Vicinage effect on the charge state of fast clusters in solids

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We present a theoretical model for the recent experimental finding by Brunelle *et al.* [Phys. Rev. A **59**, 4456 (1999)] of a reduction of average charge per ion in fast C_n clusters traversing thin carbon foils. This vicinage effect, due to the interaction of the electrons bound at each constituent ion with the neighboring ions, is described by the linear-response formalism, coupled with a statistical description of the cluster structure and the Brandt-Kitagawa variational theory for the bound electrons. The results obtained agree well with the experimental data.

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The interaction of correlated ions with the electron gas in solid targets has remained the focus of research since the early experimental [1] and theoretical [2,3] studies. It is now well documented experimentally [4] that the energy losses of fast atomic clusters, due to electron excitations in solids, are strongly influenced by the interference resulting from the spatial correlation among the cluster constituent particles. This so-called vicinage effect on the electronic stopping of clusters has been described theoretically by Arista and coworkers [2,5], who argued that the linear-response formalism [6] may be applied when cluster speeds exceed Bohr velocity, and used statistical models for the structure of randomly-oriented clusters.

Since fast atomic projectiles are efficiently stripped of their electrons soon upon entering the solid target, a cluster generally experiences the Coulomb explosion due to the dynamically screened interaction between its constituent ions, which was utilized to study the spatial arrangement of small clusters after traversing thin foils [7]. However, the evolution of the structure of large clusters under the Coulomb explosion still remains an open problem, with a potentially significant impact on the studies of cluster energy losses in solids and in fusion plasmas [8]. In this context, Nardi and Zinamon [9] have used a molecular-dynamic simulation to describe the Coulomb explosion of fast C_{60} clusters, assuming a Yukawa-type interionic interaction. We have studied recently [10,11] aspects of Coulomb explosion of large clusters by means of the concept of vicinage self-energy [12], and found that the wake pattern in the dynamical polarization of the electron gas may slow down, or even hinder, the Coulomb explosion for certain ranges of cluster sizes and speeds.

In the course of cluster penetration through a thin foil, the structure of a fast cluster is spatially dispersed due to multiple elastic collisions of each cluster constituent ion with the individual target atoms. The evolution of the cluster structure under the simultaneous action of the Coulomb explosion *and* the multiple scattering presents a difficult challenge, which was investigated by Sigmund [13] for small clusters, with an assumption of unscreened Coulomb interaction. Having in mind a possible stabilization of large clusters against Coulomb explosion due to the wake pattern, we have demonstrated that the multiple scattering plays an important role in modifying the vicinage effect on cluster stopping power [11].

So far, in theoretical studies of cluster interactions with solid targets, the cluster constituent ions have been treated as point charges, with the ion charge Q being an independent parameter, which may be obtained from empirical data for single ions in solids [3,14]. However, it has been reported very recently by Brunelle *et al.* [15] that the average charge of each cluster constituent ion is significantly *reduced* as compared to that of a single ion at the same velocity, for carbon clusters C_n ($3 \leqslant n \leqslant 10$) exiting thin carbon foils in the energy range from 1 to 4 MeV per atom. The mechanism responsible for this 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.

Here we develop a simple theoretical model for the experimental findings of Brunelle *et al.* by treating the vicinage effect along lines similar to those developed in cluster self-energy studies [10,11]. In order to handle the ion charge states, we have to adopt a localized model for the electron structure on each constituent ion that is capable of reflecting the stabilizing role of the vicinage potential due to the neighboring ions. Computations can be kept tractable by means of the simple but robust variational model for the electron structure of isolated ions traversing solid targets, developed by Brandt and Kitagawa (BK) [16]. The BK theory, which is one of the most efficient methods for dealing with electronic stopping of *heavy* ions in solids [14,17], focuses on modifications of the linear-response formalism [6] for the stopping power of a *single* ion, by taking into account the distribution of the bound electrons in a version of the statistical model

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with exponential screening. The *equilibrium* charge state of the ion is obtained in the BK theory by a variational procedure [16], coupled with the so-called energy stripping criterion [17]. We use the variational aspects of BK theory for an ion within the cluster, supplemented by the vicinage potential of neighboring ions, to deduce the average charge Q_n per ion in an n-component cluster. Atomic units are used throughout, unless otherwise indicated.

