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Feedback action of vicinage effect in cluster stopping via nonhomogeneous charge reduction

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We use a self-consistent, variational-statistical approach to analyze the vicinage effect on charge states of constituent ions in fast large clusters, passing through thin foils. The solutions of the resulting nonlinear integral equation show that the individual ion charges are reduced in comparison to isolated isotachic ions and are distributed nonhomogeneously throughout the cluster. Using a statistical description of the ion positions in conjunction with the nonhomogeneous distribution of ion charges, we evaluate the total stopping power of the cluster by means of the dielectric formalism. It is shown that interference in cluster stopping coming from the spatial correlations among the constituent ions is strongly influenced by the vicinage effect on ion charge states.

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There has been a significant increase in interest over the past decade in penetration of fast atomic or molecular clusters through solid and plasma targets, as evidenced by two recent reviews [1,2]. The progress in this area has been propelled by the advancements in experimental accelerator techniques for clusters [3] on the one hand, and by prospects of further achievements in fusion research and materials modification on the other hand. The main focus of the research is on the effects of spatial correlation among the constituent ions, commonly known as vicinage effects [1], which are manifested in several mutually inter-related phenomena: interference in cluster stopping power, the proximity effect in equilibrium charges of individual ions, and the dynamics of Coulomb explosion of a cluster in the course of penetration through a target. The most interesting effect for potential applications is the possibility of an increase in energy losses due to constructive interferences in cluster stopping [1,2]. On the other hand, both experimental [3] and theoretical [4] studies have shown that the vicinage effect is responsible for a decrease in the average charge state per ion in small carbon clusters, compared to the charge state of an isolated carbon ion moving at the same speed as the cluster. Moreover, it has been shown [5] that such charge reduction, which was observed for N_2 molecules in carbon foils [6], is intimately connected with a reduction, and even reversal, of the interference effects in the stopping power of N_2, which was documented in an earlier measurement [7]. It has also been shown that the charge reduction in N_2 molecules due to the vicinage effects may cause a slowing down of the Coulomb explosion of N_2 in carbon foils [5,8]. Thus, one may speculate about a universal role of the vicinage effect in ion charge states as a source of a negative feedback influence on the vicinage effect in stopping power and Coulomb explosion, at least for clusters made of partially stripped multielectron atoms. The situation is particularly interesting for large and fast clusters, for which the so-called resonant effect [1,9] has been associated with the existence of a maximum in cluster stopping power when the cluster radius r_{cl} satisfies the relation $r_{cl} = 1.8 v/\omega_p$, where v is the cluster speed and ω_p is the plasma frequency of the electron gas in the target.

We have recently developed a self-consistent, variational model for evaluation of the vicinage effects in ion charge distribution throughout large clusters, based on a statistical treatment of the cluster geometric structure [10]. The resulting nonlinear integral equation for the ion charge distribution function has been solved numerically, showing that, in addition to the expected reduction of the average charge per ion, the ion charges are distributed rather nonhomogeneously, such that the ions in the cluster interior have much lower charges than those in the surface region [10]. In this report, we use the nonhomogeneous ion charge distribution obtained from the integral equation to evaluate the stopping power of large clusters passing through thin solid foils at speeds in excess of Bohr's speed. In this way, we are able to explicitly describe the situation where the interference effects in stopping power due to the spatial correlation are coupled with the vicinage effects in the distribution of ion charges, thus providing a quantitative estimate of the negative feedback role of the vicinage effect. Atomic units are used throughout.

Consider a fast n-component homonuclear cluster, entering a thin solid foil with the velocity $\mathbf{v}$, and let $\mathbf{r}_j$ be the positions of the constituent ions, $1 \leqslant j \leqslant n$, in the cluster center-of-mass frame of reference. We assume that the foil is thick enough that the individual ion charge states have reached their respective equilibrium values, but also thin enough to allow us to neglect the changes in cluster geometric structure due to energy loss, Coulomb explosion, or multiple scattering in the target. Although these assumptions are not critical and can be lifted in principle, using the picture of a cluster as bunch of closely spaced ions at fixed positions, each having a fixed charge, will be helpful in understanding complicated interplay between the two aspects of the vicinage effect: one on the ion charge state distribution throughout the cluster, and the other on the interferences in cluster energy losses.

