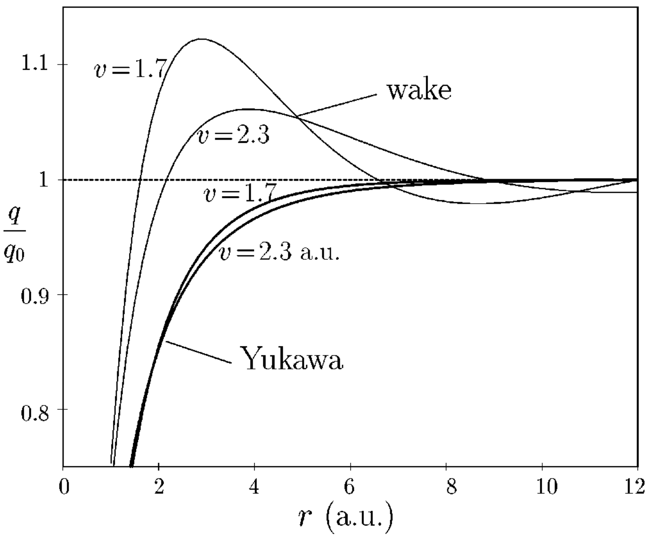

FIG. 1. Ratio of the vicinage ion charge q to the isolated-ion charge q_0 versus the internuclear distance r (in a.u.) of N_2 in carbon, for two speeds, $v=1.7$ and $v=2.3$ (in a.u.), with the Yukawa-(thick line) and the wake-(thin line) type interactions.

III. STOPPING POWER OF PARTIALLY STRIPPED MOLECULE

In the most recent development of a theoretical model for cluster energy loss, Arista [15] provides the expression for the stopping power of an n-component cluster made of partially stripped ions, viz.,

$$S_{cl}=\frac{i}{v}\int \frac{d^3\mathbf{k}}{(2\pi)^3}\,\frac{4\pi}{k^2}\,\frac{\mathbf{k}\cdot\mathbf{v}}{\epsilon(k,\mathbf{k}\cdot\mathbf{v})}\left[\sum_{j=1}^{n}|\rho_j(\mathbf{k})|^2 + \sum_{j=1}^{n}\sum_{j\neq l=1}^{n}\rho_l^*(\mathbf{k})\rho_j(\mathbf{k})e^{i\mathbf{k}\cdot(\mathbf{r}_l-\mathbf{r}_j)}\right]. \tag{9}$$

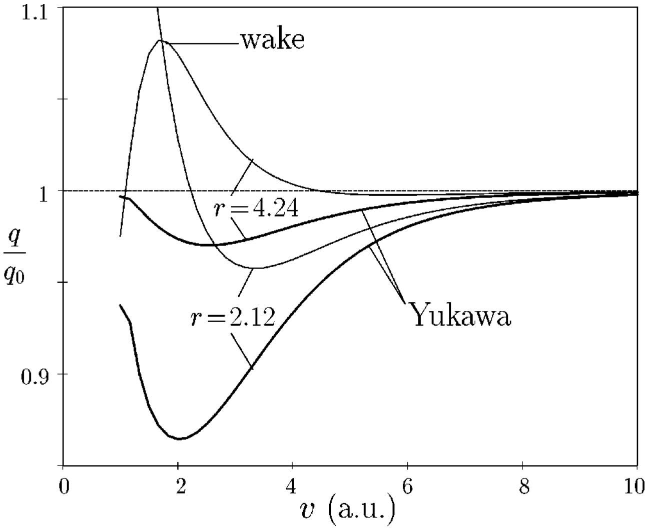

FIG. 2. Ratio of the vicinage ion charge q to the isolated-ion charge q_0 versus speed v (in a.u.) of N_2 in carbon, for two internuclear distances, $r=2.12$ and $r=4.24$ (in a.u.), with the Yukawa-(thick line) and the wake-(thin line) type interactions.

