PHYSICAL REVIEW A VOLUME 25, NUMBER 4 APRIL 1982

Vicinage effects in ion-cluster collisions with condensed matter and with single atoms

George Basbas*
Department of Physics, North Texas State University, Denton, Texas 76203

R. H. Ritchie
Health and Safety Research Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830
and Department of Physics, University of Tennessee, Knoxville, Tennessee 37916
(Received 2 September 1980)

The excitation of electronic transitions in matter by a group of swift ions traveling close together and at nearly the same velocity may depend on the spatial configuration of ions making up the cluster. We have studied theoretically the effect of configuration on the interactions of clusters with an electron gas and in collision with single atoms (vicinage effect). The latter case is explored with a classical harmonic-oscillator model and with quantum-mechanical perturbation theory. We discuss similarities between the vicinage function for energy loss of a swift cluster in an electron gas and that for the same cluster colliding with a system of noninteracting atoms at condensed-matter density. The aligning effect of the wake potential on the trailing ion of a dicluster penetrating a solid target, as first observed by Gemmell *et al.*, is not expected to occur in cluster collisions with single atoms, and does not occur in collisions with gases at ordinary pressures. Aligning forces comparable with those in solids require target densities of the same order of magnitude as those occurring in condensed matter. The data of Lurio, Anderson, and Feldman taken in a search for vicinage effects in inner-shell excitation are discussed. The effect of wake fluctuations on cluster energy loss is shown to be negligible under ordinary conditions. We evaluate the effect of residual molecular ionic structure on cluster energy loss.

I. INTRODUCTION

When a swift molecular ion impinges on a target, only a few collisions may be needed to strip the valence electrons from the projectile. The residual ionic fragments then begin to recede from one another under the influence of interionic Coulomb forces. At sufficiently small times following stripping, the ions have velocities which are nearly the same as that of the original projectile. When the fragments are traveling close together, there may be substantial vicinage (spatial configuration) effects on electronic transitions induced in the target. A component of the force on one ion arising from transitions induced in the target may be due to the presence of other ions in the cluster. Then the energy loss of the cluster will not, in general, equal the sum of energy losses experienced by each ion when at large distances from all other ions.

Typical initial interionic separations in clusters that have been studied experimentally are ≥ 1 Å. Cluster velocities v, are $\sim 2v_0 - 10v_0$, where $v_0 = e^2/\hbar = 2.19 \times 10^8$ cm/sec. For example, the mean internuclear separation in the H_2^+ ion is ~ 1.29 Å.

In this paper we analyze theoretically vicinage effects for ion diclusters penetrating an electron gas and in collision with single atoms.

Experimental studies of ion cluster penetration have so far been concerned primarily with condensed matter. Neelavathi, Ritchie, and Brandt[1] (NRB) pointed out that the oscillatory wake of electron fluctuations trailing a swift charged particle in condensed matter may give rise to (a) wake-bound electron states, (b) spatially correlated dielectron structures, and (c) spatially correlated ion clusters.[2] Brandt, Ratkowski, and Ritchie[3] showed experimentally and theoretically that the energy loss of proton clusters to valence-electron excitations in solids displays a vicinage effect. Tape *et al.*[4] measured energy losses of diproton clusters and of clusters of two oxygen ions traveling at velocities such that the charge state of each ion was $+4 \pm 1$. Their results also display a pro-

nounced dependence on dicluster configuration. Eckardt *et al.*,[5] have reported measurements of the vicinage effect using H_2^+ ions in the energy range of 12.5 to 139 keV per nucleon and have analyzed their data using the theory of Arista and Ponce.[6] Laubert[7] has studied the vicinage effect in detail by measuring the energy and angular distributions of emerging particles after thin carbon targets were bombarded by H_2^+ and HeH^+ molecular ions in the energy range of 50 to 300 keV.

Gemmell *et al.*[8] studied the angular and energy distribution of protons emerging from crystals bombarded with $(HeH)^+$ ions under planar channeling conditions.[9] They infer that the wake of the leading ion tends to focus the trailing ion in the direction of the former. More recent observations by this group demonstrate clearly the reality of this aligning force.[10] High-resolution studies of the distribution in energy and angle of the fragments produced when swift H_2^+ ions bombard thin amorphous targets show a characteristic bimodal energy distribution at directions nearly parallel to the initial ion velocity. Observed asymmetry in this distribution results from the wake forces. A number of theoretical studies relating to the spatial variation of the wake have been published[11] following the early work of NRB.

Lurio, Andersen, and Feldman[12] have searched for such effects in the x-ray yield of thin solid films under bombardment by H_2^+ projectiles. In their experiments it is primarily the effect of inner-shell excitation of individual lattice atoms that is observed, as we show below.

We employ idealized models in order to compare vicinage effects in condensed matter, where interactions among valence electrons are important, and in collisions resulting in excitation of inner-shell electrons, where interactions between electrons on different ion cores may be neglected. The energy loss of a dicluster is examined using (a) a dielectric model of condensed matter, (b) a classical harmonic-oscillator model of electronic excitation in a single atom, and (c) a quantum-mechanical description of the excitation of an atomic system. Vicinage effects on cross sections for inner-shell ionization are also studied.[13] We evaluate the effect of wake fluctuations (Sec. VI) and residual molecular ion structure (Sec. VII) on cluster energy loss.

II. THE ENERGY LOSS OF A DICLUSTER IN A VALENCE ELECTRON GAS

Suppose that two swift point charges z_1e and z_2e proceed with velocity $\vec{v}$ in a medium characterized by the dielectric function $\epsilon(k,\omega)$. Separation between the charges is specified by the vector $\vec{R}$ with components D and B in directions with, and perpendicular to $\vec{v}$, respectively. In linear-response theory and the first Born approximation, the energy loss of the cluster per unit length to electronic excitation in the medium may be written[3]

$$\mathcal{S}_c=\frac{2e^2}{\pi v^2}\int_0^\infty \kappa\,d\kappa\int_0^\infty \frac{\omega\,d\omega}{k^2}\,\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right]\left[z_1^2+z_2^2+2z_1z_2J_0(\kappa B)\cos\left[\frac{\omega D}{v}\right]\right] \tag{2.1}$$

$$\equiv(z_1^2+z_2^2)\mathcal{S}_p+2z_1z_2\mathcal{S}_v(B,D)\ . \tag{2.1a}$$

Here $\mathcal{S}_p$, which is often written as $-dE/dR$, is the energy loss per unit path length of a single proton having the same velocity as the cluster. The vicinage stopping power $\mathcal{S}_v$ becomes equal to $\mathcal{S}_p$ as $R\to 0$ (the united ion case) and goes to zero as $R\to\infty$. In Eq. (2.1), $k^2=\kappa^2+\omega^2/v^2$ and the medium is assumed to be homogeneous and isotropic. $J_0(x)$ is the Bessel function of first kind and zero order.

We do not consider energy loss to inner-shell electrons in the context of a dielectric treatment, since collective effects are much less important for these electrons than for valence electrons. Instead we employ atomic models to describe such excitations in Secs. III and IV below.

The dielectric response functions of the valence electrons of model solids have been investigated in much detail. In Ref. 2 a plasmon-pole approximation to $\epsilon(k,\omega)$ for an electron gas was used, and numerical evaluation of $\mathcal{S}_v$ was done. In order to get easily surveyed analytical results, we employ a simplified form which exhibits collective and single-particle effects. We take

$$\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right]=\frac{\pi\omega_p^2}{2\omega}\left[\delta(\omega-\omega_p)\Theta(k_c-k)+\delta\left[\omega-\frac{\hbar k^2}{2m}\right]\Theta(k-k_c)\right], \tag{2.2}$$

where $\hbar\omega_p$ is the plasma energy of the electron gas and the choice $k_c=(2m\omega_p/\hbar)^{1/2}$ allows the two δ functions in Eq. (2.2) to coincide at $k=k_c$ in the k-ω plane. The first term in Eq. (2.2) describes the response due to nondispersive plasmon excitation in the region $k<k_c$, while the second term describes free-electron recoil in the range $k>k_c$. This approximate function satisfies the sum rule $\int_0^\infty \omega\,\mathrm{Im}[-\epsilon^{-1}(k,\omega)]d\omega=\pi\omega_p^2/2$ for all values of k and gives a good qualitative account of the vicinage effect for the present purposes. The Heaviside step function is designated by $\theta(x)$ in Eq. (2.2).

In this approximation

$$\mathscr{S}_p=\frac{2e^2}{\pi v}\int_0^\infty \kappa\,d\kappa\int_0^\infty \frac{\omega\,d\omega}{\kappa^2+\omega^2/v^2}\,\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right]$$
$$\approx\frac{e^2\omega_p^2}{v^2}\ln\left[\frac{2mv^2}{\hbar\omega_p}\right], \qquad (2.3)$$

if $v>>(\hbar\omega_p/2m)^{1/2}\equiv v_p$. This agrees exactly with the Bethe stopping-power formula,[14] except that the plasmon energy of the electron gas $\hbar\omega_p$ appears instead of the usual mean atomic excitation energy. Equation (2.3) represents the contribution of valence electrons in a solid to the stopping of an ion. Contributions from excitation of inner shells should be added to this equation, of course, to obtain the total stopping power of the medium, yielding the nonrelativistic form

$$\mathscr{S}_p=\frac{4\pi e^4 Z_2 n_A}{mv^2}\left[\ln\left[\frac{2mv^2}{I}\right]-\frac{C(Z_2)}{v^2}+\cdots\right],$$

where I is the mean excitation energy, n_A is the atomic density of the medium, and Z_2 is the atomic number of the medium. The second term in the braces accounts for inner-shell effects, and corrections for the Z_1^3 effect and other higher-order effects have been neglected.

Using Eq. (2.2) in Eq. (2.1), one finds

$$\mathscr{S}_v(B,D)=\frac{e^2\omega_p^2}{v^2}\left[\cos\left[\frac{\omega_p D}{v}\right]\int_0^{\kappa_c}\frac{\kappa J_0(\kappa B)}{\kappa^2+\omega_p^2/v^2}d\kappa+\int_{k_c}^{k_2}\frac{dk}{k}\cos\left[\frac{\hbar k^2 D}{2mv}\right]J_0(QB)\right], \qquad (2.4)$$

where $Q^2=k^2-(\omega_k/v)^2$, $\kappa_c^2=k_c^2-\omega_p^2/v^2$, $\omega_k=\hbar k^2/2m$, and $k_2=2mv/\hbar$. We examine the cases $B\to 0$ and $D\to 0$ separately.

