

Ion-charge distribution in fast, partially stripped clusters passing thin foilsZ. L. Mišković,¹ W.-K. Liu,² F. O. Goodman,¹ and Y.-N. Wang³¹*Department of Applied Mathematics, University of Waterloo, Waterloo, Ontario, Canada N2L 3G1*²*Department of Physics, University of Waterloo, Waterloo, Ontario, Canada N2L 3G1*³*The State Key Laboratory for Materials Modification by Laser, Ion and Electron Beams, Dalian University of Technology, Dalian 116023, People's Republic of China*

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We provide a self-consistent theoretical model describing the vicinage effect on charge states of partially stripped ions in a large cluster traversing a thin solid foil at a high speed. Starting from Brandt-Kitagawa variational theory for the effective charge on a fast heavy ion, supplemented by the dynamically screened interionic interactions within the cluster, we obtain a nonlinear integral equation for a function describing the spatial distribution of ion charges throughout the cluster. Numerical solutions of such an equation show that the ions in the cluster interior become increasingly neutralized as the cluster size grows.

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Stopping of fast heavy ions (with atomic numbers $Z \geq 3$) is still an open problem that continues to attract theoretical attention [1], owing to the fact that such ions are only partially stripped of their valence electrons when moving through solid targets at speeds in excess of the Bohr speed. A statistical model for ion screening by the remaining bound electrons was proposed by Brandt and Kitagawa (BK) some twenty years ago [2], has been used with confidence ever since [3], and continues to play a central role in modern developments of the theory of heavy-ion stopping [1,4].

On the other hand, stopping of atomic clusters in solids is another problem with rich history, which has been reviewed recently [5,6]. Interest in cluster stopping originates from a possibility that the energy loss per constituent ion in a cluster may be enhanced, compared to the energy loss of an isolated ion at the same speed, due to close spatial correlation among the cluster constituents, which is concisely described as the vicinage effect. While this enhancement has been documented for fast hydrogen clusters traversing thin foils, the data on clusters made of heavier atoms, such as C_n and B_n , generally exhibit rather weak vicinage effect on the cluster stopping power [5,6], or even a reduction of the energy loss per ion, as in the case of N_2 [7].

The main cause of this suppression, or even reversal, of the energy-loss enhancement for heavy-atom clusters lies in the fact that the charge state of each ion is influenced by the vicinity of other ions in the cluster. Namely, experimental measurements on fast N_2 molecules [8] and C_n clusters [9] 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 [8,9], showing that the charge-state reduction diminishes with increasing penetration time on a time-scale characteristic of cluster dispersion due to Coulomb explosion and/or multiple elastic scattering of cluster constituents on target atoms.

The vicinage effect on the charge state of partially stripped ions in clusters of arbitrary size has been described recently by a theoretical model [10] that combines the self-energy calculations for atomic clusters in solids [11,12] with the variational aspects of the BK model [2,4] for the electronic structure of isolated heavy ions moving through solids. Our model [10] has successfully described recent experimental data [9] for the dependence of average ion charge on the number n of constituent particles in carbon clusters C_n ($3 \leq n \leq 10$), as well as the dependence of this charge on the target thickness, based on cluster dispersion due to multiple scattering [13]. Moreover, we have combined our report [10] with Arista's model [6] for stopping power of heavy-atom clusters, in order to study the complicated interplay between the vicinage effects on the ion-charge states and on the cluster stopping power, which is responsible for the reduction or even reversal of the above-mentioned energy-loss enhancement. For example, experimental data on charge-state reduction of N_2 constituents in carbon foils [8] have been reproduced by our report [14], leading to a successful resolution [14,15] of the long-standing puzzle of reduced energy losses of fast N_2 molecules in carbon foils [7]. Finally, it should be stressed that the vicinage effect on ion charges is expected to influence the dynamics of Coulomb explosion of heavy-ion clusters in a significant way, as shown in [15].