Let Z and N_j be, respectively, the nuclear charge and the number of electrons bound at the jth ion, such that its charge is $Q_j = Z - N_j$. The total charge density of cluster in the laboratory frame, $\rho_L(\mathbf{r},t) \equiv \rho_{cl}(\mathbf{r} - \mathbf{v}t)$, is given in terms of the instantaneous charge density in the cluster frame, $\rho_{cl}(\mathbf{r}) = \sum_{j=1}^{n} \rho_j(\mathbf{r} - \mathbf{r}_j)$, where the total (nuclear and electronic) charge density localized at the jth ion is given by $\rho_j(\mathbf{r}) = Z\delta(\mathbf{r}) - \rho_{e,j}(\mathbf{r})$, with $\rho_{e,j}(\mathbf{r})$ being the density of bound electrons normalized to $\int d^3\mathbf{r}\rho_{e,j}(\mathbf{r}) = N_j$. The total electronic energy of the cluster in the laboratory frame of reference consists of the kinetic energy of all bound electrons due to the cluster motion at speed v, the internal binding energies of the electrons in the individual ions, and the energy of dynamically screened interionic interactions [10],

$$E_L = \sum_{j=1}^{n} N_j \frac{v^2}{2} + \sum_{j=1}^{n} E_j + \frac{1}{2} \sum_{j=1}^{n} \sum_{j \neq l=1}^{n} \int \frac{d^3\mathbf{k}}{(2\pi)^3} \frac{4\pi}{k^2} \frac{\rho_j(\mathbf{k})\rho_l^*(\mathbf{k})}{\epsilon(k,\mathbf{k}\cdot\mathbf{v})} e^{i\mathbf{k}\cdot\mathbf{r}_{jl}}, \tag{1}$$

where $\epsilon(k,\omega)$ is the dielectric function of the medium, $\rho_j(\mathbf{k})$ is the space Fourier transform of $\rho_j(\mathbf{r})$, and $\mathbf{r}_{jl}$ is the relative position vector between the jth and the lth ions, $\mathbf{r}_{jl} \equiv \mathbf{r}_l - \mathbf{r}_j$. In order to determine the binding energy and the electronic structure of an individual ion, we adopt the variational-statistical model of Brandt and Kitagawa (BK) [11]. Thus, both the energy E_j and the electron density $\rho_{e,j}(\mathbf{r})$ of the jth ion are parametrized by the number N_j of electrons bound on that ion, that is, $E_j \equiv E_{BK}(N_j)$ and $\rho_{e,j}(\mathbf{r}) \equiv \rho_{e,BK}(r;N_j)$. Consequently, one may consider the Fourier transform of the total charge density at the jth ion to be parametrized by the net charge on that ion, $Q_j = Z - N_j$, that is, $\rho_j(\mathbf{k}) \equiv \rho_{BK}(k;Q_j)$, with the property $\rho_{BK}(0;Q_j) = Q_j$. Note that E_{BK}, $\rho_{e,BK}$, and ρ_{BK} are universal functions in the BK model [11].

The dynamical equilibrium charge of an *isolated* ion, $Q_0 = Z - N_0$, is obtained in the BK theory by applying the so-called *energy stripping criterion* [12], where the electron energy in the laboratory frame, $Nv^2/2 + E_{BK}(N)$, is minimized relative to N, that is, by solving the equation $v^2/2 + E'_{BK}(N_0) = 0$, where $E'_{BK}(N) \equiv dE_{BK}/dN$. Now, in order to determine the individual equilibrium ion charges throughout the *cluster* in a self-consistent manner, we generalize the energy stripping criterion and minimize the total cluster energy in the laboratory frame E_L, Eq. (1), relative to the electron populations N_j on all ions. This gives n coupled algebraic equations $\partial E_L/\partial N_j = 0$, for $1 \leqslant j \leqslant n$, which completely determine the equilibrium ion charges $Q_j = Z - N_j$ throughout the cluster [10]. These equations are simplified by invoking the point-charge approximation (PCA) for the constituent ions, that is, by setting $\rho_j(\mathbf{k}) \simeq \rho_j(\mathbf{k}=\mathbf{0}) = Z - N_j$ in Eq. (1), which is justified owing to the fact that the typical ion sizes are much smaller than the interionic distances $|\mathbf{r}_{jl}|$ within the cluster.