(i) $B=0$; one ion trails directly behind the other. In this limit and for $v>>v_p$,

$$\mathscr{S}_v(0,D)=\frac{e^2\omega_p^2}{v^2}\left\{\cos\frac{\omega_p D}{v}\ln\left[\left[\frac{2mv^2}{\hbar\omega_p}\right]^{1/2}\right]+\tfrac{1}{2}\mathrm{Ci}\left[\frac{2mv}{\hbar}D\right]-\frac{1}{2}\mathrm{Ci}\left[\frac{\omega_p D}{v}\right]\right\},$$

where $\mathrm{Ci}(x)=-\int_x^\infty \cos u\,du/u$ is a tabulated function.[15] Note that as $D\to 0$, $\mathscr{S}_v\to\mathscr{S}_p$ as it must, since in this limit the two ions coalesce and $\mathscr{S}_c\to(z_1+z_2)^2\mathscr{S}_p$. Figure 1(a) shows a plot of the ratio $g(0,\omega_p D/v)$ vs $(\omega_p D/v)$, for $v=3v_0$ and

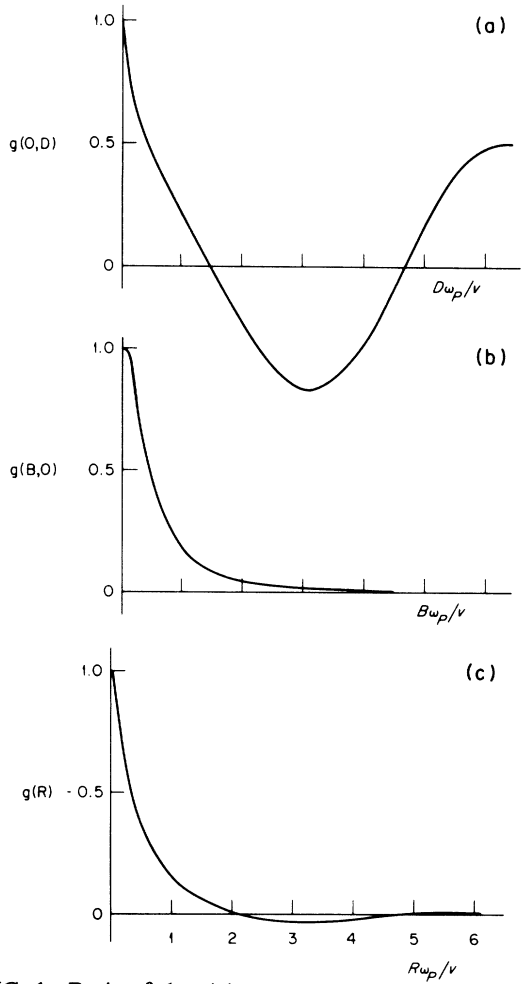

FIG. 1. Ratio of the vicinage term in the stopping power of an electron gas for a diproton cluster to the stopping power for a proton having the same velocity. The velocity is taken to be $v=3v_0$ and is assumed to be $>>(2\hbar\omega_p/m)^{1/2}$, and the plasma energy of the electron gas is $\hbar\omega_p=15$ eV. (a) gives the ratio when one proton trails directly behind the other by the distance D. (b) shows the ratio when the two protons travel directly abreast, while (c) shows this ratio averaged over all orientations.

$\hbar\omega_p = 15$ eV. We define the vicinage function $g(B,D) \equiv \mathscr{S}_v(B,D)/\mathscr{S}_p$, and similarly for other vicinage functions considered below. One sees a descent from unity at $D=0$ to a first zero at $\omega_p D/v \approx 1.5$ and a characteristic oscillatory behavior for large argument. As discussed in Ref. 3, fluctuations in the stopping power of a medium for a cluster as separation increases are due to electron density variation in the wake of the leading ion. The wavelength of these fluctuations is $2\pi v/\omega_R$, where ω_R is the collective resonance frequency of the electrons in the medium.

(ii) $D=0$; ions travel abreast. In this case we find

$$\mathscr{S}_v(B,0) = \frac{e^2\omega_p^2}{v^2}\int_0^{\kappa_c} \frac{\kappa J_0(\kappa B)d\kappa}{\kappa^2+\omega_p^2/v^2}\ , \tag{2.5}$$

if $v >> v_p$. Although the integral in this equation is not tabulated, it approaches the modified Bessel function $K_0(B\omega_p/v)$ as $2mv^2/\hbar\omega_p$ becomes large compared with one. When $B \to 0$, Eq. (2.5) shows that $g(0,0)=1$. Figure 1(b) shows a sketch of $g(\omega_p B/v, 0)$ vs $(\omega_p B/v)$ computed from Eq. (2.5).

A more accurate representation of $\mathscr{S}_v(B,D)$ may be obtained by employing the plasmon-pole approximation to $\epsilon(k,\omega)$ in

$$\mathscr{S}_v(B,D) = \frac{2e^2}{\pi v^2}\int_0^\infty \kappa\, d\kappa \int_0^\infty \frac{\omega\, d\omega}{k^2}\,\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right] \times J_0(\kappa B)\cos\left[\frac{\omega D}{v}\right]. \tag{2.6}$$

The plasmon-pole approximation has been used extensively[2,3] to represent the response function of an electron gas. It may be written

$$\epsilon(k,\omega) = 1+\omega_p^2/[\,\beta^2k^2+\hbar^2k^4/4m^2 - \omega(\omega+i\gamma)]\ , \tag{2.6a}$$

where $\beta^2 = 3v_F^2/5$ is the square of the mean propagation velocity of disturbances in the system and v_F is the Fermi velocity of the electron gas. Both collective and single-particle effects in an electron gas are represented in $\epsilon(k,\omega)$. Figures (2)–(4) show the results of calculations made using Eq. (2.6a) in Eq. (2.6). The results are represented by the vicinage function $g(B,D)=\mathscr{S}_v(B,D)/\mathscr{S}_p$, where $\mathscr{S}_p$ is taken from Eq. (2.3). The constants employed are specified in the figure captions. One sees the same kind of general trends displayed in Figs. 1(a) and 1(b). Crispations of wavelength $\sim h/mv$ corresponding to single-particle effects are prominent in the region $1 \lesssim B \lesssim 4$ and $0 < D < 4$,

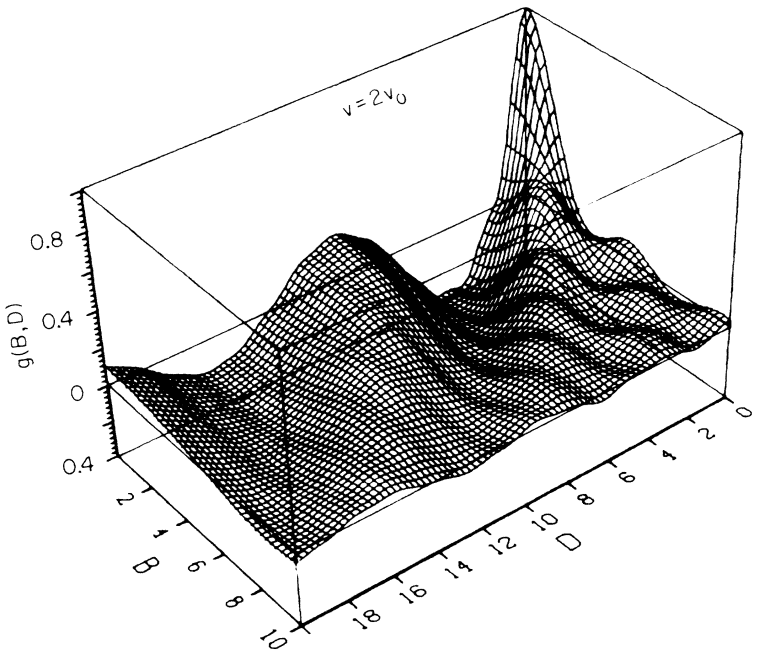

FIG. 2. A vicinage function $g(B,D)=\mathscr{S}_v(B,D)/\mathscr{S}_p$ plotted as a function of B and D for stopping of a dicluster proceeding with velocity $v=2$ a.u. in a medium characterized by plasma frequency $\omega_p=0.55$ a.u. = 15 eV. The response of the medium is taken to be given by the plasmon-pole dielectric function, Eq. (2.6a). Atomic units are used. Lengths are measured in units of 0.529 Å, while the unit of velocity $v_0=e^2/\hbar=2.19\times10^8$ cm/sec.

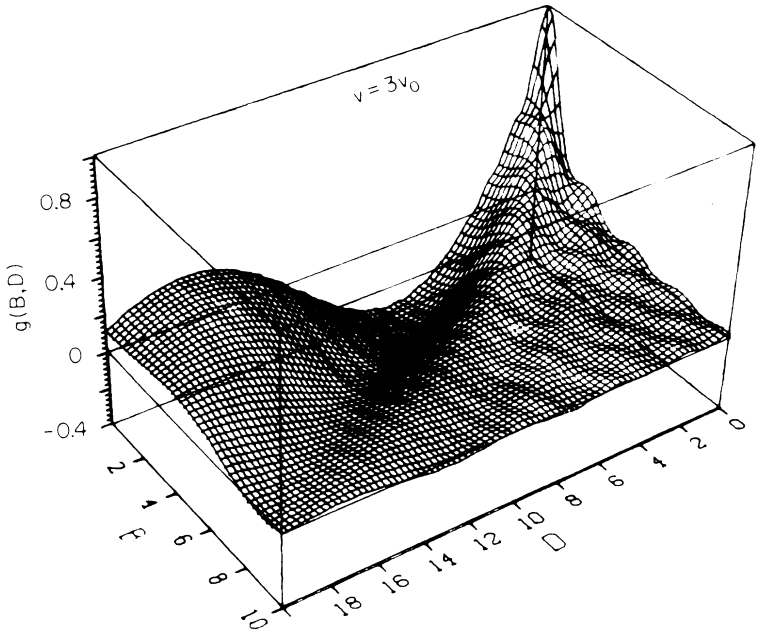

FIG. 3. A vicinage function $g(B,D)$ appropriate to a medium such as that specified in the caption of Fig. 2, and for a dicluster velocity $v=3$ a.u.

where B and D are measured in atomic units ($a_0=0.529$ Å).

In many experimental situations, clusters are formed with random orientations of $\vec{R}$. A vicinage stopping power appropriate to this situation may be obtained by carrying out a spherical average over $\vec{R}$ of the quantity $\mathscr{S}_v$ in Eq. (2.1). We find

$$\langle \mathscr{S}_c \rangle = (z_1^2+z_2^2)\mathscr{S}_P + 2z_1z_2\mathscr{S}_v(R) , \qquad (2.7a)$$

where

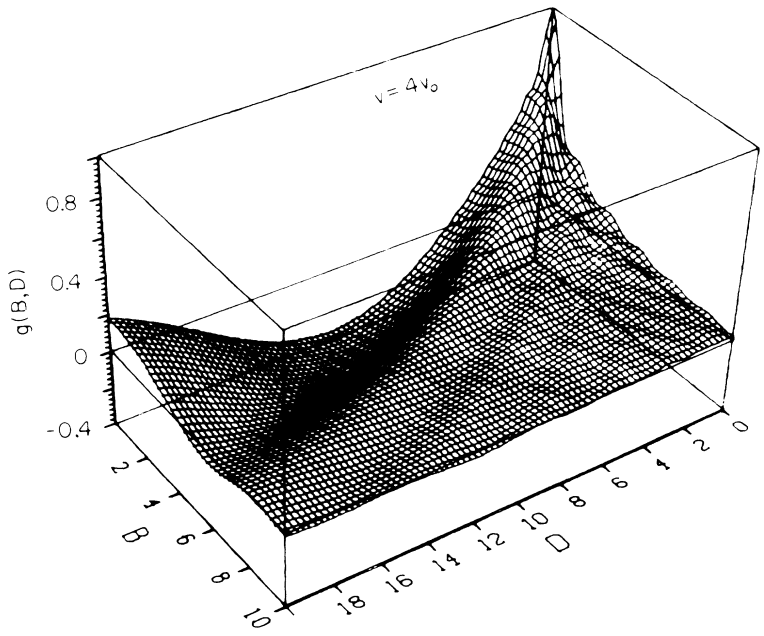

FIG. 4. A vicinage function $g(B,D)$ appropriate to a medium with properties specified in the caption of Fig. 2, and for a dicluster velocity $v=4$ a.u.