In previous implementations of our model [10,15], we have obtained the average charge per ion based on the assumption that all the constituent ions carry equal charges throughout a randomly oriented cluster, which facilitates a solution of a system of coupled algebraic equations for individual ion charges. We wish to investigate in the present report the spatial inhomogeneities, if any, of the distribution of ion charges throughout relatively large clusters. This will be achieved by introducing joint charge-state-configuration-space distribution functions for the cluster constituent ions, allowing a translation of the system of algebraic equations into a single integral equation for a function describing the spatial distribution of ion charges throughout a large randomly oriented cluster. This integral equation will be solved numerically, with the interionic interaction modeled by a dynamically screened Yukawa-type potential, for the case of

fast sodium clusters traversing a thin foil. Atomic units are used throughout unless otherwise indicated.

We consider an n -component homonuclear 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 be the atomic number of the constituent ions and N_j the number of bound electrons at the j th ion ($1 \leq j \leq n$), which is placed at the position \mathbf{r}_j in the cluster center-of-mass frame of reference. Since the typical ion sizes are much smaller than the typical interionic distances $|\mathbf{r}_l - \mathbf{r}_j|$, one may apply the point-charge approximation for the interaction energy among distinct ion pairs in the cluster [10,15]. Thus, the total electronic energy of the cluster 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 the bound electrons in isolated ions, and the interionic interaction energy, viz.,

$$E_L = \sum_{j=1}^n N_j \frac{v^2}{2} + \sum_{j=1}^n E_{iso}(N_j) + \frac{1}{2} \sum_{j=1}^n \sum_{l \neq j}^n (Z - N_j)(Z - N_l) U(\mathbf{r}_{jl}). \quad (1)$$

Here, the energy of the isolated ion with N -bound electrons is taken from the BK model [2],

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

with $a = 0.24$, while the dynamically screened interaction potential among the two-unit point charges at the relative position $\mathbf{r}_{jl} = \mathbf{r}_l - \mathbf{r}_j$ is defined by

$$U(\mathbf{r}) = \int \frac{d^3 \mathbf{k}}{(2\pi)^3} \frac{4\pi}{k^2} \frac{e^{i\mathbf{k} \cdot \mathbf{r}}}{\epsilon(k, \mathbf{k} \cdot \mathbf{v})}, \quad (3)$$

with $\epsilon(k, \omega)$ being the dielectric function of the medium. We further follow the variational approach of the BK theory [2] and apply the so-called energy stripping criterion [4], 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 achieved by solving n coupled equations $\partial E_L / \partial N_j = 0$ [10,15],

$$\frac{v^2}{2} + E'_{iso}(N_j) - \sum_{j \neq l=1}^n (Z - N_l) U(\mathbf{r}_{jl}) = 0, \quad \text{for } 1 \leq j \leq n. \quad (4)$$

Note that the self consistency of this procedure is expressed by the fact that the solution for the charge $Q_j \equiv Z - N_j$ on each ion ($1 \leq j \leq n$) becomes a function of the positions of all the ions in the cluster: $Q_j = Q_j(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_n)$.

Since the solution of Eqs. (4) becomes prohibitive for large clusters, we introduce the joint one-particle probability

distribution function $F_1(Q; \mathbf{r})$ for an ion to have charge Q and be placed at the position \mathbf{r} in the cluster by

$$F_1(Q; \mathbf{r}) = \left\langle \frac{1}{n} \sum_{j=1}^n \delta(Q - Q_j) \delta(\mathbf{r} - \mathbf{r}_j) \right\rangle \approx \delta(Q - q(\mathbf{r})) f_1(\mathbf{r}), \quad (5)$$

where the average is taken over an ensemble of all the cluster configurations in a beam of randomly oriented clusters, while $f_1(\mathbf{r})$ is the standard one-particle distribution function for the ion positions. Our goal is to determine, in a self-consistent manner, the function $q(\mathbf{r})$ describing the spatial distribution of ion charges throughout a cluster of a given structure in the configuration space. Since the pair correlation among the ion charges in this approach is a consequence of the correlation among the ion positions in the configuration space, we may write for the joint two-particle probability distribution