Even with the PCA, the solution of n coupled equations for individual ion charges Q_j may be a formidable task for large clusters. In that case, one may take advantage of the fact that the cluster beam provides an ensemble of randomly oriented clusters, such that the geometric structure of such clusters may be described by statistical means, using a one-particle distribution function $f_1(\mathbf{r})$ and a two-particle distribution function $f_2(\mathbf{r},\mathbf{r}') = f_1(\mathbf{r}) f_1(\mathbf{r}') g_2(|\mathbf{r}-\mathbf{r}'|)$, where g_2 is the familiar pair-correlation function [1,13]. It has been shown recently [10], that the distribution of individual ion charges throughout the cluster may be described by a single function $q(\mathbf{r})$ of ion position $\mathbf{r}$ in the cluster frame, which is determined by the overall cluster geometric structure. As a result of minimizing the total cluster energy in the laboratory frame, one obtains a nonlinear integral equation for $q(\mathbf{r})$, viz.,

$$v^2/2 + E'_{BK}(Z - q(\mathbf{r})) = (n-1) \int d^3\mathbf{r}' q(\mathbf{r}') f_1(\mathbf{r}') g_2(|\mathbf{r}-\mathbf{r}'|) U(\mathbf{r}-\mathbf{r}'), \tag{2}$$

with

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

being the dynamically screened interaction potential between two unit point charges, where f_1 and f_2 are taken to be each normalized to unity.

We have solved the integral equation Eq. (2) for the ion distribution function $q(\mathbf{r})$ [10] by using simple radially symmetric models for f_1 and g_2 [13], as well as a Yukawa-type potential for $U(r)$ in Eq. (3) [14]. For the sake of computation, we have chosen spherical Na$_n$ clusters ($Z = 11$) with radii $r_{cl} \simeq (d_{ex}/2) n^{1/3}$ (with $d_{ex} = 7.9$ being the exclusion distance among the constituent ions), and with sizes up to $n = 10^4$, which move through an Al target. In Fig. 1, we show the ratio of the ion charge distribution function to the charge on an isolated isotachic Na ion, $q(r)/Q_0$, versus the reduced radial distance r/r_{cl} from the center of the cluster with (a) $n = 100$ and (b) $n = 1000$ ions, moving at several speeds v. Clearly, the ion charge distribution is rather nonhomogeneous throughout the cluster, displaying an overall reduction of charge compared to isolated ions, as well as the tendency that the cluster interior grows more neutral as both the size n and the speed v increase.

In order to evaluate the cluster stopping power, or the total energy loss per unit path length, we invoke the same statistical arguments as those used to determine the vicinage effects on charge states of constituent ions. From the dielectric theory of cluster stopping [1], we deduce the expected value of the cluster stopping power as follows:

$$S_{cl} = \frac{1}{v} \int \frac{d^3\mathbf{k}}{(2\pi)^3} \frac{4\pi}{k^2} (\mathbf{k}\cdot\mathbf{v}) \mathrm{Im}\left[\frac{-1}{\epsilon(k,\mathbf{k}\cdot\mathbf{v})}\right] F_{cl}(\mathbf{k}), \tag{4}$$