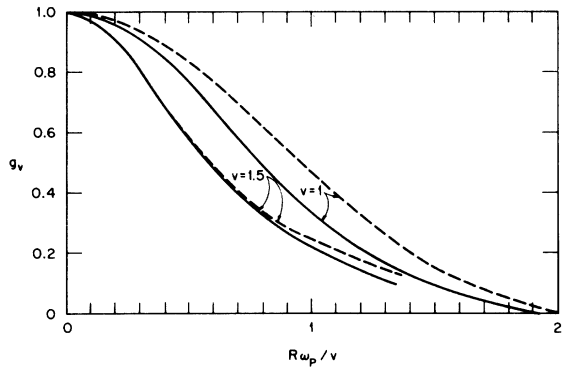

FIG. 5. A comparison of the vicinage function for randomly oriented clusters penetrating an electron gas. Computed from Eq. (2.7) employing the Lindhard dielectric function of an electron gas (solid lines) and using the approximate formula of Eq. (2.8) (dashed lines). The two upper curves correspond to a cluster velocity of $v=1$ a.u., while the lower pair correspond to $v=1.5$ a.u. The results become nearly the same for $v\gtrsim 2$ a.u.

$$\mathscr{S}_v(R)=\langle \mathscr{S}_v(R\sin\theta,R\cos\theta)\rangle = \frac{2e^2}{\pi v^2}\int_0^\infty \frac{dk}{k}\frac{\sin kR}{kR}\int_0^{vk}\omega\, d\omega\,\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right] . \tag{2.7b}$$

The approximation of Eq. (2.2) when substituted into Eq. (2.7b) yields,[16] for $v>>v_p$,

$$\mathscr{S}_v(R)=\frac{e^2\omega_p^2}{v^2}\left[\mathrm{si}_2\left(\frac{\omega_p R}{v}\right)-\mathrm{si}_2(2mvR/\hbar)\right], \tag{2.8}$$

where we take

$$\mathrm{si}_2(x)\equiv\int_x^\infty \frac{\sin u\, du}{u^2}=\frac{\sin x}{x}-\mathrm{Ci}(x) . \tag{2.9}$$

Figure 1(c) shows a plot of $g(R)\equiv\mathscr{S}_v(R)/\mathscr{S}_p$ vs $(R\omega_p/v)$ for the case $v=3v_0$ and for $\hbar\omega_p=15$ eV. As $R\to 0$, $g(R)\to 1$ and as $\omega_p R/v$ becomes large compared with 1,

$$g(R)\sim \cos(\omega_p R/v)\Big/\ln\left[\frac{2mv^2}{\hbar\omega_p}\right]\left[\frac{\omega_p R}{v}\right]^2 . \tag{2.10}$$

Oscillations in $g(B,D)$ are strongest when the trailing ion moves directly behind the leading ion, Fig. 1(a). Orientational averaging leads to strong damping of these oscillations.

The accuracy of the approximation of Eq. (2.2) to the dielectric response function of an electron gas for vicinage calculations may be checked in a straightforward way for the case of random dicluster orientation. We have evaluated numerically Eq. (2.7b) using the Lindhard dielectric function[17] of an electron gas. The density assumed was that of the conduction electrons in Al metal (the one-electron radius $r_s=2.07$, $\hbar\omega_p=15.8$ eV) and a range of velocities was studied. Figure 5 shows some of the results, which have been plotted as a function of $R\omega_p/v$ for purposes of comparison. Data computed using the Lindhard $\epsilon(k,\omega)$ are shown as solid lines, while Eq. (2.8) yields values plotted as dashed lines. For $v\geq 2$ the corresponding results are indistinguishable on this scale. When $v\leq v_F=0.93v_0$ at $r_s=2.07$, the approximation of Eq. (2.2) yields $\mathscr{S}_v(R)=0$ when substituted in Eq. (2.7b).

The effect of the presence of an energy gap in a solid upon the vicinage function may be estimated by employing the Callaway-Tosatti model dielectric function[18] of a semiconductor in Eq. (2.7b). A better approximation would be obtained using the Brandt-Reinheimer[19] dielectric function, based on the Penn model[20] of a semiconductor. However, because of the numerical complexity of this function, we have used the simpler Callaway-Tosatti approximation and have employed an energy gap which yields the same partition constant $C(r_s,\epsilon_g)$ found by Brandt and Reinheimer at the same value of r_s. Figure 6 shows a plot of $g(R)$ vs $2k_FR$ at a velocity $v=3v_0$ and for $r_s=2.07$. The equivalent energy gap value E_g is specified in the caption for

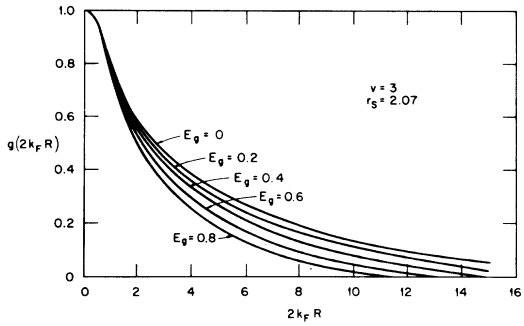

FIG. 6. The vicinage function of a randomly oriented dicluster penetrating a semiconductor. The Calloway-Tosatti model of the dielectric function of the semiconductor has been used in this calculation. The gap energy E_g is specified in units of the Fermi energy E_F, and the cluster separation is expressed in units of $(2k_F)^{-1}$. The cluster velocity $v=3$ a.u., and the one-electron radius $r_s=2.07$ in the model.

each curve and is given in units of E_F. The Fermi wave number $k_F = mv_F/\hbar$. These results indicate that cluster energy loss in material characterized by a sizable energy gap $E_g \sim 0.3$ may differ by several percent from the loss of the same cluster in a metal with the same Fermi energy but with $E_g = 0$.

III. ENERGY LOSS TO A CLASSICAL, HARMONICALLY BOUND ELECTRON

Bohr's[21] harmonic-oscillator model for the response of an atomic electron to swift charged-particle bombardment may be generalized easily to include cluster collisions. This model is very useful in displaying the physics of charged-particle interactions in matter and, as well, gives a good quantitative description of optically allowed (dipole) electronic transitions in atoms under charged-particle bombardment.

A bound classical electron, represented as an isotropic harmonic oscillator with resonance frequency ω, will evolve in time under the influence of a moving dicluster according to Newton's equation,

$$m\ddot{\vec{r}} + m\omega^2\vec{r} = \nabla_{\vec{r}}\left[\frac{z_1 e^2}{|\vec{r}-\vec{v}t-\vec{b}_1|} + \frac{z_2 e^2}{|\vec{r}-\vec{v}t-\vec{b}_2|}\right]. \quad (3.1)$$

The electron is bound to a center of force at the origin of coordinates, and the uniform rectilinear motion of the cluster is such that the projectile with charge $z_j e$ moves with impact parameter $\vec{b}_j$ relative to this center, where $j = 1$ or 2. As in the previous section, $\vec{R} = \vec{b}_2 - \vec{b}_1$ describes the orientation of the dicluster.

At large velocities the impulse approximation may be invoked; $\vec{r}$ may be set equal to zero on the right-hand side of Eq. (3.1) after the gradient operation is carried out. Then the net energy $\Delta E_c(\vec{b}_1, \vec{R})$ transferred to the electron at large times following the collision may be written

$$\Delta E_c = \frac{e^4}{2m}\left|\int_{-\infty}^{\infty} dt\, e^{i\omega t}\left[\nabla_{\vec{r}}\left[\frac{z_1}{|\vec{r}-\vec{v}t-\vec{b}_1|} + \frac{z_2}{|\vec{r}-\vec{v}t-\vec{b}_2|}\right]\right]_{\vec{r}\to 0}\right|^2, \quad (3.2)$$

where we now express ΔE_c in terms of contributions ΔE_1 and ΔE_2 from each ion plus a cross term ΔE_v. By straightforward manipulation the integrals in Eq. (3.2) may be expressed in terms of tabulated functions,

$$\Delta E_j = \frac{2}{m} z_j^2 \left[\frac{\omega e^2}{v^2}\right]^2 \left[K_0^2\left[\frac{\omega b_j}{v}\right] + K_1^2\left[\frac{\omega b_j}{v}\right]\right]. \quad (3.3)$$

Here $K_0(x)$ and $K_1(x)$ are modified Bessel functions of the second kind, and z_j and $\vec{b}_j$ are the charge number and impact parameter appropriate to projectile j, respectively. In this notation $b_1 = |\vec{b}_1|$ and $b_2 = |\vec{b}_2| = |\vec{b}_1 + \vec{R}|$. The vicinage term is

$$\Delta E_v = 4z_1 z_2 \frac{\omega^2 e^4}{mv^4} \cos\left[\frac{\omega D}{v}\right]\left[\frac{\vec{b}_1\cdot\vec{b}_2}{b_1 b_2} K_1\left[\frac{\omega b_1}{v}\right] K_1\left[\frac{\omega b_2}{v}\right] + K_0\left[\frac{\omega b_1}{v}\right] K_0\left[\frac{\omega b_2}{v}\right]\right]. \quad (3.4)$$

The trigonometric factor $\cos(\omega D/v)$ originates in the fact that the time delay D/v between impulses delivered to the electron by the ions may result in constructive interference $D/v \sim 2n\pi/\omega$, or destructive interference $D/v \sim (2n+1)\pi/\omega$. Here $n = 0, 1, 2, \ldots$.

In most experimental situations involving cluster bombardment, the range of impact parameters is effectively unlimited. To obtain comparable theoretical results, Eq. (3.2) must be integrated over all $\vec{b}_1$ in order to obtain S, the stopping cross section for this collision. Note that S has dimensions of energy times area. This integration may be carried out by utilizing the momentum representation of the Coulomb potential in Eq. (3.2), i.e.,

$$r^{-1} = \frac{1}{2\pi^2}\int\int\int \frac{e^{i\vec{q}\cdot\vec{r}}}{q^2} d^3q. \quad (3.5)$$

Then

$$\Delta E_c = \frac{e^4}{2\pi^2 mv^2} \times \left|\int\int \frac{d^2Q}{q^2}\vec{q}\, e^{-i\vec{Q}\cdot\vec{b}_1}(z_1 + z_2 e^{-i\vec{q}\cdot\vec{R}})\right|^2, \quad (3.6)$$

where $\vec{q}=\vec{Q}+\vec{v}\omega/v^2$, $q^2=Q^2+\omega^2/v^2$, and the integration is over the two-dimensional space of $\vec{Q}$. Expanding the absolute square in powers of z_1 and z_2 and integrating over $\vec{b}_1$ with $\vec{R}$ fixed, we may write

$$S=\int\int d^2b\,\Delta E_c=(z_1^2+z_2^2)S_p+2z_1z_2S_v\ . \tag{3.7}$$

Here S_p is the stopping cross section of the electron for a proton with velocity v, which in this model may be written

$$S_p=\frac{4\pi e^4}{mv^2}\int_0^{Q_m}\frac{Q\,dQ}{Q^2+\omega^2/v^2}\ , \tag{3.8}$$

where the upper limit Q_m must be found outside of the framework of the classical harmonic-oscillator model. A reasonable approximate value of $Q_m=(k_{\max}^2-\omega^2/v^2)^{1/2}$ may be obtained by setting $(\hbar k_{\max})^2/2m=\hbar\omega$. In effect this assumes that the model breaks down when the momentum transferred to the struck electron, if given to a stationary free electron, would deliver energy to the latter exceeding $\hbar\omega$, the quantum energy of the oscillator.

