DYNAMIC LOCAL FIELD CORRECTION IN THE CALCULATION OF ELECTRONIC STOPPING POWER*

I. NAGY

Quantum Theory Group, Institute of Physics,
Technical University, Budapest, H-1521

Received June 27, 1989

Abstract

Energy loss of slow charged particles in a degenerate electron gas is calculated on the basis of the dielectric theory. The influence of dynamic local field correction is examined. A straightforward extension of this theory to problems that exceed the validity of the first Born approximation is discussed.

Introduction

Accurate values of electronic energy loss of ions in solids are extremely valuable in various branches of science and technology. The agreement between theoretical and experimental results is good in the high velocity region, where it can be shown that all of the theoretical results converge to the same limit (Bethe-Bloch region). At lower velocities the agreement becomes poorer. The discrepancies in this velocity region are mainly due to the lack of information about the exact electron densities in the interatomic regions in solids *and* the stopping function.

The electron density of solids can in principle be derived quite accurately from the free atom wavefunctions using the linear combination of atomic orbitals. This formalism enables us to find the change in the wavefunction of an electron bound to a certain atom in the lattice due to overlaps with wavefunctions of neighbouring atomic electrons. In solids, and especially in metals, even overlaps of high-order neighbours are not negligible for the outer weakly bound electrons. As a result the outer electrons are delocalized and an almost constant electron density is obtained in most of the atomic volume, while the core of inner-shell electrons is only slightly affected by this interaction.

The problem of slow ions moving at constant velocity v ($v < v_F$, v_F being the Fermi velocity of electrons in the conduction band) is of special interest in cases such as the slowing and reflection of hydrogen atoms impinging on the inner wall of a controlled thermonuclear reactor as well as in astrophysical studies and in surface analytical techniques.

---

* Dedicated to Prof. J. Giber on the occasion of his 60th birthday.

Usually a many-body formalism (Fetter and Walecka, 1971; Mahan, 1981) is needed to treat the complicated nature of the interaction. An essential step is to introduce an appropriate *local* potential to describe the interaction of the electrons with the incoming ion.

The linear response theory provides a good starting point to characterize the stopping phenomenon.

The method

The main concern of this short paper consists in describing electronic excitation that means the energy (ω) and momentum (k) is transferred to the electrons of a degenerate, homogeneous system.

The absorption probability per unit volume and unit time, is given by (atomic units are used throughout this paper, except where explicitly stated):

$$P(k,\omega)=n\,|U(k)|^2 S(k,\omega)\delta(\omega-\mathbf{kv}). \tag{1}$$

Here n is the density of the electron gas, $U(k)$ is the interaction potential and $S(k, \omega)$ is the dynamic structure function of the system (Van Hove, 1954).

There is a well-known relation between $S(k, \omega)$ and the so-called density-density response function $\chi(k, \omega)$ via the famous fluctuation-dissipation theorem (Callen and Welton, 1952):

$$nS(k,\omega)=\mathrm{Im}\{\chi(k,\omega)\} \qquad \omega\geq 0; \quad T=0. \tag{2}$$

In Eq. (2) $\chi''(k,\omega)=\mathrm{Im}\{\chi(k,\omega)\}$ is the spectral function; and $\omega\chi''(k,\omega)\geq 0$ because of the causality. This inequality has a close connection with the irreversible nature of the dissipation.

The longitudinal dielectric function $\varepsilon(k,\omega)$ is defined by the density-density response function:

$$\frac{1}{\varepsilon(k,\omega)}=1-\frac{4\pi}{k^2}\chi(k,\omega), \tag{3}$$

where $(4\pi/k^2)$ is the Fourier transform of the bare Coulomb potential. Generally the stopping power (the energy loss per unit distance) is determined in the following way:

$$\frac{1}{v}\frac{dE}{dt}=\frac{1}{v}\int\frac{d^3\mathbf{k}}{(2\pi)^3}\int d\omega\,\omega P(k,\omega). \tag{4}$$

It is evident that concrete calculations require the knowledge of $\varepsilon(k,\omega)$ or $\chi(k,\omega)$ functions.

Representation

Here we adopt a formally exact representation for $\chi(k, \omega)$, which has the form of the Dyson equation

$$\chi^{-1}(k, \omega)=\chi_0^{-1}(k, \omega)+\frac{4\pi}{k^2}+\phi(k, \omega) \tag{5}$$

well-known in the theory of Green functions. In this equation $\chi_0(k, \omega)$ is the Lindhard function, or free electron polarization propagator (Lindhard, 1954). The long range correlations characteristic of the electron system have been taken into account explicitly by separating off the Coulomb interaction ($4\pi/k^2$). Thus $\phi(k, \omega)$ represents the collisional part of the total effective interaction between density fluctuations. This function is complex; it has an imaginary part because of the "off-the-energy-shell" scattering between electrons of the system.