Here, the space Fourier transform of the total charge distribution $\rho_j(\mathbf{k})$ at the jth ion in the cluster is given by Eq. (3), where the ionization degree q_j and the ion size Λ_j are determined by a solution of the equations (6) for the electron populations N_j which, in turn, depend on the coordinates of all ions. Thus, the vicinage effect on ion charges is incorporated, in a complicated way, in both the incoherent part (single summation) and the coherent, or vicinage, part (double summation) of the stopping-power expression (9). This complex picture simplifies somewhat for a homonuclear di-cluster ($n=2, Z_1=Z_2=Z$), in which case $N_1=N_2=N(\mathbf{r})$, and consequently $\rho_1(k)=\rho_2(k)=\rho(k;\mathbf{r})$, where we indicate an implicit dependence of the ionic charge distributions on the relative internuclear position $\mathbf{r}$. Thus, the stopping power formula for a di-cluster reduces to

$$S_{cl}=\frac{1}{\pi^2 v}\int d^3\mathbf{k}\frac{\mathbf{k}\cdot\mathbf{v}}{k^2}\rho^2(k;\mathbf{r})[1+\cos(\mathbf{k}\cdot\mathbf{r})]\mathrm{Im}\left[\frac{-1}{\epsilon(k,\mathbf{k}\cdot\mathbf{v})}\right]. \tag{10}$$

We note that, letting $r\rightarrow\infty$ amounts to $\cos(\mathbf{k}\cdot\mathbf{r})\rightarrow 0$ in Eq. (10), and one thus obtains the incoherent stopping power of two isolated ions

$$S_0=\frac{4}{\pi v^2}\int_0^{\infty}\frac{dk}{k}\rho_0^2(k)\int_0^{kv}d\omega\,\omega\,\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right], \tag{11}$$

which is exactly twice the result of the standard BK theory [27], with the charge distribution $\rho_0(k)$ given by Eq. (3) where the electron population N_0 of an isolated ion is to be used. For a randomly oriented molecule with a finite internuclear distance r, we have a radially symmetric $N=N(r)$ and consequently $\rho=\rho(k;r)$, as discussed in the previous section, while the factor $\cos(\mathbf{k}\cdot\mathbf{r})$ in the stopping power formula Eq. (10) is to be again replaced by $(kr)^{-1}\sin(kr)$, giving

$$S_{cl}=\frac{4}{\pi v^2}\int_0^{\infty}\frac{dk}{k}\rho^2(k;r)\left[1+\frac{\sin(kr)}{kr}\right]\int_0^{kv}d\omega\,\omega\,\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right]. \tag{12}$$

Vicinage effect on cluster energy loss is best expressed in terms of the stopping-power ratio $R=S_{cl}/S_0$, which compares the total (incoherent plus coherent) stopping power of the n-component cluster to the stopping power of n isolated ions. Note carefully that, for a randomly oriented di-cluster, S_{cl} contains the ion charges which depend on the internuclear separation r and thus are already modified by a vicinage effect, while the ion charges used in S_0 correspond to isolated ions at the same speed.

Calculations of stopping powers (11) and (12) for a randomly oriented N_2 molecule in a carbon target are performed with a plasmon-pole approximation [11] for the dielectric function, while the ionization degrees of Sec. II are used to evaluate the charge densities $\rho(k;r)$ and $\rho_0(k)$. In Fig. 3, we plot the stopping-power ratios R, obtained from the Yukawa-type (thick solid lines) and the wake-type (thin solid lines) interactions for ion charges, versus the internuclear distance