However, we may also choose a value for $k_{\max}$ such that an additional contribution is obtained from the region of small impact parameters where the struck electron may be regarded as essentially free after the collision and where large momenta may be absorbed by it. This may be done by setting $\hbar k_{\max}=2mv$, the maximum momentum which can be transferred to a free electron from a swift massive ion in a head-on collision. This yields

$$S_p=\frac{4\pi e^4}{mv^2}\ln\left[\frac{2mv^2}{\hbar\omega}\right] \tag{3.9}$$

for the bound electron, when $2mv^2>>\hbar\omega$. This atomic model may be generalized immediately to one consisting of Z harmonically bound electrons with the ith having oscillator strength f_i and resonant frequency ω_i. Then summing over i and multiplying by n_A, the density of atoms in the target, one finds

$$S_p=\frac{4\pi n_A e^4}{mv^2}\sum_i f_i\ln\left[\frac{2mv^2}{\hbar\omega_i}\right]\ ,$$

the stopping power of the target for a proton. This expression corresponds to the nonrelativistic form of the Bethe stopping-power formula.[14]

A comparable expression for the vicinage term in the energy-transfer cross section is

$$S_v(B,D)=\frac{4\pi e^4}{mv^2}\cos\left[\frac{\omega D}{v}\right]\times\int_0^{Q_m}\frac{Q\,dQ}{Q^2+\omega^2/v^2}J_0(QB)\ . \tag{3.10}$$

If $BQ_m>>1$, we may write

$$S_v(B,D)\rightarrow\frac{4\pi e^4}{mv^2}\cos\left[\frac{\omega D}{v}\right]K_0\left[\frac{\omega B}{v}\right]\ , \tag{3.11}$$

while for $B\rightarrow 0$,

$$S_v(B,D)\rightarrow\frac{4\pi e^4}{mv^2}\cos\left[\frac{\omega D}{v}\right]\ln\left[\frac{vk_{\max}}{\omega}\right]\ . \tag{3.12}$$

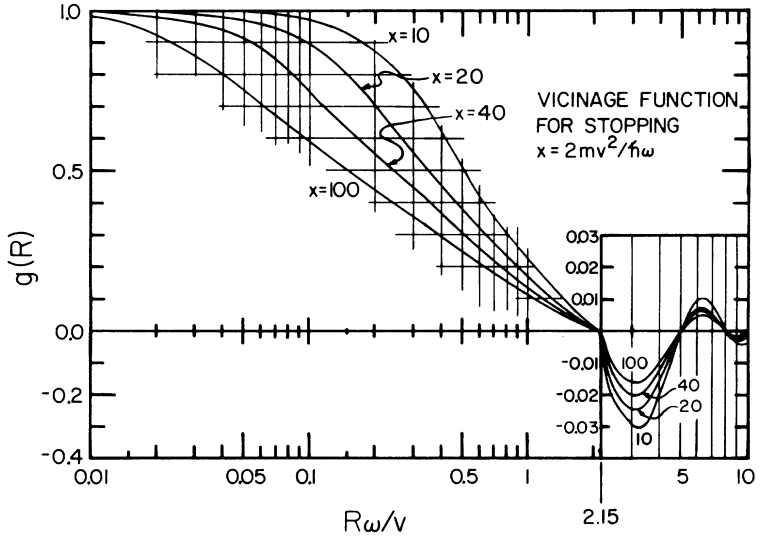

FIG. 7. A vicinage function for stopping of randomly oriented diclusters. The quantity $g(\omega R/v)=[\mathrm{si}_2(R\omega/v)-\mathrm{si}_2(xR\omega/v)]/\ln x$, where $x=2mv^2/\hbar\omega$. It is plotted as a function of $R\omega/v$ for several different values of x.

An average of S_v over directions of $\vec{R}$ yields

$$S_v(R) \equiv \langle S_v(B,D) \rangle = \frac{4\pi e^4}{mv^2}\left[\mathrm{si}_2\left[\frac{\omega R}{v}\right] - \mathrm{si}_2\left[\frac{2mvR}{\hbar}\right]\right] \tag{3.13}$$

for the case where the dicluster orientation is completely random on incidence. Figure 7 shows a plot of $g(\omega R/v)=S_v/S_p$, computed from Eqs. (3.9) and (3.13), as a function of $\omega R/v$ for several different values of $x \equiv 2mv^2/\hbar\omega$. This function g, appropriate to the stopping cross section of a single harmonically bound electron with resonant frequence ω, may be used to make estimates of the magnitude of the vicinage effect for optically allowed transitions. As one sees by referring to Eqs. (2.3) and (2.8), this function is also appropriate for diclusters with random orientation traveling in an electron gas. Note that in all of these results we have assumed that $v >> (\hbar\omega_R/m)^{1/2}$, where ω_R is the resonant frequency of the medium.

Even though the classical harmonic-oscillator model is used only for qualitative comparison, it seems clear that an interesting correspondence exists between the response to cluster bombardment of a dielectric medium and that of the single oscillator we have considered above. The similarities between Eqs. (2.4) and (3.10) for the oriented case and between Eqs. (2.8) and (3.13) for the case of random incidence are striking. In summing over impact parameters to obtain a stopping cross section, Eq. (3.7), one in effect builds up a continuous medium of noninteracting harmonic-oscillator atoms, each with resonant frequency ω. The net vicinage response of this medium is quite comparable with the response of an electron gas with plasma frequency $\omega_p=\omega$. Plasma oscillations in an electron gas are of dipole character and, though originating in long-range Coulombic interactions between nearly free electrons, have spatial extension depending in part on the speed of the ion.

The vicinage terms discussed above depend on cluster separation R primarily through the parameter $R\omega/v$, where ω is a characteristic electronic transition frequency for an atom or the plasma frequency for the case of an electron gas. A simple physical argument justifying the appearance of this parameter may be made. The quantity $\tau=R/v$ is a measure of the time delay between impulses delivered to a given electron in the medium by the ions. If τ is large compared with $T=1/\omega$, proportional to the characteristic period of an oscillator, one expects only small vicinage effects. On the other hand, if $\tau/T=\omega R/v$ is comparable with, or smaller than unity, appreciable interference between the effects of the ions making up the cluster is expected to occur.

For purposes of qualitative comparison, we may derive an expression for the cross section σ for excitation of the bound electron by the cluster. In this model we need only divide Eqs. (3.7)–(3.13) by $\hbar\omega$, the quantum energy of the oscillator, and also choose $k_{\max}=(2m\omega/\hbar)^{1/2}$ to obtain a qualitative measure of the value of momentum transfer at which the harmonic-oscillator model is expected to break down, as discussed following Eq. (3.8). We may then write

$$\sigma=(z_1^2+z_2^2)\sigma_p+2z_1z_2\sigma_v \ , \tag{3.14}$$

where

$$\sigma_p \sim \frac{4\pi e^4}{\hbar m\omega v^2}\ln\left[\left[\frac{2mv^2}{\hbar\omega}\right]^{1/2}\right] \tag{3.15}$$

and

$$\sigma_v=\frac{4\pi e^4}{\hbar m v^2\omega}\left\{\mathrm{si}_2\left[\frac{\omega R}{v}\right] - \mathrm{si}_2\left[\left[\frac{2m\omega}{\hbar}\right]^{1/2}R\right]\right\}. \tag{3.16}$$

Equations (3.15) and (3.16) may be used to obtain estimates of the vicinage effect for random orientation as manifested in the cross section per electron for excitation of an optically allowed transition. These expressions can be generalized by multiplying Eqs. (3.15) and (3.16) by f_i, the oscillator strength for the ith transition in the atom and by replacing ω by ω_i, the frequency corresponding to this transition.

IV. QUANTUM THEORY OF ATOMIC EXCITATION BY DICLUSTERS

Electronic transition of the atom from an initial state designated by the eigenket $|i\rangle$ with eigenenergy E_i to a final state corresponding to the eigenket $|f\rangle$ with eigenenergy E_f is now considered. We choose the origin of energy so that $E_i=0$. As in Sec. III above, the position of the charge z_1e at time t is taken to be $\vec{b}_1+\vec{v}t=\vec{r}$, while that of charge z_2e is $\vec{R}+\vec{r}$ and the atom is located at the origin. The probability, P_{fi}, that this transition has occurred at large times after the collision is given compactly in first-order, time-dependent perturbation theory as

$$P_{fi}(\vec{b}_1)=\frac{1}{\hbar^2}\left|\int_{-\infty}^{\infty}dt e^{iE_f t/\hbar}\left\langle f\left|\sum_{j=1}^{Z_2}\left[\frac{-z_1e^2}{|\vec{r}_j-\vec{b}_1-\vec{v}t|}+\frac{-z_2e^2}{|\vec{r}_j-\vec{b}_1-\vec{R}-\vec{v}t|}\right]\right|i\right\rangle\right|^2 \tag{4.1}$$

The sum inside the matrix element is over all coordinates, $\vec{r}_j$, of electrons on the atom of atomic number Z_2. Again employing the momentum representation of the Coulomb energy terms and carrying out the integrations over time, we obtain δ functions which may be used to reduce the integrals over momentum to components perpendicular to $\vec{v}$ only. Then

$$P_{fi}(\vec{b}_1)=\left[\frac{e^2}{\pi\hbar v}\right]^2\left|\int\int d^2Q\frac{e^{-i\vec{Q}\cdot\vec{b}_1}}{Q^2+q_0^2}F_{fi}(q)[z_1+z_2e^{-i\vec{q}\cdot\vec{R}}]\right|^2, \tag{4.2}$$

where $\vec{q}=(\vec{Q},q_0)$, $q_0=E_f/\hbar v$, and $\vec{Q}$ is defined such that $\vec{v}\cdot\vec{Q}=0$. Note the similarity between Eqs. (3.6) and (4.2). The latter, of course, reduces to the former when the dipole approximation to $F_{fi}(\vec{q})$ is made (see below). The form factor

$$F_{fi}(\vec{q})=\left\langle f\left|\sum_{j=1}^{Z_2}e^{i\vec{q}\cdot\vec{r}_j}\right|i\right\rangle=\left\langle E_f,\hat{\Omega}\left|\sum_{j=1}^{Z_2}e^{i\vec{q}\cdot\vec{r}_j}\right|i\right\rangle, \tag{4.3}$$

where the final-state vector $|E_F,\hat{\Omega}\rangle$ is denoted by its eigenenergy E_f and by the direction $\hat{\Omega}$ of the ejected electron if the final state lies in the continuum.

The cross section σ^f for the transition is obtained by integrating $P_{fi}(\vec{b})$ over all impact parameters with $\vec{R}$ fixed; thus

$$\sigma^f=\int d^2bP_{fi}(b)=\left[\frac{2e^2}{\hbar v}\right]^2\times\int\int\frac{d^2Q}{q^4}|F_{fi}(\vec{q})|^2|z_1+z_2e^{-i\vec{q}\cdot\vec{R}}|^2. \tag{4.4}$$

Integration over directions of ejection of the electrons in final states having fixed energies or summation over discrete final states having the same energy E_f may be carried out so that the form factor as written henceforth depends only on the magnitude of $\vec{q}$. In the case of continuum final states $\sigma^f\to d\sigma^f/dE_f$. Expanding the term in Eq. (4.4) that depends on z_1 and z_2, we have

$$\sigma^f=(z_1^2+z_2^2)\sigma_p^f+2z_1z_2\sigma_v^f(\vec{R}), \tag{4.5a}$$

where

$$\sigma_p^f=\sigma_v^f(0) \tag{4.5b}$$

and

$$\sigma_v^f(\vec{R})=\left[\frac{2e^2}{\hbar v}\right]^2\times\int\int\frac{d^2Q}{q^4}|F_{fi}(q)|^2\cos(\vec{q}\cdot\vec{R}). \tag{4.5c}$$

For simplicity, we do not treat the case in which the direction of $\vec{R}$ is fixed but instead proceed directly to consider the ions composing the cluster to be oriented at random. Averaging over directions of $\vec{R}$ and changing the integration variable from $\vec{Q}$ to the magnitude of $q=(Q^2+q_0^2)^{1/2}$, we find

$$\sigma_v^f(R)=2\pi\left[\frac{2e^2}{\hbar v}\right]^2\int_{q_0}^{\infty}\frac{dq}{q^3}\frac{\sin qR}{qR}|F_{fi}(q)|^2, \tag{4.6}$$

which replaces $\sigma_v^f(\vec{R})$ in an equation similar to Eq. (4.5a) for the randomly oriented case.