The connection between $\phi(k, \omega)$ and the so-called dynamic local field correction $G(k, \omega)$ is given by

$$\phi(k, \omega)=-\frac{4\pi}{k^2}G(k, \omega). \tag{6}$$

The function $G(k, \omega)$ represents the short-range correlations between electrons in the homogeneous electron gas.

The total effective potential between electrons is:

$$v_{\text{eff}}(k)\equiv\frac{4\pi}{k^2}[1-G(k, \omega)]. \tag{7}$$

In other words, the interaction between an electron (in the system) and the displaced charge is not merely given by the Hartree term but includes exchange and correlation terms that in fact depend on the electron's momentum and energy. A local deficiency of charge (Pauli and Coulomb hole) is induced by exchange and correlation around the charge density at a given point. Clearly the distortion of the hole by the applied field (by the potential field of the incoming particle) will contribute to the internal local forces determining the density response.

The particular representation for $\varepsilon(k, \omega)$ is given by

$$\varepsilon(k, \omega)=1+\frac{Q_0(k, \omega)}{1-Q_0(k, \omega)G(k, \omega)}, \tag{8}$$

where

$$Q_0(k, \omega)\equiv\frac{4\pi}{k^2}\chi_0(k, \omega), \tag{9}$$

is the Lindhard's expression. In the Random Phase Approximation (RPA) the dielectric function corresponds to an expression given by Eq. (8), but $G=0$.

Stopping for low velocities

For small ω value ($\omega = \mathbf{kv}$) we can write (Nagy, László and Giber, 1985):

$$\chi''(k,\omega) = \left[\chi_0''(k,\omega) + \frac{4\pi}{k^2}\chi_0^2(k)G''(k,\omega) \right] \Big/ \varepsilon_e^2(k) \tag{10}$$

where the static electron dielectric function is

$$\varepsilon_e(k) = 1 + \frac{4\pi}{k^2}\chi_0(k)\,[1 - G(k)]\,. \tag{11}$$

Furthermore

$$\operatorname{Im}\chi_0(k,\omega) = \left(\frac{k_F}{\pi^2}\right)\frac{\pi}{2}\frac{\omega}{kk_F} \qquad \text{for} \quad k \leq 2k_F, \tag{12}$$

$$\chi_0(z) = \left(\frac{k_F}{\pi^2}\right)\frac{1}{2}\left(1 + \frac{1-z^2}{2z}\ln\left|\frac{z+1}{z-1}\right|\right). \tag{13}$$

Here we have introduced the standard notation $z = (k/2k_F)$; and k_F is defined from the density (n)

$$n = \frac{k_F^3}{3\pi^2}\,. \tag{14}$$

The *first* explicit calculation which goes beyond the free electron representation (RPA) of the spectral function was performed *by us* (see Ref. 6). We obtained a very good agreement with high precision data (Blume, Eckstein and Verbeek, 1982) for slow *proton* stopping in Au. This result clearly demonstrates that the interacting nature of the electron bath [the significance of $G''(k,\omega)$] can give measurable deviations with respect to a free electron system for the examined quantity (dE/dR).

Beyond the first Born approximation

Although Eq. (4) is applicable for arbitrary non-relativistic velocities, its validity is questionable in dielectric representation for cases of bombarding particles of high atomic charges, or small density of the system. This is due to the fact that in the examined ($v < v_F$) limit the first Born approximation, implicitly used in *all* dielectric descriptions, is not adequate. In other words the Born series does not converge, according to the Klein-Zemach-Bargmann theorem, because of the appearance of bound states.

On the other hand, this deficiency can be saved by considering the structure of Eq. (10). The *numerator* of this equation is determined by the time dependent properties of fluctuations of the electron gas and may be treated independently of the presence of the incoming ion (long-time behaviour of the excitation spectrum). This argument is very general, and only requires the validity of the assumption that the system is of linear dissipative nature. The *numerator* of Eq. (10) is related to the amount of phase space available for the creation of elementary excitation (particle-hole generation), i.e. is a kinematical constant, independent of the details of the interaction between the external particle and the environment.

The "dressing" (in a transient period) is included in the factor [see Eqs (1), (2) and (10)]

$$|U(k)|^2 \varepsilon_e^{-2}(k). \tag{15}$$

This is essentially the Fourier transform of the screened scattering potential.