where the cluster structure factor is given by

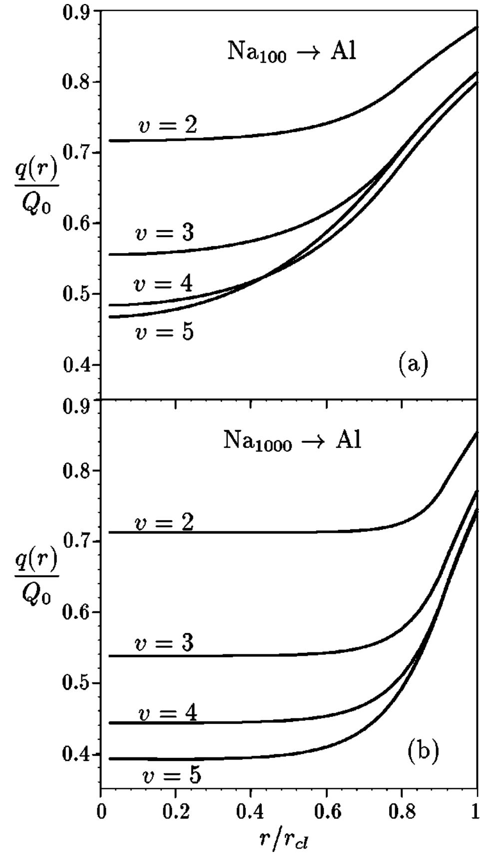

FIG. 1. Reduced distribution of ion charges $q(r)/Q_0$ versus the reduced radial distance r/r_{cl} for Na$_n$ clusters in an Al foil at several speeds v (in a.u.), with (a) $n=100$ and (b) $n=1000$.

$$F_{cl}(\mathbf{k})=\left\langle \sum_{j=1}^{n} |\rho_j(\mathbf{k})|^2+\sum_{j=1}^{n}\sum_{j\neq l=1}^{n} \rho_j(\mathbf{k})\rho_l^*(\mathbf{k})e^{i\mathbf{k}\cdot\mathbf{r}_{jl}}\right\rangle . \tag{5}$$

The ensemble average in $F_{cl}(\mathbf{k})$ must take into account the fact that the distribution of individual ion charges is determined in a self-consistent manner by the geometrical structure of the cluster. This task has been accomplished by solving the integral equation Eq. (2) for the function $q(\mathbf{r})$, which is commensurate with the distribution functions f_1 and f_2 for ion positions. We now use the BK model for the Fourier transform of the charge density on the jth ion and make the transition $\rho_j(\mathbf{k})=\rho_{BK}(\mathbf{k};Q_j)\rightarrow\rho_{BK}[\mathbf{k};q(\mathbf{r})]$ in order to express the cluster structure factor as follows:

$$\begin{aligned}F_{cl}(\mathbf{k})=n\int d^3\mathbf{r}|\rho_{BK}[\mathbf{k};q(\mathbf{r})]|^2 f_1(\mathbf{r})+n(n-1)\\ \times\int\int d^3\mathbf{r}d^3\mathbf{r}'\rho_{BK}[\mathbf{k};q(\mathbf{r})]\rho_{BK}[\mathbf{k};q(\mathbf{r}')]\\ \times f_2(\mathbf{r},\mathbf{r}')e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}.\end{aligned} \tag{6}$$

A more rigorous derivation of this result would use the *joint* one- and two-particle distribution functions for ion positions *and* ion charges, as described in [10]. It should be stressed that the result in Eq. (6) completely describes the influence of the nonhomogeneous ion charge distribution, which is itself a consequence of the vicinage effect, on the interferences in cluster energy losses due to spatial correlation among the constituent ions.