At high velocities the lower limit of the integral over q is small, and in this region the dipole approximation for $|F_{fi}(q)|^2$ may be exploited and sum rules may be invoked as in Bethe's derivation of the stopping power of atoms for single ions.[14] The analysis is facilitated by dividing the integration into two regions joined at some intermediate value of q, designated as q_I.

The stopping cross section S is obtained from

$$S=\sum_f E_f\sigma^f=(z_1^2+z_2^2)S_p+2z_1z_2S_v, \tag{4.7a}$$

where

$$S_v=\sum_f E_f\sigma_v^f(R) \tag{4.7b}$$

and S_p is the total stopping cross section of the target atom for a proton traveling with velocity v. The sum over E_f is understood to include an in-

tegral over continuum values of E_f. Appealing to the sum rule $\sum_f E_f |F_{fi}(q)|^2 = Z_2\hbar^2 q^2/2m$ and its small-q limit $\sum_f E_f |(x)_{fi}|^2 = Z_2\hbar^2/2m$, we may write

$$S_v = 2\pi\left[\frac{2e^2}{\hbar v}\right]^2\left[\sum_f E_f |(x)_{fi}|^2 \int_{E_f/\hbar v}^{q_I} \frac{dq}{q}\frac{\sin qR}{qR} + \frac{\hbar^2 Z_2}{2m}\int_{q_I}^{2mv/\hbar}\frac{dq}{q}\frac{\sin qR}{qR}\right]. \tag{4.8}$$

The dipole matrix element is

$$(x)_{fi} \equiv \langle f | \sum_{j=1}^{Z_2} x_j | i \rangle .$$

The finite upper limit in the second integral on the right-hand side of Eq. (4.8) originates from the fact that $|F_{fi}(q)|^2$ exhibits a "Bethe ridge" in the q-E_n plane for large q such that a negligible contribution to this term occurs for q larger than $2mv/\hbar$.[22] Then replacing E_f in the lower limit of the integral on the right-hand side of Eq. (4.8) by an average value $\bar{E}_f$, using the dipole sum rule to evaluate the term, and combining the two integrals, we find

$$S_v = \frac{4\pi Z_2 e^4}{mv^2}\left[\mathrm{si}_2\left[\frac{\bar{E}_f R}{\hbar v}\right] - \mathrm{si}_2\left[\frac{2mvR}{\hbar}\right]\right], \tag{4.9}$$

where $\bar{E}_f$ is defined by the equation

$$\mathrm{si}_2\left[\frac{\bar{E}_f R}{\hbar v}\right] = \frac{2m}{Z_2\hbar^2}\sum_f E_f |(x)_{fi}|^2 \mathrm{si}_2\left[\frac{E_f R}{\hbar v}\right].$$

We may take $\bar{E}_f \approx \bar{E}_v$, the mean excitation energy of valence electrons in the atom, and set $Z_2 \to Z_v$, where Z_v is the number of valence electrons in the atom. Deeper-lying shells typically have binding energies considerably larger than E_v and so will make negligibly small contributions to S_v for values of R presently obtainable in clusters. The similarity in form between Eqs. (3.13) and (4.9) is clear. When $R \to 0$, S_v approaches S_p, the stopping power of the atom for a proton with velocity v. When $R >> \hbar/2mv$, the de Broglie wavelength of an electron viewed in the frame of the moving projectile, the second term in Eq. (4.9) may be neglected compared with the first. Then further, if $\bar{E}_f R/\hbar v >> 1$,

$$S_v(R) = \frac{4\pi e^4 Z_2}{mv^2}\frac{\cos(\bar{E}_f R/\hbar v)}{(\bar{E}_f R/\hbar v)^2} . \tag{4.10}$$

We now consider the cross section σ_i for ionization of a particular shell of electrons of the atom by randomly oriented clusters. It may be obtained by integrating Eq. (4.4) over all continuum energies, after averaging over the orientation of $\vec{R}$ as in Eq. (4.6). Thus,

$$\sigma = \int d\sigma = (z_1^2 + z_2^2)\sigma_p^I + 2z_1 z_2 \sigma_v^I(R) , \tag{4.11}$$

where σ_p^I is the cross section for ionization of the shell by a proton[23] with velocity v and

$$\sigma_v^I(R) = 2\pi\left[\frac{2e^2}{\hbar v}\right]^2 \times \int dE_f \int_{q_0}^{\infty}\frac{dq}{q^3}\left[\frac{\sin qR}{qr}\right]|F_{fi}(q)|^2 \tag{4.12}$$

is the vicinage term in the total cross section for a diproton cluster. Again, we may split the integration over q into an interval from q_0 to q_I, in which the dipole approximation to $F_{fi}(q)$ is acceptable, and a region from q_I to infinity. In the latter range it is not necessary to use a finite upper limit, since for large v the contribution from this region is expected to be quite small compared with that from the interval $q_0 \le q \le q_I$. Thus

$$\sigma_v^I(R) = 2\pi\left[\frac{2e^2}{\hbar v}\right]^2 \int dE_f \left[Z_2 |(x)_{fi}|^2 \int_{q_0}^{q_I}\frac{dq}{q}\frac{\sin qR}{qR} + \int_{q_I}^{\infty}\frac{dq}{q^3}\frac{\sin qR}{qR}|F_{fi}(q)|^2\right]. \tag{4.13}$$

We may obtain an approximate value for the second integral by assuming that in the region $q_I < q$, the form factor is peaked about the Bethe ridge such that $|F_{fi}|^2 \approx \delta[E_f - (\hbar^2/2m)q^2]$. Then we may write

$$\sigma_v^I(R) = 8\pi\left[\frac{e^2}{\hbar v}\right]^2\left\{\mathcal{M}^2\left[\mathrm{si}_2\left[\frac{\bar{\omega}_f R}{v}\right] - \mathrm{si}_2(q_I R)\right] + \int_{q_I}^{\infty}\frac{dq}{q^3}\frac{\sin qR}{qR}\right\}, \tag{4.14}$$

where $\hbar\bar{\omega}_f = \bar{E}_f$ is a mean value of E_f such that

$$\mathcal{M}^2 \mathrm{si}_2\left[\frac{\bar{\omega}_f R}{v}\right] = \int dE_f \,|(x)_{fi}|^2 \,\mathrm{si}_2\left[\frac{E_f R}{\hbar v}\right] \tag{4.15a}$$

and

$$\mathcal{M}^2 = \int dE_f \,|(x)_{fi}|^2 \tag{4.15b}$$

for optically allowed transitions to final states. To proceed further it is necessary to make specific assumptions about the form of $|(x)_{fi}|^2$. It is clear again, however, that a dependence of σ_v^I on R which has the general character of $\mathrm{Si}_2(\bar{\omega}R/v)$ would be found. The limiting case $R \to 0$ yields, for the large-v limit,

$$\sigma_v^I(0) = 8\pi\left[\frac{e^2}{\hbar v}\right]^2 \mathcal{M}^2 \ln\left[\frac{q_I v}{\bar{\omega}_f}\right], \tag{4.15c}$$

which has the form of the Bethe asymptotic formula for the total inelastic cross section for excitation of the given shell by a proton.

In order to obtain detailed information about the vicinage effect in atomic collisions, it is necessary to evaluate Eq. (4.5c) or Eq. (4.12) by numerical integration, employing specific representations of the form factor $|F_{fi}(q)|^2$. Figure 8 shows the result of such an evaluation in which we have used an analytical form factor[23] describing the excitation of an electron from a hydrogenic s-orbital to hydrogenic continuum final states. The quantity $g(R) = \sigma_v^I/\sigma_p^I$, where σ_v^I is defined in Eq. (4.12) and σ_p^I is the cross section for ionization by a proton. We have taken the ionization energy $\hbar\omega = 72$ eV and $v = 6.28 v_0$, corresponding to an ion energy of 1.975 MeV/amu. In terms of the variables in Ref. 23, the constant η, the square of the scaled projectile velocity, was taken to be 0.5, and the value 0.067 was used for θ, the scaled ionization energy. Again, one sees a dependence of $g(R)$ on R similar to that found for the electron gas and for the classical harmonic oscillator.

We have also employed atomic form factors, or generalized oscillator strengths (GOS) values,[24] to evaluate $g(R)$ for the $2s$-orbital-to-continuum and the $2p$-orbital-to-continuum transitions of the aluminum atom. The results are shown in Figs. 9 and 10, respectively. Figure 9 shows the vicinage function $g(R)$ vs R for transitions from the $2s$ shell and for four different values of v. The ionization energy of electrons in this shell is taken equal to 118.6 eV. Figure 10 shows similar data for the $2p$ shell in which the ionization energy is 80.9 eV. All the curves in Figs. 9 and 10 may be scaled, to a good approximation, into a single curve by employing as independent variable the quantity $R\bar{\omega}/v$, where $\hbar\bar{\omega}$ is an energy of the order of the ionization energy of the shell. Each of the curves in both of these figures is described approx-

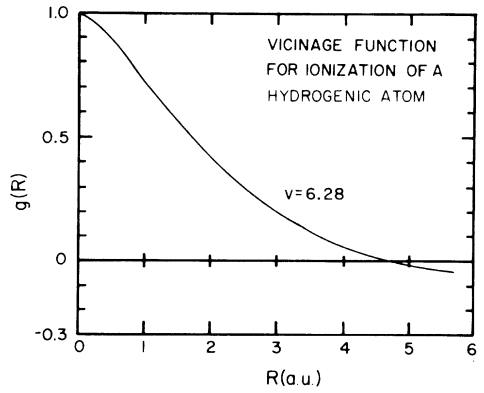

FIG. 8. Vicinage function for ionization of a hydrogenic atom, plotted as a function of interionic separation R and for projectile velocity $v = 6.28$ a.u. The parameters $\eta = 0.5$ and $\theta = 0.067$ were chosen for these calculations.

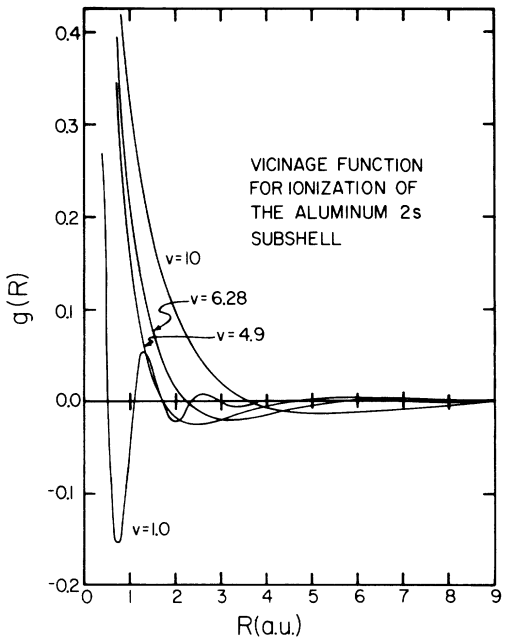

FIG. 9. Plots of the vicinage function $g(R)$ vs R for ionization of the Al $2s$ subshell for different ion velocities v (a.u.). These data were plotted using numerically specified values (Ref. 24) of the atomic GOS for ionization of this level.

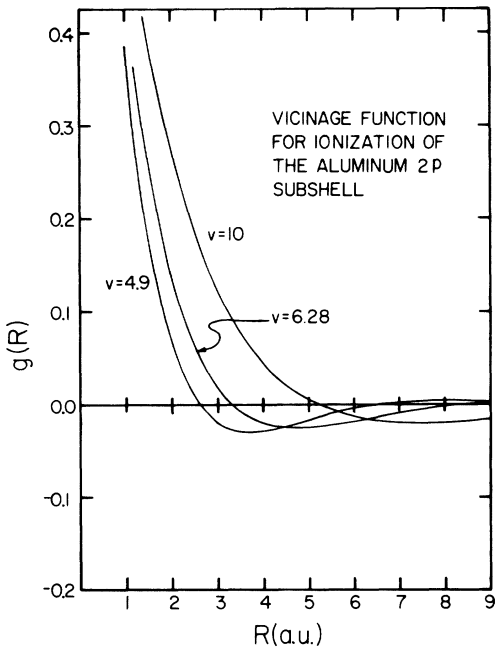

FIG. 10. Plot of the vicinage function for ionization of the Al $2p$ subshell. The data of Ref. 24 were used in obtaining these values.

imately by the expression of the form given in Eq. (4.14) divided by σ_v^I, Eq. (4.15c).

