

Dielectric theory of the electronic stopping power of fast ions in crystals

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The electronic stopping of a fast, charged particle in a lattice-periodic structure is treated by linear response theory. The energy loss in open crystal channels is found to be reduced due to destructive interference of umklapp scattering processes with small momentum transfer. A sum rule for the first reciprocal moment of the velocity-dependent stopping power is derived, which can be expressed in terms of the static inverse dielectric function. Quantitative information about the importance of the crystal potential is obtained by first-principles pseudopotential calculations. The averaged stopping power in the $\langle 110 \rangle$ channel in Si is calculated to be smaller by 30% compared to nonchanneling directions. The sum-rule calculations in the local-density approximation agree with the band-structure results within 20% and are used to discuss the electronic stopping in other semiconductors.

The stopping of fast, charged particles by an electronic system has been the topic of numerous experimental and theoretical studies, which are motivated by important applications such as ion implantation and backscattering experiments. The microscopic theory of the stopping power can be formulated in terms of the linear response of the system to the added charge.¹ It is well known that approximating the target by a homogeneous electron gas is not adequate to describe experiments. In Monte Carlo simulations of the ion implantation process, the inhomogeneity of the target crystal has been modeled by phenomenological impact-parameter-dependent expressions for the electronic stopping.² Among the first authors to describe the stopping in periodically inhomogeneous structures in linear response theory were Saslow and Reiter.³ Bonsignori and Desalvo⁴ derived the stopping power under channeling conditions. In subsequent works by the same authors⁵ and later by Desalvo and Rosa⁶ and by Burenkov *et al.*,⁷ simple models for the dielectric response function were used to calculate the stopping of ions with specified kinetic energies. Realistic band-structure calculations of the energy dissipation of fast particles in crystals have not been carried out so far.

In this paper we present a general linear response treatment of the inhomogeneous viscosity of the electron liquid in lattice-periodic structures. An expression for the position-dependent stopping power is derived and discussed. Special attention is paid to the role of umklapp processes under channeling conditions. By using a new sum rule, we focus on a velocity average of the stopping of a point charge in Si, which is calculated from first principles. Several applications of this sum rule are discussed.

We consider a classical point charge Z (in atomic units used throughout $Z = 1$ for a proton), moving at a constant velocity \mathbf{v} along $\mathbf{R} = \mathbf{R}_0 + \mathbf{v}t$, where \mathbf{R}_0 is the impact parameter. In the energy range, for a proton, of 100 keV–4 MeV, the kinetic energy is dissipated predominantly via valence electron excitations. The stopping

power S , defined as the energy loss per unit path length, is given by¹

$$S(\mathbf{R}, \mathbf{v}) = -(1/v)Z\mathbf{v} \cdot \nabla \phi^{\text{ind}}(\mathbf{r} = \mathbf{R}; t). \quad (1)$$

Here, ϕ^{ind} is the induced screening potential of the stopping medium. Linear response theory supposes that ϕ^{ind} depends linearly on the Coulomb potential of the perturbation through the inverse dielectric response function ϵ^{-1} . At low ion velocities nonlinear corrections are appreciable in metals, but presumably less so in semiconductors, which are the main subject of the present study. In periodic structures, the inverse dielectric matrix in momentum space (IDM) can be written in general as $\epsilon^{-1} = \epsilon^{-1}(\mathbf{q}, \mathbf{q} + \mathbf{G}; \omega)$, where \mathbf{G} is a reciprocal lattice vector. The position-dependent stopping power inside a periodic structure is given by^{8,9}

$$S(\mathbf{R}, \mathbf{v}) = \frac{1}{v} \int \frac{d\mathbf{q}}{(2\pi)^3} (\mathbf{q} \cdot \mathbf{v}) \sum_{\mathbf{G}} \frac{4\pi Z^2}{|\mathbf{q} + \mathbf{G}|^2} \times \text{Im}\{-\epsilon^{-1}[\mathbf{q}, \mathbf{q} + \mathbf{G}; (\mathbf{q} + \mathbf{G}) \cdot \mathbf{v}] e^{-i\mathbf{G} \cdot \mathbf{R}}\}. \quad (2)$$