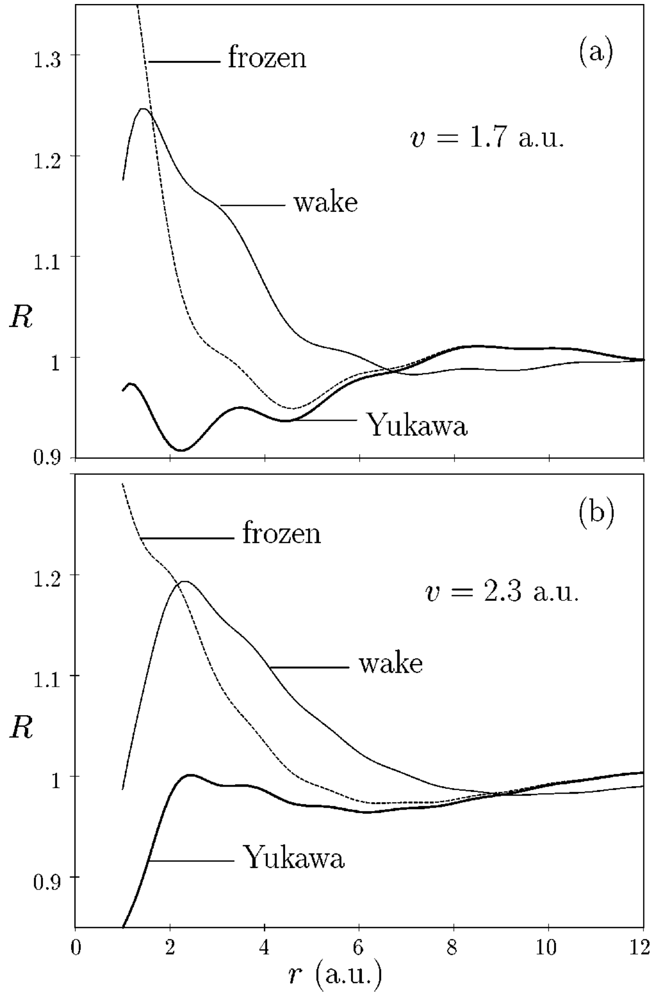

FIG. 3. Stopping-power ratio $R = S_{cl}/S_0$ of N_2 in carbon versus the internuclear distance r (in a.u.), for two speeds: (a) $v = 1.7$ and (b) $v = 2.3$ (in a.u.). Ion charges are evaluated with the Yukawa-(thick line) and the wake-(thin line) type interactions, or taken to be frozen (dashed lines) at the value q_0 for an isolated ion.

r for two speeds, (a) $v = 1.7$ and (b) $v = 2.3$. In addition, we have also calculated the stopping-power ratio R when the ion charges are frozen and equal to the charge of an isolated ion, that is, when S_{cl} in Eq. (12) is evaluated with $\rho_0(k)$ rather than $\rho(k;r)$. These results are displayed in Fig. 3 by the dashed lines, showing only the vicinage effect on stopping power, with the usual energy loss enhancement ($R > 1$) for small internuclear distances, which diminishes as the ions separate ($R \rightarrow 1$ as r increases). It is obvious that the combined vicinage effects on both the ion charges and the stopping power, as displayed by the solid lines in Fig. 3, may alter the stopping power in a profound manner when compared to the frozen-charge case. Depending on the model interaction used to obtain the vicinage effect on ion charges, we may have additional energy loss enhancement (related to the attractive parts in the wake-type potential), or energy loss deenhancement ($R < 1$) when the interactions are positive. In Fig. 4, we show the velocity dependence of the stopping power ratios for two internuclear separations, $r = 2.12$ and $r = 4.24$, when the ion charges are frozen (dashed lines), or

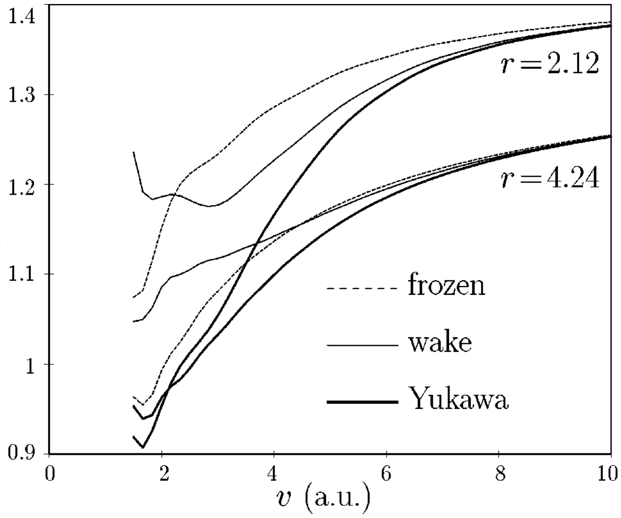

FIG. 4. Stopping-power ratio $R = S_{cl}/S_0$ of N_2 in carbon versus speed v (in a.u.), for two internuclear distances: $r = 2.12$ and $r = 4.24$ (in a.u.). Ion charges are evaluated with the Yukawa-(thick solid line) and the wake-(thin solid line) type interactions, or taken to be frozen (dashed lines) at the value q_0 for an isolated ion.

evaluated with the wake-type (thin solid lines) and the Yukawa-type (thick solid lines) interactions. At moderate speeds v, the three types of ion-charge treatments show marked differences but, as v increases, they all converge to steadily increasing energy-loss enhancements which are more pronounced at shorter internuclear distances.