We consider a homonuclear cluster of n ions with atomic number Z, traveling at a constant velocity $\mathbf{v}$, with the ions placed at the positions $\mathbf{r}_j$, $1 \leqslant j \leqslant n$, relative to the center of mass of the cluster. We assume that the electron density $\rho_e(\mathbf{r};N,\Lambda)$, centered at each ion in the cluster, is a radially symmetric function normalized to the number N of bound electrons, and parametrized by the variational size Λ of the ion. The vicinage energy of the interaction of the electrons bound at the ion j with the remaining $(n-1)$ ions in the cluster is

$$E_{vic}^{j} = -\int d\mathbf{r}\, \rho_e(\mathbf{r}-\mathbf{r}_j-\mathbf{v}t;N_j,\Lambda_j)\Phi(\mathbf{r},t), \tag{1}$$

with the Fourier transform of the potential of the remaining cluster ions given by the solution of the Poisson equation, $\Phi(\mathbf{k},\omega) = (4\pi/k^2)\rho(\mathbf{k},\omega)/\epsilon(k,\omega)$, where $\epsilon(k,\omega)$ is the dielectric function of the electron gas and $\rho(\mathbf{k},\omega)$ is the Fourier transform of the total charge density of the remaining ions

$$\rho(\mathbf{r},t) = \sum_{l\neq j}^{n} [Z\delta(\mathbf{r}-\mathbf{r}_l-\mathbf{v}t) - \rho_e(\mathbf{r}-\mathbf{r}_l-\mathbf{v}t;N_l,\Lambda_l)].$$

At this point, the problem may be made tractable if we assume that all the ion charges are equal within the cluster, i.e., $N_j = N$ and, consequently, $\Lambda_j = \Lambda$ for all j, so that the average charge per ion is $Q = Z - N$. Then, the average vicinage energy per ion $E_{vic}(N,\Lambda;D) \equiv \langle E_{vic}^{j} \rangle$ is

$$E_{vic}(N,\Lambda;D) = -(n-1)\int \frac{d\mathbf{k}}{(2\pi)^3}\frac{4\pi}{k^2}\rho_e(k;N,\Lambda) \times [Z-\rho_e(k;N,\Lambda)]\frac{F_2(\mathbf{k};D)}{\epsilon(k,\mathbf{k}\cdot\mathbf{v})}, \tag{2}$$

where $F_2(\mathbf{k};D) = \int\int d\mathbf{r}\, d\mathbf{r}'\, e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}F_2(\mathbf{r},\mathbf{r}';D)$ is the Fourier transform of the two-particle distribution function $F_2(\mathbf{r},\mathbf{r}';D)$ of the cluster constituent ions normalized to unity: $F_2(\mathbf{k}=\mathbf{0};D)=1$. Here, D is the distance the cluster has traversed through the target.

After traversing a very short target thickness D, of the order of 1 μg/cm^2, the ion charge states are already equilibrated, but the cluster structure is still very close to the initial structure of the incident cluster, because both the multiple scattering [18] and the Coulomb explosion [19] have negligible effects on cluster dispersion in the initial stages of penetration through the target. In this limit, we use the initial two-particle distribution function $F_2^0(\mathbf{r},\mathbf{r}') \equiv F_2(\mathbf{r},\mathbf{r}';D=0)$, corresponding to the structure of a randomly oriented homogeneous cluster [5,10], with $F_2^0(\mathbf{r},\mathbf{r}') = F_1^0(r)F_1^0(r')G_2^0(|\mathbf{r}-\mathbf{r}'|)$, where $F_1^0(r)$ is the normalized one-particle distribution function of ion positions in the cluster center-of-mass frame of reference, while the pair distribution function G_2^0 may be modeled by $G_2^0(r) = C\Theta(r-d_{ex})$ [5]. Here, C is a constant determined by the normalization of F_2^0, d_{ex} is the exclusion distance for interionic spacing within the cluster, and Θ is the Heaviside step function. We adopt the spherical-ball model of the cluster [5,10], with $F_1^0(r) = (4\pi r_c^3/3)^{-1}\Theta(r_c - r)$, where r_c is the cluster radius which scales with the exclusion distance approximately as $r_c \simeq (d_{ex}/2)n^{1/3}$.