Since the computation of Eq. (6) is still quite tedious, we resort to the PCA by setting $\rho_{BK}[\mathbf{k};q(\mathbf{r})]\approx\rho_{BK}[\mathbf{0};q(\mathbf{r})]=q(\mathbf{r})$. While this approximation is well justified for the interionic or interference part in Eq. (6), the contribution of individual ions in Eq. (6) will be somewhat underestimated [2]. Nevertheless, for the sake of preliminary study of the feedback of the vicinage effect in ion stopping, we proceed with the PCA, aware that the relative role of the interference term in cluster stopping will be somewhat overestimated. After using the solution $q(r)$ of the integral equation Eq. (2) to evaluate the structure factor from Eq. (6) in the PCA, we have used the so-called plasmon-pole approximation [15] for the dielectric function to evaluate the stopping power from Eq. (4) for Na$_n$ clusters moving through an Al target at various speeds v. We call this result S_{nonhom} in order to emphasize that the ion charges are distributed nonhomogeneously due to the vicinage effect, as shown in Fig. 1. Next, we have also evaluated the stopping power, called S_{homog}, for a model of homogeneously charged cluster by taking $q(r)=Q_0=\text{const}$, where Q_0 is the charge on an isolated ion moving at the same speed, and by using the same set of parameters as those used for S_{nonhom}. Finally, we have evaluated the stopping power S_0 of n isolated Na ions moving at the same speed as the cluster, given in the PCA by $S_0=nQ_0^2S_p$, where S_p is the proton stopping power obtained from Eq. (4) by setting $F_{cl}(\mathbf{k})=1$ [15]. In Fig. 2(a), we show the stopping-power ratios $R=S_{nonhom}/S_0$ (thick lines) and $R=S_{homog}/S_0$ (thin lines) versus the cluster size n for two cluster speeds $v=2$ and $v=5$. Thus, the thin lines show the interference effects in cluster stopping due only to spatial correlations among the constituent ions which are all equally charged by Q_0, whereas the thick lines reveal how the nonhomogeneity of the ion charge distribution influences the interferences in cluster stopping. It is clear that the feedback role of the vicinage effect, through the nonhomogeneous charge reduction, generally reduces cluster energy losses. Both sets of curves in Fig. 2(a) exhibit shapes similar to those obtained for hydrogen clusters, with the characteristic resonant effect, where a global maximum is achieved for a specific combination of the cluster size and speed [1]. However, the reduction of stopping power due to the vicinage effect in ion charges cannot be ascribed to a trivial scaling by a factor. This is clearly seen in the largest (first) peaks in the stopping powers for $v=5$, where S_{homog} reaches a maximum for $r_{cl}\omega_p/v\approx1.8$, in agreement with [1,9], while S_{nonhom} reaches its peak for $r_{cl}\omega_p/v\approx1.3$.

In order to further investigate how the charge reduction due to the vicinage effect affects the cluster stopping power, we have evaluated the charge reduction factor per ion, denoted by Γ, in several different ways, which are shown in Fig. 2(b) for two speeds $v=2$ and $v=5$. The dashed lines show the average charge reduction, obtained from $\Gamma=\sqrt{\langle Q^2\rangle}/Q_0$, where $\langle Q^2\rangle=\int d^3\mathbf{r}q^2(\mathbf{r})f_1(\mathbf{r})$. The thin solid

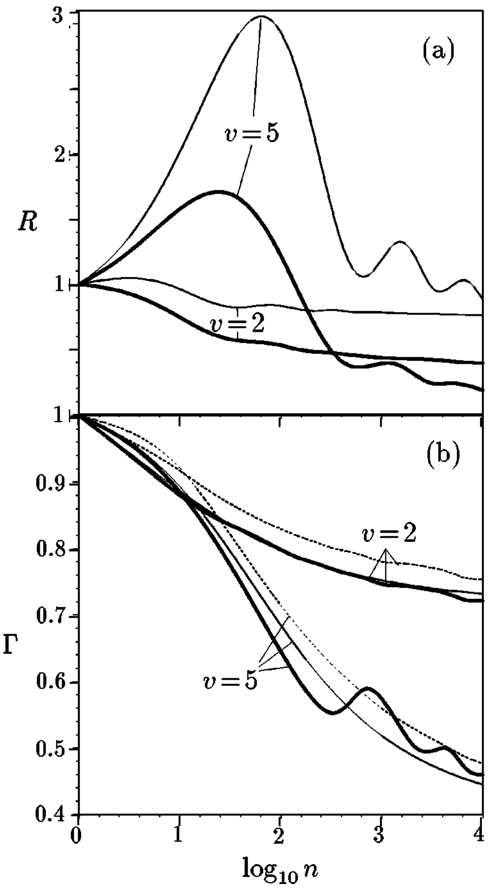

FIG. 2. (a) Stopping power ratios $R=S_{nonhom}/S_0$ (thick lines) and $R=S_{homog}/S_0$ (thin lines) versus the logarithm of cluster size n for Na_n clusters in Al foils at speeds $v=2$ and 5 a.u. (b) Effective charge reductions per ion (see text for details) $\Gamma=\sqrt{\langle Q^2\rangle}/Q_0$ (dashed lines), $\Gamma=Q_c/Q_0$ (thin solid lines), and $\Gamma=\sqrt{S_{nonhom}/S_{homog}}$ (thick solid lines) versus the logarithm of cluster size n for Na_n clusters in Al foils at speeds $v=2$ and 5 a.u.