IV. ROLE OF VARIABLE ION CHARGES IN COULOMB EXPLOSION AND MULTIPLE SCATTERING

The ion charges, obtained from the variational procedure of Sec. II, in general depend on the geometric structure of the cluster, and therefore will modify the interionic interactions responsible for the dynamics of Coulomb explosion. In particular, for a homonuclear di-cluster, we have obtained from Eqs. (5) and (7) the equilibrium charges on the constituent ions, $Q_1 = Q_2 = Q(\mathbf{r}) \equiv Z - N(\mathbf{r})$, which vary with the internuclear distance $\mathbf{r}$ in an adiabatic manner. Now, the actual Coulomb explosion is driven by the effective interaction potential between the two partially stripped ions in a di-cluster, given by $V(\mathbf{r}) = Q^2(\mathbf{r})U(\mathbf{r})$, where $U(\mathbf{r})$, Eq. (5), is the dynamically screened interaction between a pair of unit point charges. For a randomly oriented N_2 in carbon, we use the ion charges obtained from the Yukawa- and the wake-type interaction potentials (see Fig. 1), and calculate the corresponding radially symmetric effective potentials $V(r)$. The results are displayed in Fig. 5 by thick solid lines for the two speeds (a) $v = 1.7$ and (b) $v = 2.3$, and are compared to the effective interaction potentials $V(r) = Q_0^2 U(r)$ (shown by thin solid lines), with the ion charges frozen at the value Q_0, corresponding to an isolated ion at the same speed. It is seen that the vicinage effect on ion charges makes all the repulsive interactions in the effective potentials softer than in the case of ions with frozen charges. As predicted in Ref. [13], this softening of the interaction potential will make Coulomb explosion slower than what would be expected if the charges

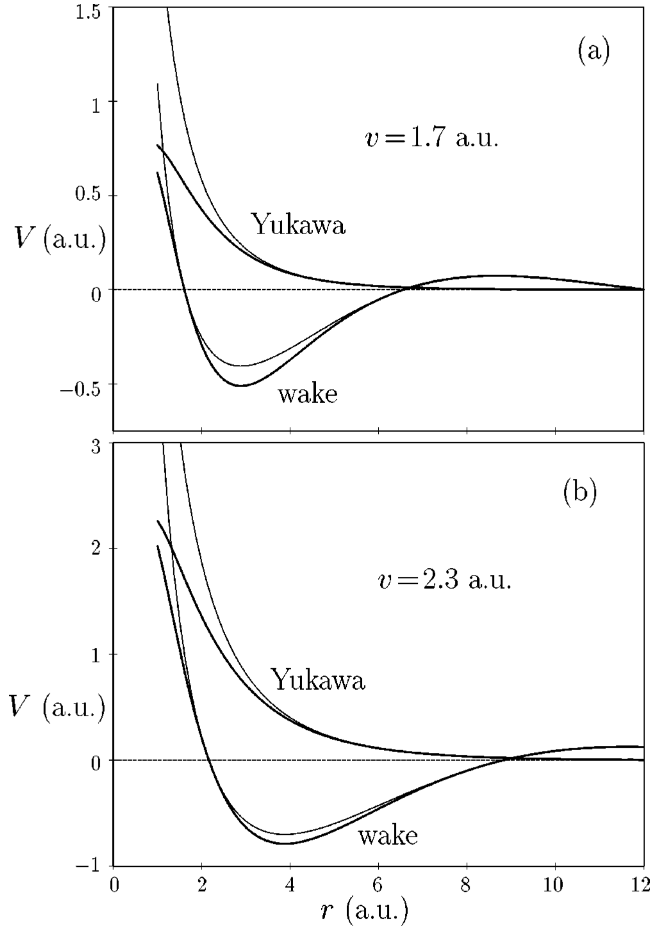

FIG. 5. Effective interaction potentials V (in a.u.) for a randomly oriented N_2 in carbon versus the internuclear distance r (in a.u.), for two speeds: (a) $v = 1.7$ and (b) $v = 2.3$ (in a.u.), based on the Yukawa- and the wake-type interactions. Thin solid lines are obtained with the frozen ion charges q_0, while the thick solid lines show the modification of the potentials due to vicinage effect on ion charges.