Regarding the evolution of the cluster structure with increasing penetration depth D, we assume that the multiple scattering at the target atoms is the dominant mechanism, as compared to the Coulomb explosion. This may be rationalized by a possible slowing down of the Coulomb explosion, due to the stabilizing effect of the wake pattern in the interionic interaction [10,11], or by the fact that the Coulomb explosion, under the dynamically screened Yukawa-type interaction, quickly enters the ballistic regime, after traveling very short distances in the target [9]. Of course, the experimental finding [15] of the charge state reduction, due to the vicinage effect itself reduces the intensity of the Coulomb explosion to some extent, while the multiple scattering remains unaffected by this charge reduction.

Multiple scattering of a *single* fast ion proceeds through a random sequence of small-angle deflections, with a negligible change in the ion velocity $\mathbf{v}$. Under the assumption of small-angle elastic scattering at target atoms, the two-dimensional Fourier transform $H(\mathbf{K};D)$ of the distribution of ion displacements in a plane perpendicular to $\mathbf{v}$, at the depth D, may be obtained in a closed form [11,20], which satisfies the normalization condition $H(\mathbf{K}=\mathbf{0};D) = H(\mathbf{K};D=0) = 1$. Statistical independence of the multiple scattering of each constituent ion of the cluster then allows the two-particle distribution function to be written as $F_2(\mathbf{k};D) = |H(\mathbf{K};D)|^2 F_2^0(k)$ [11]. Using an inverse-square radially symmetric scattering potential [14,20] further yields a simple yet realistic result, $H(K;D) = \exp(-\alpha D^2 K)$, where α is a constant [11].

It may be shown from the procedure outlined below that the ion size parameter Λ is by far the shortest length scale in the vicinage energy (2), so that the Fourier transform of the electron density may be approximated by its $k \to 0$ limit, i.e., $\rho_e(k;N,\Lambda) \simeq N$. This gives the point-charge approximation for the average vicinage energy (2) as $E_{vic}(N;D) \simeq -N(Z-N)(n-1)\tilde{E}_v(D)$, where

$$\tilde{E}_v(D) = \frac{2}{\pi v}\int_0^\infty \frac{dk}{k}F_2^0(k)\int_0^{kv} d\omega\, H^2(\sqrt{k^2-\omega^2/v^2};D) \times \mathrm{Re}\left[\frac{1}{\epsilon(k,\omega)}\right] \tag{3}$$

is a generalization of the reduced vicinage self-energy of a cluster of unit point charges, in the notation of [10]. Note that $\tilde{E}_v$ describes, in an average manner, the effective interaction potential between a pair of cluster constituent ions,

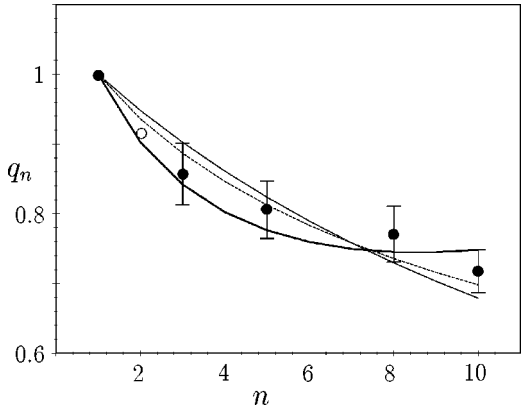

FIG. 1. Charge state ratio $q_n = Q_n/Q_1$ versus the number n of constituent ions in C_n clusters with the energy 2 MeV/atom passing through the carbon foil of thickness 2.2 μg/cm^2. Points with the error bars are experimental data from [15]. Theoretical results are shown for PLA (thick solid line, $d_{ex}=1.9$ a.u.), Yukawa (dashed line, $d_{ex}=3.8$ a.u.) and Coulomb (thin solid line, $d_{ex}=18$ a.u.) models.