This expression can be interpreted as an integral over electronic scattering processes with given energy and momentum transfer from the ion to the electronic system. The electron to which a momentum \mathbf{q} is transferred can instantaneously be Bragg-reflected to receive additional momentum \mathbf{G} from the lattice. The sum over reciprocal lattice vectors \mathbf{G} in Eq. (2) provides the rate at which the ion loses momentum, regardless of the final momentum state of the electronic system. The corresponding energy loss is well approximated¹ by $\mathbf{q} \cdot \mathbf{v}$. Hence, Eq. (2) is the expectation value for the total energy loss. For the special case of a homogeneous system ($\mathbf{G} = \mathbf{0}$), Eq. (2) reduces to the standard result.¹

Note that the umklapp scattering introduces a phase factor. As a consequence, the instantaneous energy loss is partially determined by the real (elastic) part of the

dielectric response, which is counterintuitive in an energy loss expression. It does not contribute to the permanent energy loss, however: The local stopping power in Eq. (2) can be separated into two components according to their behavior in time as the charge moves through the crystal. The periodic energy loss component is determined by those \mathbf{G} with $\mathbf{G} \cdot \mathbf{v} \neq 0$. These contributions are characterized by an oscillating phase factor $\exp[-i\mathbf{G} \cdot \mathbf{v}t]$ and hence vanish on average. The other component, the permanent or average energy loss, is determined by \mathbf{G}_\perp , i.e., \mathbf{G} satisfying $\mathbf{G} \cdot \mathbf{v} = 0$ (Ref. 5). The periodic energy loss component causes a periodic acceleration and deceleration of the particle, causing emission of coherent electromagnetic radiation. The radiated intensity is largest under channeling conditions, contributing to the coherent bremsstrahlung radiation, which is traditionally calculated by considering just the atomic Hartree potentials in the crystal.¹⁰ The radiation discussed here is therefore a dielectric contribution to the coherent bremsstrahlung which to the best of our knowledge has not been discussed before. We notice that its importance in different dynamic regimes needs to be established, but disregard radiative losses in the following.

The periodic components vanish on averaging the stopping power $\langle S(\mathbf{R}_0, \mathbf{v}) \rangle$ along the given trajectory⁸

$$\langle S(\mathbf{R}_0, \mathbf{v}) \rangle = \frac{1}{v} \int \frac{d\mathbf{q}}{(2\pi)^3} (\mathbf{q} \cdot \mathbf{v}) \sum_{\mathbf{G}=\mathbf{G}_\perp} \frac{4\pi Z^2}{|\mathbf{q}+\mathbf{G}|^2} \times \text{Im}[-\epsilon^{-1}(\mathbf{q}, \mathbf{q}+\mathbf{G}; \mathbf{q} \cdot \mathbf{v})] \cos(\mathbf{G} \cdot \mathbf{R}_0). \quad (3)$$

The most important contribution in Eq. (3) is provided by the term $\mathbf{G} = \mathbf{0}$. The magnitude of the other terms depends on the direction of \mathbf{v} . For most directions, the condition $\mathbf{G} \cdot \mathbf{v} = 0$ is never satisfied (irrational angles), or satisfied only by a few large \mathbf{G}_\perp , with negligible contributions. $\langle S(\mathbf{R}_0, \mathbf{v}) \rangle$ is then equal to or very well approximated by the leading term $\mathbf{G}_\perp = \mathbf{0}$. In these general directions¹¹ we obtain the “random” stopping power $\langle S(\mathbf{v}) \rangle_R$,

$$\langle S(\mathbf{v}) \rangle_R = \frac{1}{v} \int \frac{d\mathbf{q}}{(2\pi)^3} (\mathbf{q} \cdot \mathbf{v}) \frac{4\pi Z^2}{q^2} \text{Im}[-\epsilon^{-1}(\mathbf{q}, \mathbf{q}; \mathbf{q} \cdot \mathbf{v})], \quad (4)$$

which is independent of \mathbf{R}_0 . From the nearly isotropic diagonal elements of the IDM (Ref. 12) it follows that the remaining dependence of the random stopping on the direction of \mathbf{v} is very weak. For a few highly symmetric directions, some $\mathbf{G}_\perp \neq \mathbf{0}$ exist which are small enough to contribute significantly in Eq. (3). For these *channeling* directions, one has to carry out the sum over reciprocal lattice vectors. The extra terms are negative if \mathbf{R}_0 is chosen near the channel axis. The reduced electronic stopping power for channeled ions can be explained by destructive interference of symmetry-allowed, low-index umklapp processes. The alternative, real-space explanation in terms of the reduced electron density in the channels (see below) is more intuitive, but less accurate.