were frozen at the value of isolated ions. On the other hand, the attractive wells in the wake-type potential are deepened by the vicinage effect on ion charges, thus further strengthening possible wake-assisted bound states of the two constituent ions [42]. In either case, it seems that the vicinage effect on ion charges renders Coulomb explosion a less effective mechanism of the cluster spatial dispersion, while the multiple scattering should not be affected by the variable ion charges.

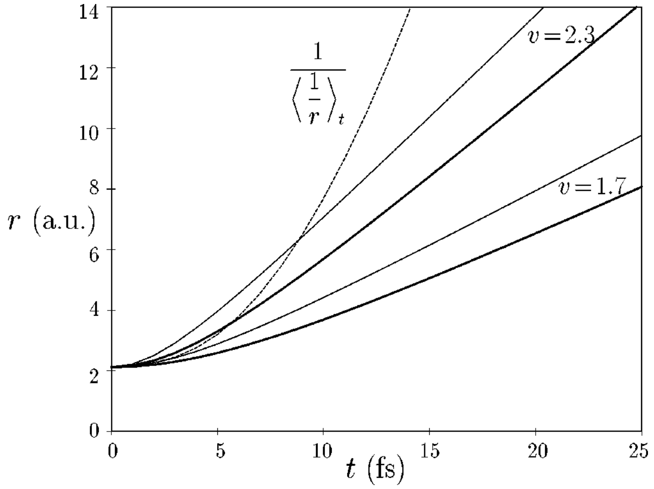

FIG. 6. Dependence of the internuclear separation r (in a.u.) on time t (in fs) in Coulomb explosion governed by Yukawa-type interaction for N_2 in carbon for two speeds, $v = 1.7$ and $v = 2.3$ (in a.u.). Thin solid lines are obtained with the frozen ion charges q_0, while the thick solid lines show the modification of the Coulomb explosion due to vicinage effect on ion charges. Effective internuclear separation due to multiple scattering, $(\langle r^{-1} \rangle_t)^{-1}$, is shown by the dashed line.

While these conclusions add to a debate on relative roles of Coulomb explosion and multiple scattering [13,18], we wish to comment here on modeling the interaction potential, Eq. (5), based on a model for the electronic response of the target. In particular, the role of core electrons in the interionic interactions within a cluster seems to be unclear at present. Apart from several studies of cluster energy losses, which include the contributions of core electrons [30,31,36], there seem to exist only two very recent studies of modifications of the induced potential by a fast ion due to core electrons [32,33]. It has been shown that [33], in a more realistic description of the target dielectric function, the oscillatory wake pattern in the induced potential may be damped compared to the induced potential when only the free-electron gas response is considered. Such a conclusion may be rationalized based on the fact that the screening lengths due to the core electrons, v/ω_0, are much shorter than the length of a free-electron gas response, v/ω_p, owing to much higher resonance excitation frequencies ω_0 of core electrons than the plasma frequency ω_p [14]. It may then be expected that the wake-type model potential, used in the previous two sections, overemphasizes the oscillations in the wake pattern, and that the Yukawa-type potential serves a better role in modeling the vicinage effect on ion charges in a cluster. While no definite judgment can be made at this point from the theoretical side, it is suggested by the results presented in Sec. II (Figs. 1 and 2) that an experimental affirmation of strong wake effects on ion charges in a cluster would require observations of both reduced *and* increased ion charges, as compared to the isolated ions at the same speed. As mentioned before, so far only charge reduction has been observed experimentally [22,23], providing an empirical affirmation of the Yukawa-type interaction. Led by this discussion, we have solved the equation of motion for Coulomb explosion of a N_2 molecule in carbon, using the effective interaction potential $V(r)$ of the Yukawa type. The results for internuclear distance r versus time t are shown in Fig. 6 for two speeds, $v = 1.7$ and $v = 2.3$, where the thick solid lines depict the effect of variable ion charges on Coulomb explosion, as compared to the case of frozen ion charges, shown by the thin solid lines. These curves clearly demonstrate a quantitative effect of slowing down of Coulomb explosion due to the vicinage effect on ion charges.

