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Electronic stopping power of periodic crystals

I. Campillo [a], J.M. Pitarke [a,*], A.G. Eguiluz [b,c], Alberto García [d]

[a] *Materia Kondentsatuaren Fisika Saila, Zientzi Fakultatea, Euskal Herriko Unibertsitatea, 644 Posta kutxatila, 48080 Bilbo, Basque Country, Spain*

[b] *Department of Physics and Astronomy, The University of Tennessee, Knoxville, TN 37996-1200, USA*

[c] *Solid State Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831-6032, USA* [1]

[d] *Departamento de Física Aplicada II, Facultad de Ciencias, Universidad del País Vasco, Apartado 644, 48080 Bilbao, Spain*

Abstract

The electronic stopping power suffered by ions penetrating periodic crystals is investigated within linear response theory. A full treatment of the dynamical response of valence electrons is made, starting from a realistic description of the one-electron band structure and using ab initio non-local pseudopotentials. The dynamical density-response function is obtained in the random-phase approximation. Results for the stopping power of aluminum averaged over impact parameters are presented. © 1998 Elsevier Science B.V.

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1. Introduction

The stopping power of charged particles penetrating a solid has been the subject of considerable theoretical and experimental research, since the beginning of this century [1–4]. The electronic stopping power of valence electrons has been calculated for many years on the basis of a jellium model of the electron gas [5], in which electronic states are described by plane waves. In a more realistic approach to the stopping power of real solids, valence electrons move in a periodic potential, electronic states are described by Bloch states, and the spectrum of one-electron excitations splits into so-called energy bands.

Recently, Quong and Eguiluz [6] carried out an ab initio evaluation, within the random-phase approximation (RPA) [7], of the plasmon dispersion relation in Al, and found the crystal lattice to lower the plasmon frequency for large wave vectors by as much as 4 eV, in agreement with the experimental data [8]. Subsequently, Fleszar et al. [9] evaluated the dynamical structure factor of Al crystal for large wave vectors and concluded that an experimentally observed [10] two-peak loss structure is a band-structure effect. Both calculations and similar recent work by other authors [11,12], have established the importance of band-structure effects in the response of valence electrons, even in the case of simple free-electron-like metals such as Al.

* Corresponding author. E-mail: wmppitoj@lg.ehu.es.

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On the basis of approximate treatments of the dielectric matrix of periodic crystals [13,14], the stopping power of valence electrons in real solids has been described [15–19] over the years within linear response theory. More recently, the low-velocity limit has been investigated on the basis of a static treatment of the density-response [20] and ab initio band-structure based dynamical calculations of the electronic stopping power have been presented [21].

In this paper we first outline, in Section 2, our method for the evaluation of the stopping power of valence electrons in periodic crystals. In Section 3, we present RPA results for the random stopping power of Al crystal.

2. Theory

We consider a particle of charge Z_1 (atomic units are used throughout, i.e., $\hbar = e = m_e = 1$) moving in a periodic crystal with velocity $\mathbf{v}$ at a given impact parameter $\mathbf{b}$.

The position-dependent stopping power is obtained as the energy loss per unit path length of the projectile, and in the case of periodic crystals with inversion symmetry it is found to be given by the following expression [18,21]:

$$\left[-\frac{dE}{dx}\right]_{\mathbf{b}} = -\frac{8\pi}{\Omega v}Z_1^2 \times \sum_{\mathbf{q}}^{BZ}\sum_{\mathbf{G}}\sum_{\mathbf{K}}^{*} e^{i\mathbf{K}\cdot\mathbf{b}}\,\mathrm{Im}\,\epsilon^{-1}_{\mathbf{G},\mathbf{G}+\mathbf{K}}[\mathbf{q},(\mathbf{q}+\mathbf{G})\cdot\mathbf{v}] \times \frac{(\mathbf{q}+\mathbf{G})\cdot\mathbf{v}}{|\mathbf{q}+\mathbf{G}+\mathbf{K}|^2}, \quad (1)$$

where Ω represents the normalization volume, the first sum runs over $\mathbf{q}$ vectors within the first Brillouin zone, and $\mathbf{G}$ are vectors of the reciprocal lattice. The star in the third summation indicates that only those reciprocal vectors which are perpendicular to the velocity of the projectile, i.e., $\mathbf{K}\cdot\mathbf{v} = 0$, are included. Finally, $\epsilon^{-1}_{\mathbf{G},\mathbf{G}'}(\mathbf{q},\omega)$ is the inverse dielectric matrix in momentum space,

$$\epsilon^{-1}_{\mathbf{G},\mathbf{G}'}(\mathbf{q},\omega) = \delta_{\mathbf{G},\mathbf{G}'} + \frac{4\pi}{|\mathbf{q}+\mathbf{G}|^2}\chi_{\mathbf{G},\mathbf{G}'}(\mathbf{q},\omega), \quad (2)$$

$\chi_{\mathbf{G},\mathbf{G}'}(\mathbf{q},\omega)$ representing the Fourier coefficients of the dynamical density-response function.