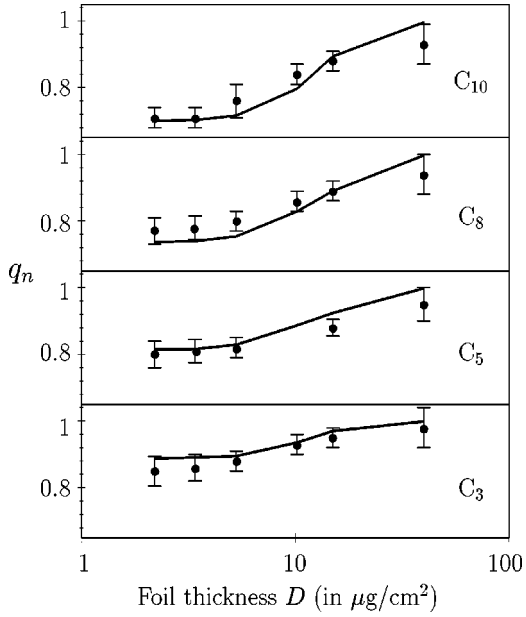

FIG. 2. Charge state ratio $q_n = Q_n/Q_1$ versus the foil thickness D (in μg/cm^2) for the 2 MeV/atom clusters C_n with $n=3$, 5, 8, and 10. Points with the error bars are experimental data from [15], while the theoretical results are shown by the thick solid lines.

with appropriate weighting of the near and distant neighbors. It should be also noted that the point-charge approximation (3) is justified for the vicinage energy of heavy ions, where only the long-ranged interactions, as compared to the ion size, are important, while the stopping power calculations for heavy ions require both the distant and close interactions. Since the dependence of $\tilde{E}_v$ on cluster structure is determined by the model used for the dielectric function $\epsilon(k,\omega)$, several cases are considered. First, we use the plasmon-pole approximation (PLA) [6] for the dielectric function, which exhibits a wake pattern in the interionic interaction within the cluster. Next, we model the dynamically screened Yukawa-type interaction between the ions by using $\mathrm{Re}[1/\epsilon(k,\omega)] \simeq k^2/(k^2+k_s^2)$, with a velocity-dependent screening length k_s^{-1} [21]. Finally, the bare Coulomb interaction is obtained by taking $\epsilon(k,\omega)=1$.

We now turn to the determination of the structure of electrons bound at an ion within the cluster. In the frame of reference moving with the cluster, the total electron energy at each ion consists of the energy arising from the interactions localized at the isolated ion, plus the vicinage energy due to the interactions with the neighboring ions, that is, $E_{tot}(N,\Lambda)=E_{iso}(N,\Lambda)+E_{vic}(N;D)$. In the BK theory [16], the variational electron density is modeled by $\rho_e(\mathbf{r};N,\Lambda) = N\exp(-r/\Lambda)/(4\pi\Lambda^2 r)$, and the electron energy of an isolated ion is obtained as $E_{iso}(N,\Lambda)=aN^{5/3}/\Lambda^2+(\lambda/4)N^2/\Lambda - ZN/\Lambda$, where the three terms represent, respectively, the electron kinetic energy with $a=0.24$, the electron-electron interaction weighted for correlation in an average manner by the variational parameter λ, and the electron-nucleus interaction. Following the variational procedure of BK [16], the condition $\partial E_{tot}/\partial\Lambda=0$ gives the ion size $\Lambda(N) = (2a/Z^{1/3})\mu^{2/3}/(1-\lambda\mu/4)$, where $\mu \equiv N/Z$, while the parameter λ is obtained from the requirement that $\partial E_{tot}/\partial N = 0$ when $N=Z$, that is, when the cluster is neutral. We obtain $\lambda=(4/7)(4-3\sqrt{1+\Delta})$, where $\Delta=(28/3)(a/Z^{1/3})(n-1)\tilde{E}_v(D)$, with $\tilde{E}_v(D)$ given by Eq. (3). The final step in the BK theory is to obtain the velocity-dependent average number N of the bound electrons from the energy stripping criterion, where the total energy of bound electrons in the laboratory frame of reference, $E_{lab}(N)=Nv^2/2 + E_{iso}(N,\Lambda(N))+E_{vic}(N;D)$, is minimal [17]. The condition $\partial E_{lab}/\partial N=0$ yields the equation for $\mu=N/Z$, namely,

$$2a\left(\frac{v}{Z^{2/3}}\right)^2 = \frac{(1-\lambda\mu/4)(1-7\lambda\mu/4)}{3\mu^{2/3}} + \frac{3}{7}(1-2\mu)\Delta, \tag{4}$$

which is easily solved, resulting in an average charge $Q_n = Z(1-\mu)$ per ion in an n-component cluster.