Direct evaluation of Eq. (3) is complicated due to the highly singular nature of the dynamic dielectric function and its inverse.¹³ Instead, consider the first reciprocal moment

$$\langle A(\mathbf{R}_0, \hat{\mathbf{v}}) \rangle = \int_0^\infty dv \frac{1}{v} \langle S(\mathbf{R}_0, \mathbf{v}) \rangle. \quad (5)$$

$\langle A(\mathbf{R}_0, \hat{\mathbf{v}}) \rangle$ is a velocity average of the stopping power with emphasis on the low velocity regime. In the homogeneous electron gas the stopping power increases linearly with v at low energies, i.e., the integrand in Eq. (5) becomes constant and proportional to the slope. By making use of one of the Kramers-Kronig relations,¹⁴ Eq. (5) can be rewritten in terms of the real part of ϵ^{-1} :⁸

$$\langle A(\mathbf{R}_0, \hat{\mathbf{v}}) \rangle = \frac{Z^2}{4\pi} \int d\mathbf{q} \sum_{\mathbf{G}=\mathbf{G}_\perp} \frac{|\mathbf{q} \cdot \hat{\mathbf{v}}|}{|\mathbf{q}+\mathbf{G}|^2} \times \{\delta_{G0} - \text{Re}[\epsilon^{-1}(\mathbf{q}, \mathbf{q}+\mathbf{G}; 0)]\} \times \cos(\mathbf{G} \cdot \mathbf{R}_0). \quad (6)$$

The main advantage of working with this velocity average rather than the stopping power itself is the considerable saving of time and effort to produce first-principles numerical results. The sum rule is useful to compare channel stopping and random stopping, to investigate the impact parameter dependence, to test approximations and as a measure for the effect of, e.g., an electronic band gap.

First-principles pseudopotential theory¹⁵ was used to calculate the static IDM for Si (Ref. 16). Exchange-correlation corrections¹² were disregarded (random-phase approximation). Equation (6) was evaluated numerically for “random” particles as well as for particles in the center of the $\langle 100 \rangle$ and $\langle 110 \rangle$ channels. We obtain for the ratio of the sum rule for the “best channeled” to the

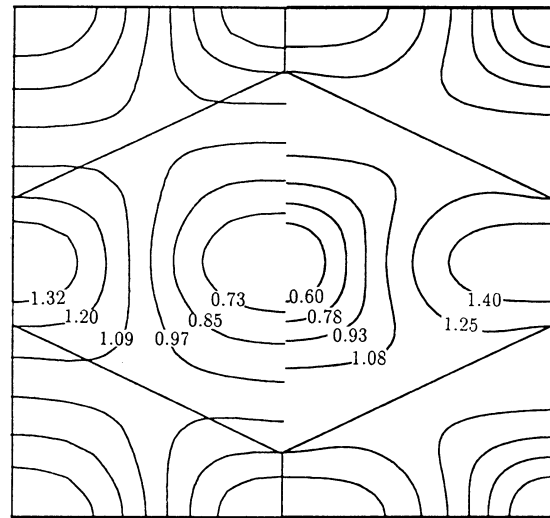


FIG. 1. Contour plot of $\langle A \rangle_{110} / \langle A \rangle_R$ vs impact parameter \mathbf{R}_0 . On the left are results of the first-principles band-structure calculations. The results on the right side have been obtained in the LDA, i.e., are based on the Hybertsen-Louie dielectric function. The projection of the lines connecting nearest neighbor atom cores are also indicated.

random ions $\langle A \rangle_{100}/\langle A \rangle_R = 0.98$, $\langle A \rangle_{110}/\langle A \rangle_R = 0.69$, where $\langle A \rangle_R/Z^2 = 0.39$ a.u. Although the $\langle 100 \rangle$ channel is geometrically well defined, the channel and random stopping powers are almost equal. This result is consistent with the fact that the average electron density in this channel does not differ much from the volume average. A much clearer contrast is provided by the $\langle 110 \rangle$ channel. Ratios of 0.88 for 100 keV (Ref. 6) and 0.43 (Refs. 17 and 18) for 4 MeV have been found experimentally, where the best channeled particles were identified by the leading edge of the channeled energy-loss spectra. The present result of 0.69 for the velocity average appears to be consistent with the experimental data.