It is also interesting to compare the temporal evolutions of the internuclear separation r due to Coulomb explosion and due to multiple scattering of a di-cluster, in order to gain an idea about the relative strengths of the two mechanisms of cluster dispersion. The stochastic nature of multiple scattering is best described by a distribution function $F(\mathbf{r};t)$ of internuclear separations **r** after the penetration time *t*, for a di-cluster which was initially oriented in random directions, with an equilibrium internuclear distance r_0. Multiple elastic scattering of each constituent ion in the di-cluster by the target atoms can be represented by a random sequence of small-angle deflections in the laboratory frame, with negligible change of the ion velocity **v**. As a result, in the cluster center-of-mass frame, each ion undergoes two-dimensional displacements **R** in a plane perpendicular to **v**, which are distributed according to a function $H(\mathbf{R};t)$ after the penetration time *t*. Assuming that the random deflections of the cluster constituent ions are statistically independent from each other, one can obtain a three-dimensional Fourier transform of the distribution function $F(\mathbf{r};t)$ as follows: $F(\mathbf{k};t) = |H(\mathbf{K};t)|^2(kr_0)^{-1}\sin(kr_0)$, where $H(\mathbf{K};t)$ is a two-dimensional Fourier transform of the multiple scattering distribution function for a single ion $H(\mathbf{R};t)$, with **K** being the component of the wave vector **k** perpendicular to **v**, such that $\mathbf{k}=(\mathbf{K},k_\parallel)$ and $\mathbf{k}\cdot\mathbf{v}=k_\parallel v$ [20]. With the normalization $H(\mathbf{K}=0;t)=1$ of the multiple scattering distribution, we ensure that the distribution of inter-nuclear separations is normalized as well, $F(\mathbf{k}=0;t)=1$, for all $t \geqslant 0$. The analytical expression for $H(\mathbf{K};t)=H(K;t)$, after traversing the distance vt through the target, is obtained by using the small-angle approximation for a central-potential elastic scattering of the projectile ion on target atoms [19,20]. On using an inverse-square scattering potential, one further obtains a simple, yet reliable, result $H(K;t)=\exp(-cKt^2)$, which does not depend on the cluster speed v and where *c* is a constant determined by the atomic parameters of both the target and the projectile particles [19,20]. Unfortunately, the first moment of the internuclear separation, $\langle r\rangle_t$, taken with the distribution $F(\mathbf{r};t)$, does not exist in such a simple model of multiple scattering. However, it makes sense to evaluate the temporal evolution of the average of the inverse separation, $\langle r^{-1}\rangle_t$, due to multiple scattering, when comparison is made with the change of *r* with time in a Coulomb explosion. Namely, $(\langle r^{-1}\rangle_t)^{-1}$ is such a separation that would give the same (bare) Coulomb interaction among unit charges as the true average of that interaction, taken with the distribution $F(\mathbf{r};t)$. The time dependence of $(\langle r^{-1}\rangle_t)^{-1}$ is shown in Fig. 6 by a dashed line for a randomly oriented N_2 molecule, as a measure of the increase in internuclear separation due to multiple scattering in a carbon foil. Clearly, the effect of multiple scattering is comparable to the effect of (dynamically-screened) Coulomb explosion in initial stages of penetration through the target, but the multiple scattering takes over for prolonged penetration times [13].