One set of experimental results by Brunelle *et al.* [15] deals with a very thin carbon foil of thickness 2.2 μg/cm^2, and provides the data for the average charge state *ratio* $q_n = Q_n/Q_1$, where Q_1 is the average charge state of a single ion, for C_n clusters with the energy 2 MeV/atom. For such a thin foil, the dispersion of the cluster ions is relatively small, so that the cluster structure may be considered close to its initial structure at the entrance to the foil, with the estimated value of the exclusion distance of about $d_{ex}\simeq 3.8$ [15]. At the present level of understanding of the initial transient stages of the cluster breakup in the target, it is difficult to make a more reliable estimate of d_{ex}. Thus, we neglect the multiple scattering effects by setting $D=0$ in Eq. (3) and treat d_{ex} in the initial two-particle distribution F_2^0 as a free parameter, testing its values against various models for the dielectric function. In Fig. 1 we present the experimental points from [15] for q_n against the number of cluster constituents n, together with the results of our calculations for the PLA model (thick solid curve), Yukawa-type interaction (dashed curve), and the bare Coulomb interaction (thin solid curve). As

pointed out in [15], q_n is a decreasing function of n, regardless of the detailed cluster structure, in support of our spherical-ball model. In order to make the best fit to the experimental data for each model, we chose $d_{ex}=1.9$ for the PLA model, $d_{ex}=3.8$ for the Yukawa model, and $d_{ex}=18$ for the bare Coulomb case. It is clear that the required values of d_{ex} for the PLA and Yukawa models are within a realistic expectation of the exclusion distance at the exit from a very thin foil, while the bare Coulomb interaction may be ruled out. Whether the Yukawa interaction suffices, or the PLA model should be used, may be clarified when the data on the actual distribution of charges per ion, preferably in larger clusters, become available. In the following, we use the Yukawa model, because it is simpler and it gives the results for q_n in Fig. 1, which are less sensitive to variations in d_{ex} than the PLA model.

Another set of experimental data by Brunelle *et al.* [15] follows the dependence of the average reduced charge q_n per ion on the carbon foil thickness D in the range 2.2 to 40 μg/cm^2, for several C_n clusters with energy of 2 MeV/atom. We use the Yukawa model in Eq. (3), and take the cluster structure to be dispersed by the multiple scattering on the target atoms, starting from the initial spherical-ball cluster model with $d_{ex}=3.8$. In Fig. 2 we show the experimental points for the C_n clusters with $n=3$, 5, 8, and 10, together with the theoretical results, represented by the solid lines. As expected, the weakening of the vicinage effect, due to the dispersion of the interionic distances within the cluster during target penetration, leads to a weakening of the charge-state reduction with the increasing foil thickness D, such that q_n slowly approaches unity in thick foils, for all n. Given the simplicity of the present theory and the fact that no free parameters were used, the agreement with the experiment is surprisingly good.

In conclusion, we have developed a composite, but self-consistent, theoretical model for the vicinage effect on the average charge state per ion in fast clusters traversing thin solid foils. Reduction of ion charges is obtained from the variational-statistical approach of the BK theory [16,17] as a result of stronger binding of electrons in an ion due to the vicinage interaction with the neighboring ions, which is equivalent to the enhanced electron capture probabilities in the kinetic-equation-based phenomenological approach of Maor *et al.* [22]. Our theory is essentially parameter-free and the calculations are easily tractable, requiring at most one numerical integration. The results obtained agree well with the experimental data [15] for ion charges in C_n clusters traversing a very thin foil, if the appropriate dynamical polarization of the electron gas in the solid is taken into account, together with the realistic values of interionic distances within the cluster. Furthermore, we succeeded in reproducing the observed ion charge dependence on increasing foil thickness, based only on the multiple scattering mechanism of the cluster dispersion in the foil. In order to provide further evidence of the domination of the multiple scattering over the Coulomb explosion, a detailed molecular dynamics simulation including both these processes should be performed. However, such a study becomes complicated by the need to couple the equations of motion for cluster constituent ions with a self-consistent solution for their charges, in order to take into account the vicinage effect in large clusters.

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