The average of Eq. (1) over impact parameters gives the following expression for the random stopping power:

$$\left[-\frac{dE}{dx}\right]_{\text{random}} = -\frac{8\pi}{\Omega v}Z_1^2\sum_{\mathbf{q}}^{BZ}\sum_{\mathbf{G}}\frac{(\mathbf{q}+\mathbf{G})\cdot\mathbf{v}}{|\mathbf{q}+\mathbf{G}|^2}\,\mathrm{Im}\,\epsilon^{-1}_{\mathbf{G},\mathbf{G}}[\mathbf{q},(\mathbf{q}+\mathbf{G})\cdot\mathbf{v}], \quad (3)$$

which is identical to the $\mathbf{K} = 0$ part of the position-dependent stopping power.

If S is a symmetry operation of the point group of the periodic crystal, one can take advantage of the symmetry properties of the dielectric matrix under rotation and write

$$\epsilon_{\mathbf{G},\mathbf{G}'}(S\mathbf{q},\omega) = \epsilon_{S^{-1}\mathbf{G},S^{-1}\mathbf{G}'}(\mathbf{q},\omega). \quad (4)$$

Thus the random stopping power can be evaluated from the knowledge of the dielectric matrix for wave vectors $\mathbf{q}$ lying in the irreducible element of the Brillouin zone, according to

$$\left[-\frac{dE}{dx}\right]_{\text{random}} = -\frac{8\pi}{\Omega v}Z_1^2\sum_{\mathbf{q}}^{IBZ}\sum_{S}\sum_{\mathbf{G}}\frac{(S\mathbf{q}+\mathbf{G})\cdot\mathbf{v}}{|S\mathbf{q}+\mathbf{G}|^2} \times \mathrm{Im}\,\epsilon^{-1}_{S^{-1}\mathbf{G},S^{-1}\mathbf{G}}[\mathbf{q},(S\mathbf{q}+\mathbf{G})\cdot\mathbf{v}], \quad (5)$$

where the first sum runs over $\mathbf{q}$ vectors within the irreducible element, and the second sum runs over the symmetry operations which generate the wave vectors in the star of each $\mathbf{q}$.

In the RPA, the coefficients $\chi_{\mathbf{G},\mathbf{G}'}(\mathbf{q},\omega)$ of Eq. (2) are obtained by solving a linear matrix equation which relates them to the Fourier coefficients $\chi^0_{\mathbf{G},\mathbf{G}'}(\mathbf{q},\omega)$ of the density-response function for non-interacting electrons [22],

$$\chi^0_{\mathbf{G},\mathbf{G}'}(\mathbf{q},\omega) = \frac{1}{\Omega}\sum_{\mathbf{k}}^{BZ}\sum_{n,n'}\frac{f_{\mathbf{k},n} - f_{\mathbf{k}+\mathbf{q},n'}}{E_{\mathbf{k},n} - E_{\mathbf{k}+\mathbf{q},n'} + \hbar(\omega + i\eta)} \times \langle\phi_{\mathbf{k},n}|e^{-i(\mathbf{q}+\mathbf{G})\cdot\mathbf{r}}|\phi_{\mathbf{k}+\mathbf{q},n'}\rangle\langle\phi_{\mathbf{k}+\mathbf{q},n'}|e^{i(\mathbf{q}+\mathbf{G}')\cdot\mathbf{r}}|\phi_{\mathbf{k},n}\rangle, \quad (6)$$

where the second sum runs over the band structure for each wave vector $\mathbf{k}$ in the first Brillouin zone, $f_{\mathbf{k},n}$ are Fermi factors, and $\phi_{\mathbf{k},n}$ represent one-electron Bloch states with energies $E_{\mathbf{k},n}$.

The input to our entire calculation of the random stopping power given by Eq. (5) is a self-consistent solution of the ground state of the periodic crystal. The one-electron Bloch states $\phi_{\mathbf{k},n}$ are expanded in a plane wave basis with a kinetic-energy cutoff of 12 Ry. Then we solve for the coefficients of this expansion self-consistently with an effective one-electron potential consisting only of the average electrostatic interaction between the electrons and the electronic–ionic background electrostatic interaction (Hartree approximation). The electron–ion interaction is described by a non-local, norm-conserving pseudopotential generated according to the Hamann et al. scheme [23]. The sum over the BZ in both the polarizability of Eq. (6) and the stopping power of Eq. (5) has been performed on $10 \times 10 \times 10$ Monkhorst–Pack meshes [24], which we have found to be enough for convergence in the case of projectiles moving with velocities up to $v = 2$ a.u. In this velocity regime, i.e. $0 < v < 2$ a.u., on the order of 30 bands are required for convergence.