$$F_2(Q, Q'; \mathbf{r}, \mathbf{r}') = \left\langle \frac{1}{n(n-1)} \sum_{j=1}^n \sum_{l \neq j}^n \delta(Q - Q_j) \times \delta(Q' - Q_l) \delta(\mathbf{r} - \mathbf{r}_j) \delta(\mathbf{r}' - \mathbf{r}_l) \right\rangle \approx \delta[Q - q(\mathbf{r})] \delta[Q' - q(\mathbf{r}')] f_2(\mathbf{r}, \mathbf{r}'). \quad (6)$$

Here, the two-particle distribution function in the configuration space may be factorized in the usual way [6,12] as $f_2(\mathbf{r}, \mathbf{r}') = f_1(\mathbf{r}) f_1(\mathbf{r}') g_2(|\mathbf{r} - \mathbf{r}'|)$, with g_2 being related to the familiar pair-correlation function [6]. Note that the distribution functions $f_1(\mathbf{r})$ and $f_2(\mathbf{r}, \mathbf{r}')$ are chosen to be each normalized to unity, in order to emphasize the role of the cluster size n in the following.

With such a statistical approach, it is possible to use Eq. (1) to express the ensemble average of the total electron energy per ion in the laboratory frame, $\mathcal{E}_L \equiv \langle E_L / n \rangle$, as follows:

$$\begin{aligned} \mathcal{E}_L[q(\mathbf{r})] &= \frac{v^2}{2} \int d^3 \mathbf{r} [Z - q(\mathbf{r})] f_1(\mathbf{r}) \\ &+ \int d^3 \mathbf{r} E_{iso}[Z - q(\mathbf{r})] f_1(\mathbf{r}) + \frac{1}{2} (n-1) \\ &\times \int \int d^3 \mathbf{r} d^3 \mathbf{r}' q(\mathbf{r}) q(\mathbf{r}') U(\mathbf{r} - \mathbf{r}') f_2(\mathbf{r}, \mathbf{r}'). \end{aligned} \quad (7)$$

In the spirit of the above variational approach, we minimize the functional $\mathcal{E}_L[q(\mathbf{r})]$ by taking the functional derivative and solving the equation $\delta \mathcal{E}_L / \delta q(\mathbf{r}) = 0$, which gives the nonlinear integral equation for the unknown function $q(\mathbf{r})$,

$$\begin{aligned} -\frac{v^2}{2} + E'_0[q(\mathbf{r})] + (n-1) \int d^3 \mathbf{r}' q(\mathbf{r}') f_1(\mathbf{r}') \\ \times g_2(|\mathbf{r} - \mathbf{r}'|) U(\mathbf{r} - \mathbf{r}') = 0, \end{aligned} \quad (8)$$

where $E'_0(q)$ is the derivative of the function $E_0(q) \equiv E_{iso}(Z-q)$ for $q > 0$, while we take $E'_0(q) = 0$ for $q < 0$ to prevent the formation of negative ions. Equation (8) is our main result, which replaces the system of Eqs. (4) in the statistical approach. We note that setting $n=1$ in Eq. (8) gives an algebraic equation for the velocity-dependent effective charge Q_0 on an isolated ion [4].

In general, the integral Eq. (8) contains complete information on the cluster structure through the distribution functions f_1 and g_2 , as well as the details of the dynamical screening of the interaction potential via Eq. (3). Although this interaction may exhibit strong anisotropy due to wake effects [16], even for randomly oriented clusters, we adopt here an approach that is more popular in the studies of Coulomb explosion of clusters, that is, we model the interionic interaction potential by a Yukawa-like form $U(r) = r^{-1} \exp(-k_s r)$, where k_s^{-1} is a velocity-dependent screening length [17], with the high-velocity limit $k_s^{-1} \approx v/\omega_p$, ω_p being the electron plasma frequency of the target. Moreover, assuming a randomly oriented cluster, one may use a simple spherical-ball model of a homogeneous cluster [12,18], such that $f_1(\mathbf{r}) \equiv f_1(r) = (4\pi r_{cl}^3/3)^{-1} \Theta(r_{cl} - r)$, where Θ is the unit step function and $r_{cl} \approx (d_{ex}/2)n^{1/3}$ is the cluster radius with d_{ex} being the exclusion distance among the cluster constituent ions, while the pair correlation is described by $g_2(r) = C\Theta(r - d_{ex})$, where the constant C fixes the normalization [18]. With such assumptions, the unknown function q in Eq. (8) is rendered dependent only on the radial distance r from the center of the cluster, so that the integral Eq. (8) reduces to