V. VICINAGE EFFECTS IN INNER-SHELL IONIZATION

The search by Lurio, Andersen, and Feldman[12] (LAF) for cluster effects in the production of inner-shell vacancies was unsuccessful; no such effects were observed within experimental error ($\sim 1\%$). Although their experiments were carried out with solid targets, the adiabatic distance v/ω, where $\hbar\omega$ is the relevant excitation energy of the inner-shell electron, was small compared with lattice dimensions. Hence, collective effects between atoms on different lattice sites should be small and atomic models of these excitations should be appropriate.

If the vicinage effect is to be significant in an experiment, the considerations above lead to the requirement that the parameter $\bar{R}\bar{\omega}/v \lesssim 1$, where $\bar{R}$ is a mean interionic distance during the Coulomb explosion, v the ionic velocity, and $\hbar\bar{\omega}$ an effective excitation energy for the level or levels involved. In the experiments of LAF, the values of $\bar{R}\bar{\omega}/v$, appropriate for the various bombarding energies and the several target materials and x-ray energies, were, with a single exception, appreciably greater than 1, if $\bar{R}\bar{\omega}/v$ is estimated by taking $\hbar\bar{\omega}$ equal to the observed x-ray line energy, v to be the entrant velocity of the ion and $\bar{R}$ the mean initial separation of protons after stripping (~ 1.29 Å). The exception occurs in a measurement corresponding to the excitation of L electrons in Al by 1.975 MeV H_2^+ ions. For this case $\bar{R}\bar{\omega}/v \approx 1.05$. This estimate is expected to be low since it does not take into account the continuous increase in R following stripping in the film. However, for the experiments of LAF this amounts to only a small correction. The vicinage term $g(1.05)$ estimated from Fig. 7 for $x = 2mv^2/\hbar\omega \sim 29.2$, corresponding to the experiments of LAF, has the value ~ 0.16. A much better estimate can be found from Fig. 10 which is calculated from theoretical values of the GOS for excitation from the $2p$ shell of the aluminum atom. One finds $g(2.44) \sim 0.07$ for $R = 1.29$ Å and $v = 6.28 v_0$, yielding a vicinage effect which exceeds by a factor of ~ 7 that which would have been experimentally detectable. Note that for a proton dicluster $z_1 = z_2 = 1$ and the vicinage function for ionization is

$$[\sigma_{di}(R) - \sigma_{di}(\infty)]/\sigma_{di}(\infty) = \frac{2z_1 z_2}{z_1^2 + z_2^2} g_v^I(R) = g_v^I(R) , \tag{5.1}$$

where $\sigma_{di}(R)$ is the cross section for ionization by a dicluster with initial separation R, Eq. (4.11).

An explanation for the discrepancy between the LAF results and the theory described above may be found by noting that information relevant to excitation of the Al L shell was inferred by LAF from a study of the high-energy satellite of the K_α line. The energy of the normal K_α line from Al is ~ 1490 eV. A high-energy satellite of this emission is a line at ~ 1500 eV and is thought to originate from a double ionization process in which a projectile causes vacancies in both the K and L shells. The energy required to ionize the K shell when an L vacancy exists is somewhat larger than when the L electron is present. In order for a swift ion to eject both K and L electrons, it is necessary for the ion to make a small impact-parameter collision with the atom, i.e., it should pass within the K-orbital radius if there is to be appreciable probability for the double process.[25] This means, of course, that the ion should pass close to the center of the L-electron cloud. Such a collision tends to give rise to very energetic contin-

uum final states of the L electron. Qualitatively speaking, the probability of such transitions is expected to be insensitive to the presence of the other ion in a dicluster as long as R is appreciably greater than the L-shell radius.

This is confirmed in an approximate calculation of $g(R)$ for the case of a swift dicluster incident on an atomic system such that one of the ions is constrained to pass directly through the center of the atom, while the other ion assumes random orientation at constant distance from the first. This may be done by setting $\vec{b}_1=0$ in Eq. (4.2) and approximating the final-state vector $\langle \vec{r}\,|f\rangle$ by a plane wave, viz., $\langle \vec{r}\,|f\rangle \sim \exp\{i\vec{k}\cdot\vec{r}\}/\Omega^{1/2}$, where Ω is the normalization volume. The initial state $\langle \vec{r}\,|i\rangle$ is taken proportional to $e^{-\alpha r}$. For the probability amplitude of this transition, we find

$$a_f=\frac{4\sqrt{\pi}i(Z_2^*)^{3/2}}{v\Omega^{1/2}}\frac{d}{d\alpha}\times\int_0^1\frac{du}{\Lambda^2(u)}[z_1+z_2e^{-i\vec{R}\cdot\vec{A}}\Lambda R K_1(\Lambda R)]\,, \tag{5.2}$$

where

$$\Lambda^2(u)=\left[[\alpha^2+k^2(1-u)]u+\left[\frac{\omega}{v}-\vec{k}\cdot\vec{v}u/v\right]^2\right], \tag{5.3}$$

$$\vec{A}=u\vec{k}+(\omega-u\vec{k}\cdot\vec{v})\vec{v}/v^2\,, \tag{5.4}$$

and $\omega=\omega_0+k^2/2$ is the difference in energy between the final and the initial state. Atomic units are used throughout this section. After differentiation, α is set equal to Z_2^*, where $(Z_2^*)^{-1}$ is the effective screening length of the initial-state orbital. $K_1(x)$ is the modified Bessel function of first order. The first integral may be carried out exactly. The probability of the specified double ionization is obtained by summing $|a_f|^2$ over all final states and averaging over directions of $\vec{R}$. After some algebra, we find the approximate expression

$$P_0(z_1,z_2)=\frac{32}{\pi v^2\alpha^3}\int_0^\infty k^2\left[A^2+B^2-4AB\left[\frac{v}{R\omega}\right]\sin\left[\frac{R\omega}{v}\right]\right]dk\,, \tag{5.5}$$

where

$$A=z_1\left[\frac{v^2\alpha^2}{v^2d+\omega^2}-\ln\left[\frac{v^2\alpha^2+\omega^2}{\omega^2}\right]\right],$$

$$B=z_2\frac{R^2\alpha^4}{(R\alpha^2+2\omega/v)^2}\left[\frac{2\pi v}{R\omega}\right]^{1/2}e^{-\omega R/v}.$$

If we take the effective atomic number of the L shell in Al to be $Z_2^*\approx 8.85$ from Slater's rules, $\omega_0=2.65$ a.u., $v=6.28v_0$ to correspond to the experiment of LAF and $R=1.29$ Å,[3] we find the vicinage effect to be measured by the ratio

$$R\equiv\frac{P_0(1,1)}{P_0(1,0)}\equiv 1+\delta=1+0.045\,,$$

which is to be compared with the data of LAF, who find $\delta\approx -0.019\pm 0.02$ and $\delta\approx -0.089\pm 0.058$ in experiments with two different foil thicknesses. We note that the calculation leading to Eq. (5.5) is intended to illustrate only the qualitative dependence of P_0 on the parameters of that equation. It predicts that the vicinage term depends on the ratio $(a_L/R)^2$, the square of the ratio of the L-shell radius to the cluster separation, and on $R\omega_0/v$. One expects this to be confirmed in a more accurate calculation. In view of the approximate character of our Eq. (5.5), the comparison with experiment seems reasonable.

To recapitulate, in order to observe vicinage effects in inner-shell excitations, one must carry out experiments at large enough projectile velocities or small enough excitation energies $\hbar\bar{\omega}$ so that $\bar{R}\bar{\omega}/v\lesssim 1$. It appears that the single-excitation experiments which have been carried out to date do not satisfy this criterion. The multiexcitation experiment of LAF, although satisfying this criterion, does not determine the vicinage effect in single-particle excitation. Further experimental work at larger bombarding energies and, consequently, larger v, would be very desirable.

VI. FLUCTUATIONS IN CLUSTER ENERGY LOSS

The vicinage effect discussed above is represented mathematically by a statistical average of a quantity subject to quantum fluctuations. A measure of such fluctuations for a cluster may be

found by computing Ω, the variance of energy loss about the mean loss.

As Niels Bohr has shown,[21(b)] if dE is the energy loss experienced by a charged entity in traversing an infinitesimal thickness of a target, then $\Omega^2=\langle(dE)^2\rangle$, where the brackets indicate an average over the probability distribution of energy losses.

The appropriate probability distribution of energy losses for the case of clusters penetrating a valence electron gas may be obtained from Eq. (2.1) for the oriented dicluster, or from Eq. (2.7) in the case of the spherically averaged cluster. Each of these equations describes the mean energy loss per unit path length and is written as an integral over frequency ω. To reach an expression for the distribution of energy losses, we must recognize that the quantum of energy corresponding to the frequency ω is $\hbar\omega$. Then we may write for $\langle dP/d\omega\rangle$, the probability that the spherically averaged dicluster should lose a quantum of frequency ω, while traversing the path length dR,

$$\left\langle \frac{dP}{d\omega} \right\rangle = dR \left\{ (z_1^2+z_2^2)\frac{2e^2}{\pi\hbar v^2}\int_{\omega/v}^{\infty}\frac{dk}{k}\,\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right] + \frac{4z_1z_2e^2}{\pi\hbar v^2}\int_{\omega/v}^{\infty}\frac{dk}{k}\left[\frac{\sin kR}{kR}\right]\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right]\right\} . \tag{6.1}$$

Note that the second term in Eq. (6.1) may be negative under some conditions. This is related to the fact that the wake of the leading ion may deliver energy to the trailing ion. The first term of Eq. (6.1) originates from the reaction of each wake on its causative ion. The second term describes the distribution of energy loss (or gain) of one ion due to the wake of the other. We take into account the fact that contributions to the straggling from these two mechanisms of energy transfer must be statistically independent of one another by writing $\Omega^2=(z_1^2+z_2^2)\Omega_s^2+2z_1z_2\Omega_v^2$, where

$$\Omega_s^2=\frac{2\hbar e^2}{\pi v^2}\int_0^{\infty}\omega^2 d\omega\int_{\omega/v}^{\infty}\frac{dk}{k}\,\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right]dR \tag{6.2}$$

is the straggling parameter of a proton and

$$\Omega_v^2=\frac{2\hbar e^2}{\pi v^2}\int_0^{\infty}\omega^2 d\omega\int_{\omega/v}^{\infty}\frac{dk}{k}\frac{\sin kR}{kR}\,\mathrm{Im}\left[\frac{-1}{\epsilon(k,\omega)}\right]dR \tag{6.3}$$

originates in the vicinage effect. We may evaluate Ω_s^2 using a model dielectric function related to that employed in obtaining Eq. (2.2), viz.,

$$\mathrm{Im}[-\epsilon^{-1}(k,\omega)]=\frac{\pi\omega_p^2}{2\omega_k}\delta(\omega-\omega_k) ,$$

where

$$\omega_k=(\omega_p^2+\hbar^2k^4/4m^2)^{1/2} .$$

We find

$$\begin{aligned}\Omega_s^2/dR &= \frac{\hbar e^2\omega_p^2}{v^2}\int_{\omega_p/v}^{2mv/\hbar}\frac{\omega_k}{k}dk \\ &= \frac{\hbar e^2\omega_p^3}{2v^2}\left[(1+y)^{1/2}-\left[1+\frac{1}{y}\right]^{1/2}-\ln\left\{[(1+y)^{1/2}+1]\left[\left[1+\frac{1}{y}\right]^{1/2}-1\right]\right\}\right] ,\end{aligned} \tag{6.4}$$

where $y=(2mv^2/\hbar\omega_p)^2$. In the limit $y>>1$,

$$\Omega_s^2/dR\approx 4\pi e^4 n , \tag{6.5}$$

where $n_0=m\omega_p^2/4\pi e^2$ is the electron density in the medium. The approximation of Eq. (6.5) agrees with a result obtained by Bohr[21(b)] on the basis of an atomic model. An expression for Ω_v^2 may be found by a similar procedure; we obtain

$$\frac{\Omega_v^2}{dR}=\frac{\hbar e^2\omega_p^2}{v^2}\left[\omega_P\left\{\mathrm{si}_2\left[\frac{\omega_p R}{v}\right]-\mathrm{si}_2\left[\left[\frac{2m\omega_p}{\hbar}\right]^{1/2}R\right]\right\}+\frac{\hbar}{2mR^2}\left\{\cos\left[R\left[\frac{2m\omega_p}{\hbar}\right]^{1/2}\right]-\cos\left[\frac{2mvR}{\hbar}\right]\right\}\right]. \tag{6.6}$$

This approximate expression is valid for $v\gtrsim(\hbar\omega_p/m)^{1/2}$.