$$\epsilon_{\text{HL}}^{-1}(\mathbf{q}, \mathbf{q} + \mathbf{G}, 0) = \frac{1}{2} \left\{ \frac{1}{V_c} \int_{V_c} d\mathbf{r} \epsilon_{\text{LL}}^{-1}(|\mathbf{q} + \mathbf{G}|, \rho(\mathbf{r})) e^{i\mathbf{G} \cdot \mathbf{r}} + \frac{|\mathbf{q} + \mathbf{G}|^2}{|\mathbf{q}|^2} \frac{1}{V_c} \int_{V_c} d\mathbf{r} \epsilon_{\text{LL}}^{-1}(|\mathbf{q}|, \rho(\mathbf{r})) e^{i\mathbf{G} \cdot \mathbf{r}} \right\}, \quad (7)$$

where V_c is the unit cell volume. The local screening is described by the Levine-Louie reciprocal dielectric function $\epsilon_{\text{LL}}^{-1}$ for semiconductors,²¹ which is a function of two parameters, the macroscopic dielectric constant ϵ_0 and the (local) valence electron density $\rho(\mathbf{r})$. To test the accuracy of this model for the present purposes, we calculate the sum rule for the stopping power from Eqs. (6) and (7) and compare it with the result from the band-structure calculations. The inhomogeneous charge density is constructed by a Fourier synthesis of valence structure factors. For consistency we used the ϵ_0 and structure factors from the band-structure calculations.²² Omitting $\mathbf{G}_\perp \neq 0$, the sum rule in random directions becomes

$$\begin{aligned} \langle A \rangle_R^{\text{LDA}} &= \frac{Z^2}{2} \int_0^\infty dq q \left[1 - \frac{1}{V_c} \int_{V_c} d\mathbf{r} \epsilon_{\text{LL}}^{-1}(q, \rho(\mathbf{r})) \right] \\ &= \frac{1}{V_c} \int_{V_c} d\mathbf{r} \langle A \rangle_{\text{LL}}(\rho(\mathbf{r})), \end{aligned} \quad (8)$$

where we have defined a local stopping $\langle A \rangle_{\text{LL}}(\rho)$ determined by the Levine-Louie dielectric function with local electron density $\rho(\mathbf{r})$. For the $\langle 100 \rangle$ channeling direction, i.e., $\mathbf{R} = (X, Y_0, Z_0)$ and $\mathbf{R}_0 = (0, Y_0, Z_0)$, one finds after some manipulations⁸

$$\begin{aligned} \langle A \rangle_{100}^{\text{LDA}} &= \frac{Z^2}{2} \int_0^\infty dq q \left[1 - \frac{1}{a} \int_a dx \epsilon_{\text{LL}}^{-1}(q, \rho(x, Y_0, Z_0)) \right] \\ &= \frac{1}{a} \int_a dx \langle A \rangle_{\text{LL}}(\rho(x, Y_0, Z_0)), \end{aligned} \quad (9)$$

where a is the lattice parameter. Analogous results are obtained for other directions. For the specific velocity average of the energy loss considered here the Hybertsen-Louie IDM gives results which are identical to that of the local-density approximation²³ (LDA), often used in simulations of the ion implantation process²⁴ and also for description of the stopping power of channeled ions.²⁵ This result is a strong indication that the velocity-dependent stopping will not be very different in both models.

Numerical evaluation of Eqs. (8) and (9) yields $\langle A \rangle_{100}^{\text{LDA}}/\langle A \rangle_R^{\text{LDA}} = 1.08$, $\langle A \rangle_{110}^{\text{LDA}}/\langle A \rangle_R^{\text{LDA}} = 0.52$, and $\langle A \rangle_R^{\text{LDA}} = 0.41$ a.u. The impact parameter dependence

$\langle A \rangle_{110}/\langle A \rangle_R$ as a function of impact parameter \mathbf{R}_0 is depicted on the left-hand side of Fig. 1. The figure clearly shows the stopping power minimum in the center of the channel and the gradual increase as one moves away from the channel axis.