lines show $\Gamma=Q_c/Q_0$, where Q_c is obtained by the procedure outlined in [4], that is, by *assuming* $q(\mathbf{r})=Q_c=\mathrm{const}$ in the integral equation Eq. (2), which, after multiplication by $f_1(\mathbf{r})$ and integration over $\mathbf{r}$, gives an algebraic equation for Q_c, which is then solved numerically. Finally, the thick solid lines show $\Gamma=\sqrt{S_{nonhom}/S_{homog}}$, where the interferences in stopping power due to spatial correlation among ions are eliminated to some extent, so that this definition of Γ generalizes the concept of the effective stopping charge [11] to clusters. It is evident from Fig. 2(b) that, although the detailed shapes of the three types of curves are different, there exists overall agreement in general trends. This may be quite encouraging news for computations where only an approximate estimate is required for cluster stopping, since one may bypass the complications related to the use of a solution of the integral equation Eq. (2) in the cluster structure factor Eq. (6). In such a situation, it would be sufficient to just evaluate an appropriate average charge per ion, dependent on cluster size and speed.

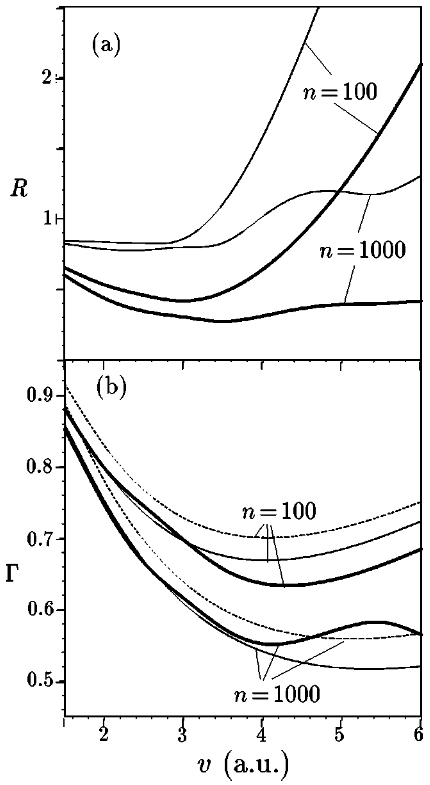

FIG. 3. (a) Stopping power ratios $R=S_{nonhom}/S_0$ (thick lines) and $R=S_{homog}/S_0$ (thin lines) versus the cluster speed v (in a.u.) for Na_{100} and Na_{1000} clusters in Al foils. (b) Effective charge reductions per ion (see text for details) $\Gamma=\sqrt{\langle Q^2\rangle}/Q_0$ (dashed lines), $\Gamma=Q_c/Q_0$ (thin solid lines), and $\Gamma=\sqrt{S_{nonhom}/S_{homog}}$ (thick solid lines) versus the cluster speed v (in a.u.) for Na_{100} and Na_{1000} clusters in Al foils.

Finally, we show in Fig. 3 the same quantities as in Fig. 2 versus the cluster speed, for two cluster sizes $n=100$ and 1000. One may deduce conclusions from the results shown in Fig. 3 that are similar to those regarding Fig. 2. In particular, one notices in Fig. 3(a) that the increase in stopping power with increasing cluster speed, which is expected due to the prevalence of collective excitations for fast clusters [1], appears to be heavily damped when the nonhomogeneous charge reduction is taken into account. As for Fig. 3(b), the three sets of curves are again closely placed, showing broad minima for certain speeds. This effect can be qualitatively attributed to the existence of a minimum of the average charge per ion as the cluster speed v increases, coming from competition between the intensified electron stripping when the kinetic-energy term in Eq. (2), $v^2/2$, increases and the pronounced mutual neutralization of the constituent ions caused by the increasing screening length v/ω_p of the potential U in Eq. (2). As a final comment, we note that the oscillatory behavior of the thick solid lines, shown in both Fig. 2(b) and Fig. 3(b) for larger clusters and higher speeds, may be related to differences in the stopping-power interferences for clusters with nonhomogeneous ion charge distribution compared to clusters with homogeneous ion charge distributions.