Numerical evaluation of Eqs. (6.4) and (6.6) for $v=3v_0$, $\hbar\omega_p=24.5$ eV, and $R=1.3$ Å yields $\Omega_s^2/dR=0.81$ a.u. $=1.133\times10^3$ (eV)^2/Å and $\Omega_v^2/dR=0.064$ a.u. $=89.9$ (eV)^2/Å. Thus the straggling parameter Ω^2 for cluster energy loss differs only by about 10% from that for the isolated ions at separations commonly encountered. We expect this to be true for collision with single atoms as well. The smallness of Ω_v^2/Ω_s^2 is due to the fact that the long-range part of the wake, which is the major contributor to the vicinage effect at representative values of R, is associated with rather small energy and momentum transfers to the wake. Thus the wake-trailing ion interactions are expected to be gentle ones.

VII. THE EFFECT OF QUANTAL COHERENCE ON CLUSTER INTERACTIONS

In the foregoing we have assumed that the ions of a cluster are classical point charges moving on deterministic trajectories. In part this is justified by the fact that ionic masses exceed the electron mass by several orders of magnitude. Thus in the course of a collision with an electron, quantal uncertainty of ionic motion may be neglected to a very good approximation, and the effect of a given ion on the electronic system may be calculated as if the ion were executing classical motion. Following this idea we have computed vicinage effects above for two point charges at fixed separations, averaging over orientation to correspond approximately to conditions of cluster bombardment in which ion clusters are incident on a target with no selection as to the direction of their figure axis: this is the usual experimental situation.

In fact, molecular ions used in experiment are expected to be in various states of rotational and vibrational motion when they arrive at a target. Upon losing valence electrons, the ions undergo Coulomb explosion: The wave function describing relative motion must expand and disperse as time increases after stripping. One may compute the time evolution of the wave function in a standard way by computing the projection of the initial wave packet on eigenstates of relative motion in a repulsive Coulomb potential. This Coulomb interaction may be modified appreciably in a condensed medium due to the dynamic properties of the system. The sum of these projection coefficients, each multiplied by the corresponding Coulomb eigenfunction and weighted by the factor $\exp\{-iE_kt/\hbar\}$, where E_k is the energy of an eigenstate of the Coulomb potential corresponding to the quantum index k in center-of-mass coordinates, gives the wave function at time t, if dynamic screening by the medium may be neglected.

We do not enter into the details of this time evolution but instead focus on structure effects at a given instant of time. Velocities attained during ionic explosion are quite small compared with the center-of-mass velocity in all cases of experimental interest.

For concreteness we consider a dicluster with constituent ions that may be of charge, mass, and coordinate $(z_1e,M_1,\vec{r}_1)$ and $(z_2e,M_2,\vec{r}_2)$, respectively. We represent it by the state vector

$$\langle R,\vec{r}\,|\,i\rangle=\frac{e^{i\vec{\kappa}\cdot\vec{R}}}{\sqrt{\Omega}}u_0(\vec{r}),$$

where $\hbar\vec{\kappa}$ is the center-of-mass momentum, $\vec{R}$ is the coordinate of the center of mass, and $u_0(\vec{r})$ describes the wave function of internal motion of the dicluster in the initial state. The normalization volume of the system is designated by Ω. After interaction, a target atom is excited from the state described initially by the state vector $|0\rangle$ to the final state represented by $|n\rangle$. We assume that the final wave function may be written as a product of a momentum eigenfunction, that describes motion in the center of mass of the two ions, and an eigenfunction in the relative coordinate $\vec{r}$, that is a solution, corresponding to energy E_f, of Schrödinger's equation with a repulsive Coulomb potential:

$$\langle\vec{R},\vec{r}\,|\,f\rangle=\frac{e^{i\vec{\kappa}_f\cdot\vec{R}}}{\sqrt{\Omega}}\phi_f(\vec{r}).$$

We take the energy of interaction between the

dicluster and the atom to be given by

$$V=\sum_{j=1}^{Z_2}\left[\frac{-z_1e^2}{\left|\vec{R}+\frac{M_2}{\mu}\vec{r}-\vec{\rho}_j\right|}+\frac{-z_2e^2}{\left|\vec{R}-\frac{M_1}{\mu}\vec{r}-\vec{\rho}_j\right|}\right], \tag{7.1}$$

where $\vec{\rho}_j$ is the coordinate of the jth electron in the atom and $\mu=M_1+M_2$.

In Golden Rule perturbation theory, the cross section for the specified excitation may be written

$$\sigma_{n0}=\frac{\Omega}{v}\frac{2\pi}{\hbar}\sum_f |\langle f,n|V|0,i\rangle|^2\delta(\epsilon_i-\epsilon_f), \tag{7.2}$$

where ϵ_i and ϵ_f are the initial and final energies of the total system, respectively, and $v=\hbar\kappa/\mu$. Using the Bethe integral form of the plane-wave matrix element of the Coulomb interaction, we may write

$$\sigma_{n0}=\frac{32\pi^3e^4}{\hbar^2v\Omega}\sum_{\vec{q}}\sum_f\frac{|F_{n0}(\vec{q})|^2}{q^4}\left|\int d^3r\phi_f^*(\vec{r})\left[z_1e^{-i(M_2/\mu)\vec{q}\cdot\vec{r}}+z_2e^{i(M_1/\mu)\vec{q}\cdot\vec{r}}\right]u_0(\vec{r})\right|^2 \times\delta\left[\omega_{n0}-\frac{1}{\hbar}(\bar{E}_0-E_f)-\vec{v}\cdot\vec{q}+\frac{\hbar q^2}{2\mu}\right]. \tag{7.3}$$

In Eq. (7.3) $\omega_{n0}=\epsilon_n-\epsilon_0$ is the transition energy of the target atom in being excited from the ground state to the nth excited state, and $\bar{E}_0$ is the energy of the dicluster in the initial wave-packet state. The form factor $F_{n0}(\vec{q})$ is specified in Eq. (4.3). We now argue that the wave packet $u_0(\vec{r})$ is expected to have a spatial extension corresponding to atomic dimensions, e.g., 1 Å. Thus its momentum transform should be quite small for momenta $\vec{k}_f$ much larger than 1 Å^{-1}. Since E_f, the eigenenergy of the repulsive Coulomb eigenfunction, should have the form $E_f\simeq\hbar^2k_f^2/2\mu$ when k_f is large, it should be an excellent approximation to neglect both E_f and the term $\hbar q^2/2\mu$ in the argument of the δ function in Eq. (7.3). This neglect allows us to carry out the sum over f using closure to obtain

$$\sigma_{n0}=\frac{4e^4}{\hbar^2v}\int d^3r|u_0(\vec{r})|^2\int\frac{d^3q}{q^4}|F_{n0}(\vec{q})|^2|z_1+z_2e^{-i\vec{q}\cdot\vec{r}}|^2\delta(\omega_{n0}-\vec{v}\cdot\vec{q}-E_0). \tag{7.4}$$

The sum over q has been converted to an integral in the limit $\Omega\to\infty$ and an irrelevant factor of $e^{-i(M_2/\mu)\vec{q}\cdot\vec{r}}$ has been extracted from the absolute-squared expression.

This interesting result is in accord with the semiclassical picture that during the early expansion phase of a dicluster, before it has had time to expand appreciably, its cross section for exciting a transition in a given system may be calculated by averaging the cross section for excitation of that system by a classical dicluster with separation $\vec{r}$ over the probability density $|u_0(\vec{r})|^2$ that the dicluster is found with that separation.

It is straightforward to generalize this approach to apply to the calculation of the energy loss of a molecular dicluster penetrating a solid target. If one writes for $\mathscr{S}_c(B,D)$, the energy loss of a classical point dicluster with separation D along the direction of motion and separation B perpendicular to that direction, then the energy loss of the quantally coherent cluster may be written as

$$\langle\mathscr{S}_c\rangle=\int d^3r|u_0(\vec{r})|^2\mathscr{S}_c(r\sin\theta,r\cos\theta), \tag{7.5}$$

where θ is the angle between $\vec{r}$ and the direction of motion and where $\mathscr{S}_c$ is given by Eq. (2.1).

We note that the great disparity in mass between electrons making up the medium and the ions of the dicluster results in nearly complete decoupling between the cluster explosion and energy loss. We then expect it to be an excellent approximation to generalize Eq. (7.4) to yield a cross section $\sigma_{n0}(t)$ at time t following the beginning of the cluster explosion. This time dependence is obtained by replacing $|u_0(\vec{r})|^2$ by the time-dependent probability density $|\sum_f\langle f|u_0\rangle\phi_f(r)e^{-iE_ft/\hbar}|^2$. The resulting expression could be averaged over the time of adiabatic expansion and should be valid until times such that elastic nuclear scattering has destroyed the coherence of the exploding wave packet.

To illustrate the magnitude of this molecular

coherence effect, we have averaged the vicinage function $g \equiv g(R\omega/v)$, defined immediately below Eq. (3.13) and plotted in Fig. 7, over the squared wave function describing the internuclear separation of the hydrogen molecular ion[26] in various states of vibration and excitation. The populations of the vibrational states were taken from the Franck-Condon—type calculations of Walters *et al.*[27] The results of this averaging are shown in Fig. 11. The solid curve labeled $\langle g \rangle$ was computed as described above for an electron density corresponding to a plasma energy of 23.2 eV (equivalent to an amorphous carbon medium) and is plotted as a function of the speed v of the cluster in atomic units. For comparison we show as the dashed curve the vicinage function as a function of v evaluated at the mean internuclear separation, also calculated from the wave functions given in Ref. 26. The dot-dash curve shows the vicinage function evaluated at the rms internuclear separation.

It is seen that the more realistic calculation of the mean vicinage function gives larger values than the function evaluated at either of the mean separations. For $v \gtrsim 2$ a.u. the differences are $\sim 15\%$ but can become considerably larger when v is smaller.

It seems clear that if the time evolution of the wave packet representing the cluster were known, one should be able to compute vicinage effects with reasonable accuracy by averaging the appropriate vicinage function over the time-dependent probability of finding the cluster separation in d^3r at $\vec{r}$.