We interpret Eq. (15) in terms of the scattering theory by means of the substitution:

$$\left|\frac{U(k)}{\varepsilon_e(k)}\right|^2 \to 4\pi^2 |f(\theta)|^2, \tag{16}$$

where $f(\theta)$ is the single elastic scattering amplitude and $k = 2k_F \sin\theta/2$ is the momentum transfer. Furthermore, we determine $G''(k, \omega)$ (see Eq. 10) according to the prescription of Gross and Kohn (1985):

$$G''(k, \omega) = a(n) \frac{k^2}{4\pi} \omega, \tag{17}$$

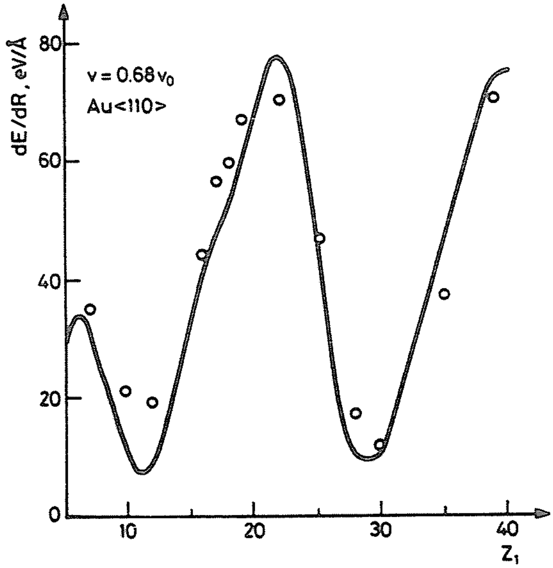

Fig. 1. Stopping power, in units of eV/Å, for best channelled ions (Z_1) in the $\langle 110 \rangle$ channel of Au. The circles are the experimental data. The curve is the theoretical result. See the text for details

where $a(n)$ depends only on the density of the system. Performing the integrations ($\omega = kvy$) in Eq. (4) we obtain

$$\frac{1}{v}\frac{dE}{dt} = \frac{1}{v}\frac{1}{2}\left(\frac{k_F}{\pi}\right)^2 \int_0^1 dy \int_0^\pi d\sigma(\theta, k_F)\left[2vk_F y \sin\frac{\theta}{2}\right]^2\left[1+\frac{12n}{\pi}a(n)\sin\frac{\theta}{2}\right]. \quad (18)$$

In this equation $d\sigma(\theta, k_F)$ is the usual differential cross section:

$$d\sigma(\theta, k_F) = 2\pi |f(\theta)|^2 \sin\theta \, d\theta. \quad (19)$$

Within the above picture a self-consistent calculation is straightforward in the knowledge of scattering phase shifts for the regular, screened potential (Echenique, 1987; Nagy, Arnau and Echenique, 1988).

We have studied the Z_1 (bombarding ion atomic charge) dependence of the stopping power (see Fig. 1) comparing the experimental data of Bøttiger and Bason (1969) for the $\langle 110 \rangle$ channelling direction in gold target with theoretical calculations based on Eq. (18).

The phase shifts are determined for a self-consistently screened potential obtained within the framework of the Density Functional Theory (DFT). The $\langle 110 \rangle$ channel was characterised by the density parameter $r_s = 2$; the ion velocity is $v = 0.68 v_0$. For this particular value of the density parameter ($r_s = 1.92/k_F$)

$$\frac{12n}{\pi} a(n) = 0.30. \quad (20)$$

The calculated phase shifts satisfy the Friedel sum rule with high accuracy. The theoretical results (see Ref. 12) show rather good agreement with the experimental data.

Acknowledgement

With this short communication the author would like to express his gratitude for continuous, stimulating discussions and conversations to Prof. János Giber.

References

1. Fetter, A. L.—Walecka, J. D. (1971): Quantum Theory of Many-Particle Systems (McGraw-Hill, New York).
2. Mahan, G. D. (1981): Many-Particle Physics (Plenum, New York).
3. Van Hove, L. (1954): Phys. Rev. *95*, 249.
4. Callen, H. B.—Welton, R. F. (1952): Phys. Rev. *86*, 702 (1952).

5. LINDHARD, J. (1954): K. Dan. Vidensk. Selsk. Mat. Fys. Medd. *28*, n° 8.
6. NAGY, I., LÁSZLÓ, J.—GIBER, J. (1985): Z. Phys. *A321*, 221.
7. BLUME, R., ECKSTEIN, W.—VERBEEK, H. (1982): Nucl. Instr. Meth. *194*, 67.
8. GROSS, E. K. U.—KOHN, W. (1985): Phys. Rev. Lett. *55*, 2850.
9. ECHENIQUE, P. M. (1987): Nucl. Instr. Meth. *B27*, 256.
10. NAGY, I., ARNAU, A.—ECHENIQUE, P. M. (1988): Phys. Rev. *B38*, 9191.
11. BØTTIGER, J.—BASON, F. (1969): Rad. Effects *2*, 105.
12. NAGY, I., ARNAU, A.—ECHENIQUE, P. M. (1989): Phys. Rev. *A40*, 987.

Dr. István NAGY H-1521, Budapest