In conclusion, we have considered fast clusters made of "heavy" ions, such that the variational-statistical model of Brandt and Kitagawa for the equilibrium charge state of multielectron ions can be generalized to include the vicinage effects in clusters. As a result of minimizing the total electronic energy of the cluster, we have found a strong reduction of ion charges which is distributed nonhomogeneously. By using the point-charge approximation, we have obtained the cluster structure factor, taking into account both the distribution of ion positions and the nonhomogeneous distribution of ion charges. As a result, the dielectric theory gives the stopping power for clusters which is strongly reduced compared to the case of a cluster with the same geometric structure, but with all ions having the charge of an isolated, isotachic ion of the same kind. This can be characterized as a negative feedback action of the vicinage effect in cluster stopping via nonhomogeneous reduction of the constituent-ion charges.

There are several points in the present theory that may be refined, such as the use of more appropriate models of the cluster structure, allowing inclusion of its evolution in the course of penetration through the target, and time-dependent effects and fluctuations in individual ion charges, as well as a more realistic description of the target response. Although the results obtained are of a qualitative nature, the effect described is rather robust and appears to be at the core of observations of energy losses of clusters made of heavy ions, which are not as much increased as one might expect from considering just the spatial interferences in cluster stopping.

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[1] N. R. Arista, Nucl. Instrum. Methods Phys. Res. B **164-165**, 108 (2000).

[2] J. Jensen and P. Sigmund, Phys. Rev. A **61**, 032903 (2000).

[3] A. Brunelle, S. Della-Negra, J. Depauw, D. Jacquet, Y. Le Beyec, and M. Pautrat, Phys. Rev. A **59**, 4456 (1999).

[4] Z. L. Mišković, S. G. Davison, F. O. Goodman, W.-K. Liu, and Y.-N. Wang, Phys. Rev. A **61**, 062901 (2000).

[5] Z. L. Mišković, S. G. Davison, F. O. Goodman, W.-K. Liu, and Y.-N. Wang, Phys. Rev. A **63**, 022901 (2001).

[6] D. Maor, P. J. Cooney, A. Faibis, E. P. Kanter, W. Koenig, and B. J. Zabransky, Phys. Rev. A **32**, 105 (1985).

[7] M. F. Steuer, D. S. Gemmell, E. P. Kanter, E. A. Johnson, and B. J. Zabransky, IEEE Trans. Nucl. Sci. **NS-30**, 1069 (1983).

[8] H.-W. Li, Y.-N. Wang, and Z. L. Mišković, Nucl. Instrum. Methods Phys. Res. B **193**, 209 (2002).

[9] F. J. Pérez-Pérez, I. Abril, N. R. Arista, and R. Garcia-Molina, Nucl. Instrum. Methods Phys. Res. B **115**, 18 (1996).

[10] Z. L. Mišković, W.-K. Liu, F. O. Goodman, and Y.-N. Wang, Phys. Rev. A **64**, 064901 (2001).

[11] W. Brandt and M. Kitagawa, Phys. Rev. B **25**, 5631 (1982).

[12] R. J. Mathar and M. Posselt, Phys. Rev. B **51**, 107 (1995).

[13] M. Vicanek, I. Abril, N. R. Arista, and A. Gras-Marti, Phys. Rev. A **46**, 5745 (1992).

[14] A. F. Lifschitz and N. R. Arista, Phys. Rev. A **57**, 200 (1998).

[15] P. M. Echenique, F. Flores, and R. H. Ritchie, Solid State Phys. **43**, 235 (1990).