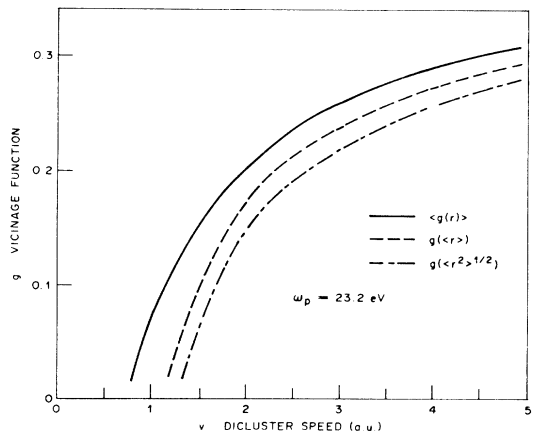

FIG. 11. Graphs of the vicinage function g vs cluster speed v averaged over the probability distribution corresponding to that of hydrogen molecular ions emerging from an ion source (Ref. 27). For comparison the vicinage function evaluated at the mean internuclear separation and at the rms internuclear separation are shown also.

VIII. DISCUSSION

Theoretical expressions for energy loss of ion clusters in condensed matter and in collision with atoms at condensed-matter densities show strong similarity when the atomic transitions induced by the cluster are of dipole character (optically allowed). It appears that the mathematical process of summing energy transfer over the impact parameter of a projectile with respect to an atom is equivalent to building up a continuous dielectric medium of noninteracting oscillators in which the projectile may be considered to travel. Thus a formal resemblance exists between the results for energy loss to an atom and to a dielectric medium.

However, one would not expect the cluster alignment effect, observed in solids by Gemmell and co-workers,[8] to manifest itself in collisions of clusters with atoms in gases at ordinary pressures. Indeed, no alignment effect could be found in experiments on molecular ions bombarding gas targets.[28] The origin of this pronounced difference lies in the fact that the force on a trailing ion due to the wake of a leading ion in a solid may be thought of as a coherent superposition of displacement fields associated with density fluctuations in the medium and is directed toward the track of the leading ion when $D \lesssim v/\omega_p$. In the case of a cluster collision with a single atom, the force on a trailing ion depends on the impact parameters of the ions ($\vec{b}_1$ and $\vec{b}_2$ of Sec. III above) relative to the atom, as one may see by considering the electric force on the trailing ion due to a dipole moment induced in the atom by the leading ion. This force is not, in general, such as to align the trailing ion when $D \lesssim \pi v/\omega$, where $\hbar\omega$ is the excitation energy of the atom. Thus, unless a large number of collisions with gas atoms or molecules occur during the period after a cluster is stripped and before it has expanded appreciably in a Coulomb explosion, there will not be a net force tending to align the trailing ion behind the leading one.

We now estimate the gas density sufficient to ensure that alignment can occur when $D \lesssim \pi v/\omega$. During a time t_e in which Coulomb explosion causes the ions of a dicluster to change their relative separation from R_0 to $(1+f)R_0$, the dicluster will travel a distance vt_e. The number of gas atoms passing the leading ion within the adiabatic

distance, v/ω, during this time can be written as

$$\pi\left[\frac{v}{\omega}\right]^2 v t_e n \ ,$$

where n is the density of gas atoms. This number should be greater than N, the minimum number of atoms in collision needed to cause cancellation of nonaligning forces to acceptable variance. We choose $f << 1$ to restrict the averaging process to times in which the effects of Coulomb explosion are small. This results in the following criterion for the gas density n:

$$n > \frac{N}{\pi}\frac{\omega^2}{v^3}\left[\frac{z_1 z_2 e^2}{2fMR_0^3}\right]^{1/2} \ ,$$

where z_1 and z_2 are the ion charges and M is the reduced mass of the dicluster. The time t_e is calculated assuming, for simplicity, that acceleration in Coulomb explosion is constant over the time interval considered. If we choose $N=10^3$, $\hbar\omega=14$ eV, $z_1=z_2=1$, $R_0=1$ Å, $v=5v_0$, and $f=0.1$, we find that n should be greater than $0.017/a_0^3$, which is typical of condensed-matter densities.

Thus we estimate that in order for alignment to occur in dicluster collisions with gas atoms when $D \lesssim \pi v/\omega$, the density of the gas should probably be of the order of that occurring in condensed matter.

ACKNOWLEDGMENTS

The authors are grateful for helpful conversations with Werner Brandt and other participants in the Informal Workshop on Wake Phenomena, held at New York University, and to L. C. Feldman and Pedro Echenique for discussions in this connection. We thank Roger Rice for assistance in the calculations done for Fig. 8 and V. E. Anderson for his work in obtaining the vicinage functions displayed in Figs. 2–4 above. This research was sponsored in part by the Office of Health and Environmental Research, U. S. Department of Energy, under Contract No. W-7405-ENG-26 with the Union Carbide Corporation.

*Present address: Physical Review Letters, Box 1000, Ridge, N. Y. 11961

[1]V. N. Neelavathi, R. H. Ritchie, and W. Brandt, Phys. Rev. Lett. 33, 302 (1974); 33, 670(E) (1974); 34, 560(E) (1975). See, also, V. N. Neelavathi and R. H. Ritchie, in *Proceedings of Conference on Atomic Collisions in Solids,* edited by S. Datz, B. R. Appleton, and C. D. Moak (Plenum, New York, 1975), p. 289.

[2]R. H. Ritchie, W. Brandt, and P. M. Echenique, Phys. Rev. B 14, 4808 (1976), have studied the wake in more detail than was done in Ref. 1.

[3]W. Brandt, A. Ratkowski, and R. H. Ritchie, Phys. Rev. Lett. 33, 1325 (1974); 35, 130(E) (1975). A review of these topics has been given by W. Brandt and R. H. Ritchie, Nucl. Instrum. Methods 132, 43 (1976).

[4]J. W. Tape, W. H. Gibson, J. Remillieux, R. Laubert, and H. E. Wegner, Nucl. Instrum. Methods 132, 75 (1976).

[5]J. C. Eckardt, G. Lantschner, N. R. Arista, and R. A. Baragiola, J. Phys. C 11, L851 (1978).

[6]N. R. Arista and V. H. Ponce, J. Phys. C 8, L188 (1975); N. R. Arista, Phys. Rev. B 18, 1 (1978).

[7]R. Laubert, IEEE Trans. Nucl. Sci. 26, 1020 (1979).

[8]D. S. Gemmell, J. Remillieux, J.-C. Poizat, M. J. Gaillard, R. E. Holland, and Z. Vager, Phys. Rev. Lett. 34, 1420 (1975). See also Z. Vager, D. S. Gemmell, and B. J. Zabransky, Phys. Rev. A 14, 638 (1976). See also the series of recent papers by Gemmell, Vager, and co-workers, in The Proceedings of the 8th International Conference on Atomic Collisions in Solids, Hamilton, Canada, August 1979 [Nucl. Instrum. Methods 170, 41–99 (1979)].

[9]Earlier workers studying the channeling of molecular ions [T. A. Tombrello and J. M. Caywood, Phys. Rev. B 8, 3065 (1973)] recognized the effect of the Coulomb explosion on alignment properties but did not discuss the role of the wake of density fluctuations in this connection. Also, A. D. Bacher, E. A. McClatchie, M. S. Zisman, T. A. Weaver, and R. A. Tombrello, Nucl. Phys. A181, 453 (1972), showed that momentum gained by the fragments in a cluster explosion accounts for the spread with energy in yield curves of protons scattered in the excitation of the 14-MeV resonance in the $^{12}C(p,p)^{12}C$ reaction. Golovchenko and Laegsgaard [Phys. Rev. A 9, 1215 (1974)] measured the energy and angular distribution of protons transmitted through a thin foil when 2-MeV H_2^+ ions are incident.

[10]Z. Vager and D. S. Gemmell, Phys. Rev. Lett. 37, 1352 (1976); A. Faibis, R. Kaim, I. Plesser, and Z. Vager, Nucl. Instrum. Methods 170, 99 (1980).

[11]M. H. Day, Phys. Rev. B 12, 514 (1975); R. H.

Ritchie, W. Brandt, and P. M. Echenique, *ibid.* 14, 4808 (1976); Z. Vager and D. S. Gemmell, Phys. Rev. Lett. 37, 1352 (1976); M. Kitagawa and Y. H. Ohtsuki, Phys. Rev. B 16, 5321 (1977); R. H. Ritchie, P. M. Echenique, W. Brandt, and G. Basbas, IEEE Trans. Nucl. Sci. 26, 1001 (1979); M. H. Day and M. Ebel, Phys. Rev. B 19, 3434 (1979); P. M. Echenique, R. H. Ritchie, and W. Brandt, *ibid.* 20, 2567 (1979); A. Faibis, R. Kaim, I. Plesser, and Z. Vager, Nucl. Instrum. Methods 170, 99 (1980); A. Mazarro, P. M. Echenique, and R. H. Ritchie (unpublished).

[12]A. Lurio, H. H. Andersen, and L. C. Feldman, Phys. Rev. A 17, 90 (1978).

[13]Work by Arista [Phys. Rev. B 18, 1 (1978)] on the vicinage effect in stopping of diclusters in an electron gas is concerned with some of the points which we discuss here. Similarly, Steinbeck and Dettman [J. Phys. C 11, 2907 (1978)] describe theoretically the interaction of diclusters with single atoms.

[14]H. A. Bethe, Ann. Phys. (N.Y.) 5, 325 (1930).

[15]M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions,* NBS Applied Math Series No. 55 (U. S. GPO, Washington, D. C., 1964).

[16]N. R. Arista and V. H. Ponce [J. Phys. 8, L188 (1975)] computed a vicinage function for energy loss in an electron gas, using a different approach, and showed that the spherical average may be carried out analytically.

[17]J. Lindhard, K. Dan. Vidensk. Selsk. Mat.-Fys. Medd. 28, No. 8 (1954).

[18]See J. Callaway, Phys. Rev. 116, 1368 (1950) and E. Tosatti and G. P. Parravicini, J. Phys. Chem. Solids 32, 623 (1971). This model has been used by Emerson *et al.* [Phys. Rev. B 7, 1798 (1973)] in a study of electron slowing-down spectra in silicon.

[19]W. Brandt and J. Reinheimer, Phys. Rev. B 2, 3104 (1970); 4, 1395(E) (1971).

[20]D. Penn, Phys. Rev. 128, 2093 (1962).

[21](a) N. Bohr, Philos. Mag. 25, 10 (1913); (b) 30, 581 (1915); K. Dan. Vidensk. Selsk. Mat.-Fys. Medd. 18, No. 8 (1948).

[22]U. Fano, Ann. Rev. Nucl. Sci. 13, 1 (1963).

[23]E. Merzbacher and H. W. Lewis, in *Handbuch der Physik,* edited by S. Flugge (Springer, Berlin, 1958), Vol. 34, p. 166.

[24]S. T. Manson, Phys. Rev. A 6, 1013 (1972). The authors are very grateful to Dr. Manson for providing us with numerical tables of the GOS for these shells.

[25]D. H. Madison and E. Merzbacher, in *Atomic Inner Shell Processes,* edited by B. Crasemann (Academic, New York, 1975), Vol. I, p. 55, Eq. (73).

[26]S. Cohen, J. R. Hiskes, and R. J. Riddell, Jr., Phys. Rev. 119, 1025 (1960); Report No. UCRL-8871, 1959 (unpublished).

[27]W. L. Walters, D. G. Costello, J. G. Skofronick, D. W. Palmer, W. E. Kane, and R. G. Herb, Phys. Rev. 125, 2012 (1962).

[28]E. P. Kanter, P. J. Cooney, D. S. Gemmell, K.-O. Groeneveld, W. J. Pietsch, A. J. Ratkowski, Z. Vager, and B. J. Zabransky, Phys. Rev. A 20, 834 (1979).