Finally, let us discuss the relative roles in energy losses of N_2 in carbon target, played by Coulomb explosion, driven by the Yukawa-type interaction, and the multiple scattering. We perform calculations separately for these two mechanisms of cluster spatial dispersion, keeping the ion charges modified by the vicinage effect due to increasing internuclear distances in each case. This is achieved for Coulomb explosion simply by recalculating the stopping power (12) versus the penetration time, $S_{cl}=S_{cl}(t)$, by using the relations $r=r(t)$, shown in Fig. 6 by thick solid lines. For the multiple scattering case, however, we have to modify the interaction potential Eq. (5) for a di-cluster, as follows:

$$U(t)=\int \frac{d^3\mathbf{k}}{(2\pi)^3}\,\frac{4\pi}{k^2}\cos(\mathbf{k}\cdot\mathbf{r_0})|H(\mathbf{K};t)|^2\mathrm{Re}\left[\frac{1}{\epsilon(k,\mathbf{k}\cdot\mathbf{v})}\right], \tag{13}$$

where $\mathbf{r}_0$ is the initial internuclear separation. On using Eq. (13) in Eq. (7), we obtain the electron populations and the charge distributions in terms of the penetration time, $N=N(t)$ and $\rho=\rho(k;t)$. Next, in the case of multiple scattering, the expression (10) has to be modified as well, giving a time-dependent stopping power

$$S_{cl}(t)=\frac{1}{\pi^2 v}\int d^3\mathbf{k}\frac{\mathbf{k}\cdot\mathbf{v}}{k^2}\rho^2(k;t)[1+\cos(\mathbf{k}\cdot\mathbf{r}_0)|H(\mathbf{K};t)|^2] \times \mathrm{Im}\left[\frac{-1}{\epsilon(k,\mathbf{k}\cdot\mathbf{v})}\right]. \tag{14}$$

Using the analytical result for the multiple scattering distribution $H(\mathbf{K};t)=H(K;t)$, obtained from an inverse-square scattering potential, along with the assumption of an N_2 molecule which is initially oriented in random directions [20], we evaluate both the ion charges and the stopping power in terms of penetration time *t* through a carbon target.

Finally, to make contact with the energy-loss measurements, we evaluate

$$\langle S_{cl}(T)\rangle=\frac{1}{T}\int_0^T dt\, S_{cl}(t), \tag{15}$$

where *T* is the actual penetration time through a target of thickness vT, and present the results for the averaged stopping power ratios $\langle R(T)\rangle=\langle S_{cl}(T)\rangle/S_0$ in Fig. 7, for N_2 in a carbon target with two speeds: (a) $v=1.7$ and (b) $v=2.3$. The thick solid lines in Fig. 7 represent the data for Coulomb explosion (CE) and multiple scattering (MS), including variable ion charges due to increasing interionic distances, while the thin solid lines show the cases when the ion charges are taken frozen at the value corresponding to isolated ions at the same speeds. In addition, we also show in Fig. 7 the points presenting the experimental data from Ref. [21], which were among the first demonstrations of the energy loss deenhancement in molecular stopping power. It is seen that both Coulomb explosion and multiple scattering exhibit energy loss deenhancement, compatible with the experimental data, when the ion charges are reduced due to vicinage effect determined by the Yukawa-type interaction. When the vicinage effect on ion charges is ignored by using frozen charges of isolated ions, a constructive interference in the stopping power due to the vicinage effect takes over and gives rise to the familiar energy loss enhancement.