$$-\frac{v^2}{2} + E'_0[q(r)] + \frac{3(n-1)C}{2k_s r_{cl}^3} \int_0^{r_{cl}} dr' r' q(r') [e^{-k_s(r'+r)} - e^{-k_s\rho(|r'-r|)}] \Theta(r'+r-\rho(|r'-r|)) = 0, \quad (9)$$

where $\rho(|r'-r|) \equiv \max(d_{ex}, |r'-r|)$.

Clearly, Eq. (9) has to be solved numerically. In our procedure, Eq. (9) was discretized into a matrix equation of the form $-v^2/2 + E'_0(q_\mu) + \sum_\nu K_{\mu\nu} q_\nu = 0$ and solved by successive iterations, governed by $-v^2/2 + E'_0(q_\mu^{(i+1)}) + \sum_\nu K_{\mu\nu} q_\nu^{(i)} = 0$. Here, matrix $K_{\mu\nu}$ replaces the kernel of the integral term in Eq. (9), while $q_\mu^{(i)}$ stands for the i th iteration of the vector q_μ replacing the function $q(r)$. Owing to the smoothness of the kernel in Eq. (9) and the monotonicity of the function $E'_0(q)$, a stable and unique solution for q was obtained after about ten iterations with the size of discretization being 20, regardless of the initial iteration $q_\mu^{(0)}$, with the most practical choice being $q_\mu^{(0)} = Q_0$.

We have solved Eq. (9) for sodium clusters ($d_{ex} = 6.9$), with the sizes up to $n=200$, which move through an aluminum target at the speed $v=3$. Although the choice of the parameters does not necessarily fall into the range of experimental feasibility, we nevertheless show the results for the reduced charge function $q(r)/Q_0$ versus the reduced radial distance r/r_{cl} in Fig. 1, in order to discuss the qualitative features of the ion-charge distribution. It is obvious that this distribution depends on the ion position in the cluster in such

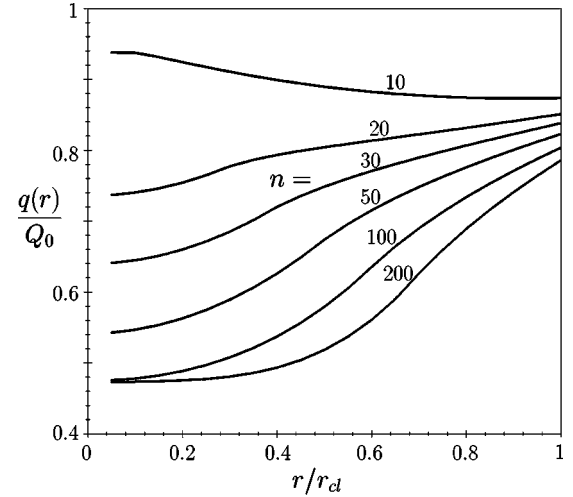


FIG. 1. Reduced distribution of ion charges $q(r)/Q_0$ in Na_n clusters versus the reduced radial distance r/r_{cl} for cluster speed $v=3$ a.u. in an Al foil.

a way that the interior of the cluster tends to become more neutral in larger clusters. Interestingly, there is little variation with the cluster size of charge states for those ions occupying the surface region of the cluster. This then implies that the Coulomb explosion of large clusters would initially peel off the outer regions with higher ion charges, which would eventually raise the charges of the remaining ions emerging on the cluster surface, thus providing a new fuel for bursting off the remaining of the cluster.