3. Results and discussion

Fig. 1 exhibits the velocity dependence of the random RPA stopping power of valence electrons in Al crystal, together with the corresponding result for a homogeneous electron gas with the same

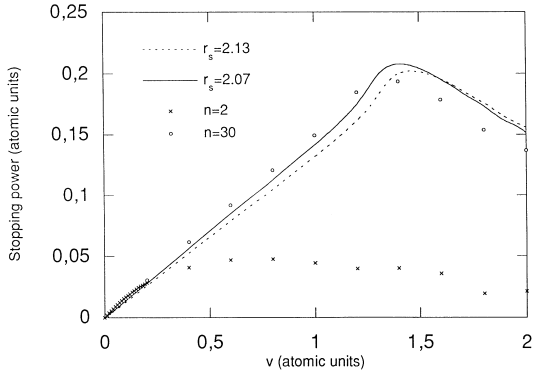

Fig. 1. RPA electronic stopping powers, as a function of the velocity of the projectile. n-band calculations for Al crystal are represented by crosses ($n = 2$) and circles ($n = 30$). Calculated stopping powers for jellium with $r_s = 2.07$ and $r_s = 2.14$ are represented by solid and dotted lines, respectively.

average valence electron density, i.e., $r_s = 2.07$. For comparison, the RPA stopping power of jellium electrons with $r_s = 2.14$ (the effective electron density parameter for which the plasmon energy for $\mathbf{q} = 0$ is the same as in Al crystal, $\omega_p = 15$ eV) is represented in the same figure by a dotted line. Stopping powers of both crystal and jellium Al have been evaluated from Eq. (5), on the basis of crystal and jellium calculations of the polarizability of Eq. (6), with the sum over $\mathbf{G}$ extended to 15 vectors of the reciprocal lattice, the magnitude of the maximum momentum transfer $\mathbf{q} + \mathbf{G}$ being $2.8q_F$ (q_F is the Fermi momentum). The sampling of the BZ required in Eq. (6) was done in conjunction with a choice of a finite value of η of 0.65 eV.

Our results show that the stopping power of Al crystal is, for projectile velocities smaller than the Fermi velocity, a linear function of the velocity and about 3% higher than the stopping power of jellium electrons. This slight enhancement of the stopping power is a consequence of the fact that the imaginary part of the polarizability $\chi^0_{\mathbf{G},\mathbf{G}}(\mathbf{q},\omega)$ is slighlty enhanced at low frequencies (where it increases linearly with frequency) when the band structure of the valence electrons is taken into account [9].

We remark that at velocities below $v = 0.1$ a.u. just two bands are required to account for the energy loss; this is a plausible result. At larger velocities excitations to higher bands become possible, and we find that convergence is achieved, for $v < 2$ a.u., if one considers on the order of 30 bands.

The threshold velocity for which plasmon excitation becomes possible is 1.27 and 1.23 a.u. in the case of electrons in jellium with $r_s = 2.07$ and $r_s = 2.14$, respectively, resulting in a shift in the jellium curves of Fig. 1 for these velocities. In the case of valence electrons in Al crystal a similar, though less intense, peak appears at a lower velocity. This is a consequence of the threshold velocity of heavy projectiles in Al crystal being smaller than predicted within a jellium model of the target with $r_s = 2.07$ and also with $r_s = 2.14$, as can be concluded from an inspection of the plasmon dispersion curve for Al crystal of Ref. [6], which we have reproduced.

At high frequencies the imaginary part of the density-response function $\chi_{\mathbf{G},\mathbf{G}}(\mathbf{q},\omega)$ for band elec-

trons is, within the electron–hole pairs continuum, smaller than in the case of jellium electrons [9]. Preliminary results show that band structure corrections lower the stopping power of electrons in jellium by about 10% at and just above the threshold velocity.

In summary, we have presented new results of ab initio calculations of random stopping power of Al crystal, as a function of the velocity of the projectile, for velocities up to $v = 2$ a.u. Our results indicate that band structure corrections for this free-electron-like material are small but non-negligible. While at low velocities the stopping power is slightly enhanced when the full band structure is taken into account, for velocities on the order of the plasmon threshold velocity smaller values for the stopping power are expected, band structure corrections being on the order of 10%. A more detailed presentation of our ab initio calculations of both random and position-dependent electronic stopping powers of Al for a wide range of non-relativistic projectile velocities will be published elsewhere [25].

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