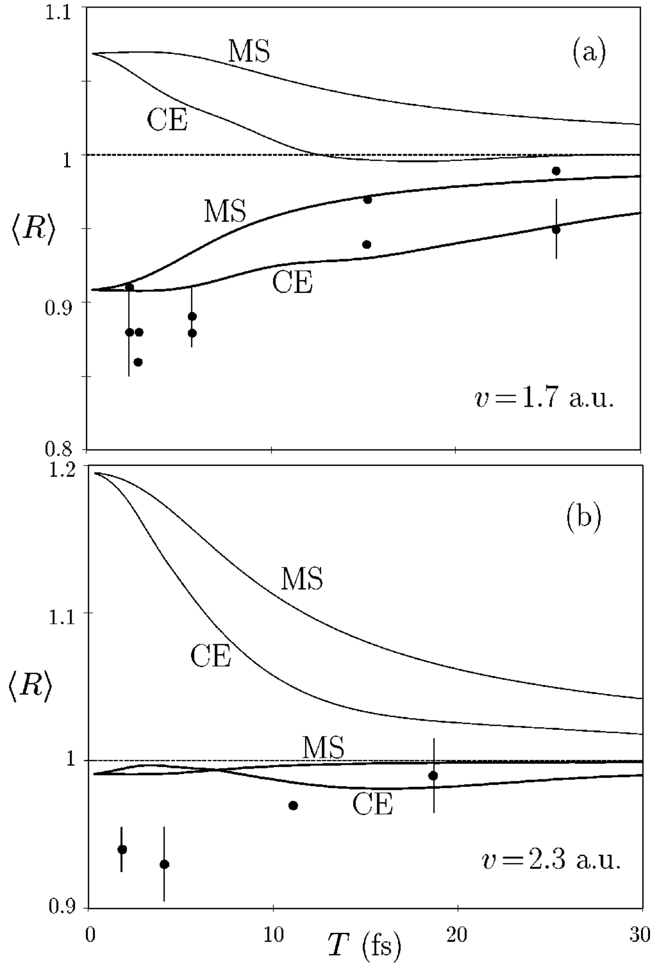

FIG. 7. Averaged stopping-power ratio $\langle R\rangle=\langle S_{cl}\rangle/S_0$ of N_2 in carbon versus the penetration time T (in fs) for two speeds: (a) $v=1.7$ and (b) $v=2.3$ (in a.u.). Thin solid lines are obtained with the frozen ion charges q_0, while the thick solid lines show the role of the vicinage effect on ion charges obtained from Yukawa-type interaction. Cluster dispersions due to Coulomb explosion (CE) and multiple scattering (MS) are treated separately. Experimental points are taken from Ref. [21].

V. CONCLUDING REMARKS

We have presented a consistent variational approach to the vicinage effect on ion charges in a heteronuclear *n*-component cluster of partially stripped ions, which move at the velocity **v** through a solid target. The model is based on the BK theory [27] for isolated ions, supplemented by dynamically screened interionic interactions in the cluster frame of reference. A systematic solution of a system of algebraic equations provides the ion charges which depend on the instantaneous structure of the cluster constituent ions. An important conclusion follows that the dynamics of Coulomb explosion, in general, is no longer governed by pairwise interionic interactions. For a homonuclear di-cluster, the ion charges have been evaluated by means of a Yukawa-like interaction, which gives rise to a charge reduction compared to the charge of isolated ions at the same speed, while a dynamically screened interaction, exhibiting a wake pattern, gives rise to reduced or increased ion charges, in the regions where the interaction potential takes positive or negative values. Based on these results, the ion charge distributions were evaluated from the BK model and used to calculate the cluster stopping power, employing the recent model of Arista [15]. It has been demonstrated that the vicinage effect on ion charges is strong enough to significantly alter the vicinage effect on cluster stopping power, giving rise to additional energy loss enhancement or overall energy loss deenhancement, depending on the model interaction used and the ranges of cluster speeds and internuclear distances. In addition, the vicinage effect on ion charges has been shown to soften the effective interionic potentials, resulting in a slower Coulomb explosion than expected for a pair of ions with the charges frozen at the value corresponding to isolated ions. Finally, we have demonstrated that the energy loss of N_2 in a carbon foil exhibits a deenhancement, compared to the energy loss of two isolated N ions at the same speed, when either Coulomb explosion or a multiple scattering mechanism of cluster dispersion is considered along with the ion charges determined by the Yukawa-type interaction.

Our intention was to demonstrate a variety of possible effects, rather than giving definite answers, regarding the stopping power and Coulomb explosion, when the vicinage effect on ion charges is treated in a consistent manner. A number of improvements of the present theory are possible and required. First, the variational approach to the determination of equilibrium charges on the ions within a cluster can be improved by incorporating the properties of the target [28]. Next, the problem of modeling the dynamically screened interactions, responsible for the vicinage effect on ion charges and Coulomb explosion, needs to be clarified, especially in view of a possible role of core-electron excitations of the target [32,33]. Also, the dynamics of Coulomb explosion needs to be carefully reexamined, in view the complexity of interionic interactions, when the ion charges depend on internuclear positions. At this stage, however, one may suggest a reaffirmation of the role of multiple scattering, based on a softening of the repulsive interionic interactions due to the ion-charge reduction and a consequent slowing down of Coulomb explosion. In addressing these issues, one would greatly benefit from a more abundant pool of systematic experimental data.

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