It is interesting to speculate about the solution for $n=10$ in Fig. 1, which shows an increase of the charge distribution in the cluster interior, contrary to all other cases of larger clusters that exhibit charge reduction in the interior. Physically, in such small clusters, practically all constituent ions lie on the cluster surface [18], and a spherical-shell model [12] for $f_1(r)$ would be more appropriate. Nevertheless, the $n=10$ solution is not entirely unphysical, since the configuration-space factor f_2 in the joint distribution function F_2 , Eq. (6), will preclude population of the cluster interior by ions in such small clusters. In a more general context, while the function $q(\mathbf{r})$ is defined for all positions \mathbf{r} , within the cluster or even outside the cluster, the actual probability for an ion to occupy a small volume $d^3\mathbf{r}$, centered at a position \mathbf{r} , and to have the charge $Q=q(\mathbf{r})$, is constrained by factors describing the spatial distribution of the constituent ions within the cluster.

Furthermore, the average charge per ion has been evaluated as $\langle Q \rangle = \int d^3\mathbf{r} q(\mathbf{r}) f_1(r)$, and the results are displayed in Fig. 2 for the speeds $v=2, 3, 4$, and 5, in a wide range of cluster sizes n . It is seen that the strong increase of ion charges in small clusters with the speed is attenuated through a steady reduction of those charges with increasing cluster size. Finally, we have tested the validity of the assumption made in [10] that the average charge per ion q_{av} may be approximately obtained by taking all ion charges in the cluster to be equal, and found a very good agreement between q_{av} and $\langle Q \rangle$. We note that q_{av} is obtained in the present context from Eq. (8) by first setting $q(\mathbf{r}) = q(\mathbf{r}') = q_{av}$

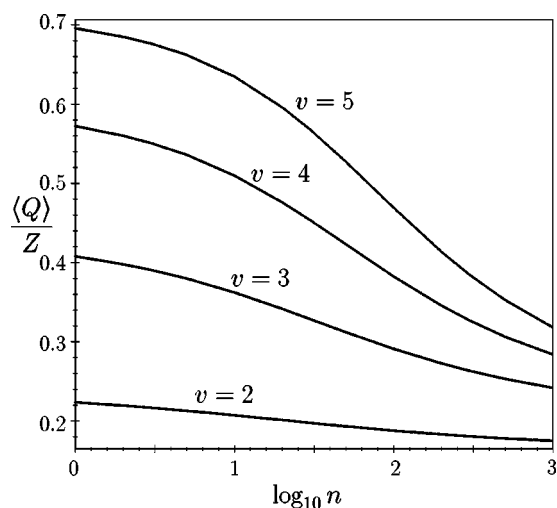


FIG. 2. Ratio of average charge per ion to atomic number, $\langle Q \rangle / Z$, versus the cluster size in Na_n clusters moving at speeds $v = 2, 3, 4$, and 5 a.u. through an Al foil.

= const., then multiplying by $f_1(\mathbf{r})$, and finally integrating over \mathbf{r} , which is fully equivalent to the procedure used in [10].

In conclusion, we have shown that the ion charges will generally decrease as the cluster size increases due to the vicinage effect, but the spatial distribution of those charges throughout the cluster may be quite nonhomogeneous. One may expect that such effects will generally reduce the cluster energy loss and will slow down the Coulomb explosion [15], compared to the picture in which the charge states of the cluster constituent ions are considered frozen. It may be further expected that the nonhomogeneity of the ion-charge distribution will be even more pronounced when an anisotropically screened interaction potential $U(\mathbf{r})$, exhibiting a wake pattern, is used in Eq. (8), thus further accentuating the anisotropy of the Coulomb explosion patterns [19]. We finally note that our goal in future work is to explore possible advantages in using the integral-equation formulation of the problem of ion-charge distribution in calculations of the stopping power of large, heavy-ion clusters.

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