Self-consistent band-structure calculations of Eq. (5) are still very time consuming. The Hybertsen-Louie dielectric function^{19,20} has been introduced to simplify calculations of the dynamical self-energy in semiconductors and found to be sufficiently accurate for this application. The Hybertsen-Louie IDM (Ref. 19) is a nonlocal model containing a unit cell integration of the local screening, according to

of the latter is shown on the right-hand side of Fig. 1. Comparison with results obtained from Eq. (6) exposes the limited accuracy of the LDA (or HL-IDM) for the calculation of the anisotropic stopping power. The overestimated anisotropy of the viscosity is attributed to the neglect of distant collisions in the LDA,⁵ which can be treated only by band-structure calculations. Yet, the disagreement is not very large (20%) [which is much smaller than the errors in the individual matrix elements in Eq. (7)], and the general features are qualitatively correct. The LDA is found to be a reasonable approximation for semiconductors, certainly for random energy loss.

Having established the (in)accuracy of the local-density approximation we now pursue a parametric study within the LDA to compare the average energy loss, i.e., the $\langle A \rangle$'s, of different tetrahedral semiconductors. The calculations were repeated using the experimental ϵ_0 's and electron density structure factors. The results are summarized in the tables. Table I illustrates the effect of the energy gap on $\langle A \rangle_R^{\text{LDA}}$ [Eq. (8)] by comparing the results of the Levine-Louie dielectric function with that from the Lindhard dielectric function for the homogeneous electron gas, obtained by letting $\epsilon_0 \rightarrow \infty$. An increasing gap reduces the stopping power, as expected.²⁶ According to Table II the effect of channeling is largest for diamond but very important for all three semiconductors. But, as discussed above, these numbers somewhat overestimate the real effect.

In conclusion, we have investigated the electronic stopping power of fast ions in tetrahedral semiconductors

TABLE I. Calculated $\langle A \rangle_R^{\text{LDA}}/Z^2$, using the Levine-Louie dielectric function, which models the effect of a gap, and the Lindhard dielectric function of the free electron gas. The last column shows the difference in percent. Units are Hartree/Bohr radius (a.u.).

	ϵ_0	$\langle A \rangle_R^{\text{LDA}}/Z^2$ Levine-Louie	$\langle A \rangle_R^{\text{LDA}}/Z^2$ Lindhard	Difference
C	5.5	0.64	0.80	+24%
Si	11.7	0.39	0.45	+15%
Ge	15.8	0.38	0.43	+12%

TABLE II. $\langle A \rangle_{\text{ch}}^{\text{LDA}}/Z^2$ calculated for the $\langle 100 \rangle$ and $\langle 110 \rangle$ channels, along with the differences compared to the $\langle A \rangle_{\text{R}}^{\text{LDA}}/Z^2$ in Table I. The average electron densities $\rho_{\text{ch,av}}$ in the channel are also shown.

Channel		$\langle A \rangle_{\text{ch}}^{\text{LDA}}/Z^2$	Difference	$\rho_{\text{ch,av}}$
$\langle 100 \rangle$	C	0.58	-10%	0.072
	Si	0.36	-10%	0.020
	Ge	0.34	-12%	0.018
$\langle 110 \rangle$	C	0.29	-56%	0.017
	Si	0.21	-47%	0.007
	Ge	0.21	-46%	0.007

with emphasis on Si. By focusing on the first reciprocal moment of the velocity-dependent stopping it is possible to quantitatively assess the importance of band-structure effects on the electronic stopping power. Significant deviations (20%) between the result obtained from first-principles band-structure calculations and a simple local-density approximation are found. The LDA is used to get an impression of the effect of the electronic band gap and umklapp scatterings in other IV-IV semiconductors. The inaccuracies of the LDA are presumably tol-

erable in Monte Carlo simulations of ion propagation in crystals. It requires only a reliable electron density distribution and a macroscopic dielectric constant as input and is simple enough for implementation into simulation routines. The accuracy as established here and the absence of adjustable parameters make this approximation attractive for practical applications, such as ion implantation in crystalline solids.

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