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VICINAGE EFFECT IN SLOW-ION-DICLUSTER STOPPING POWER

I. NAGY *, A. ARNAU and P.M. ECHENIQUE

Departamento de Física de Materiales, Universidad del País Vasco, Apartado 1072, San Sebastián 20080, Spain

Approximations to treat the screening and scattering of a slow dicluster by independent electrons of a homogeneous system are used to investigate the vicinage effect in the stopping power for the composite. A partial wave expansion is employed for the scattering amplitudes to treat the coherent elastic scattering of the electrons by the two independent scattering centers. For proton dicluster stopping, a simple analytical expression is obtained in the one-phase-shift approximation. Good agreement with the available experimental data is found.

1. Introduction

Accurate values of electronic energy losses of ions in metals are extremely valuable in various branches of science and technology. The theoretical description for homogeneous electron gases, to represent valence electrons in metals, is well developed in the case of atomic ions. For this type of ions the agreement between theoretical and experimental results is good in the high-velocity range. At lower velocities the agreement becomes poorer. The discrepancy in this velocity region is mainly due to the lack of knowledge of the stopping function. The standard methods for low velocities ($v < v_F$, v_F being the Fermi velocity) are the dielectric formalism [1] and the scattering (kinematical) approach [2]. The latter (with correct phase-shift analysis) usually gives better agreement with the experimental data [4].

One may expect that in the case of molecular ions new effects appear. Because of the closeness of the constituents in the bombarding cluster, interference effects in the scattering process must be taken into account. The dielectric formalism has been commonly used [5] to treat high-velocity molecular ion stopping. For these velocities interference effects are (mainly) the result of the coherent excitation of the wake of electron density fluctuations trailing the fast charged particles.

Little attention has been paid to the case of low-velocity molecular ion stopping. The calculations are restricted to the dielectric formalism [6]. The relative velocity of the colliding partners is moderate at metallic densities of the electron gas. Thus the validity of the first Born approximation (implicitly used in all dielectric description) is questionable for typical screened potentials in this density range. On the other hand the theories to treat the electron–molecule scattering at vacuum conditions are well developed [7].

Our aim with this short paper is to investigate the spatial configuration (vicinage) effect in slow-dicluster stopping in a homogeneous electron gas. We combine electron–molecule scattering methods, for randomly oriented constituents of the beam, with kinematical constraints to include the influence of the environment. We shall use atomic units throughout this paper.

* Permanent address: Quantum Theory Group, Institute of Physics, Technical University of Budapest, 1521 Budapest, Hungary.

2. Theoretical model

We employ idealized models in order to characterize vicinage effects in condensed matter. The energy loss per unit path length of a slow dicluster (dE/dR) is examined using a quantum mechanical scattering description of the excitations (electron–hole pairs) of a homogeneous electron gas system. Our assumptions to treat the elastic scattering of electrons at the Fermi level (kinematical constraint; Pauli exclusion principle) by the dicluster are:

(i) each spherically screened constituent of the dicluster scatters independently;
(ii) multiple scattering with the dicluster is negligible (on-the-energy-shell representation; coherent scattering);
(iii) the incident beam "particles" are randomly oriented (this is the usual experimental situation);
(iv) the cluster velocity remains essentially constant.

Within these assumptions, in the examined low-velocity limit, our problem becomes very similar to the case of atomic ion stopping [2]. Using similar ideas to those in ref. [3] we can write:

$$\frac{dE}{dR} = n_0(vk_F)\int_0^{\pi}(1-\cos\theta)\,d\sigma(\theta, k_F), \qquad (1)$$

where n_0 is the density of the system and $d\sigma$ is the

differential cross section. The factor $(1-\cos\theta)$ corresponds to the usual weighting factor suppressing the contribution from scattering in the forward direction, and $k_F=(3\pi^2 n_0)^{1/3}$. If we use, as a particular realization, the first Born approximation for the scattering amplitudes we arrive at the result previously obtained by Arista [6].

For a randomly oriented dicluster (not neccesarily homonuclear) the differential cross section is given by [7]:

$$\frac{d\sigma}{d\Omega}=|f_1|^2+|f_2|^2+(f_1f_2^*+f_2f_1^*)\frac{\sin(kR)}{kR}, \quad (2)$$

where the f_i's are the scattering amplitudes, f^* denotes the complex conjugate of f, $k=2k_F\sin(\theta/2)$ is the momentum transfer, θ is the scattering angle, and R is the distance between the cluster constituents and $d\Omega=2\pi\sin\theta\, d\theta$. In partial wave expansion the expression for the scattering amplitude is [8]:

$$f(\theta)=\frac{1}{2ik_F}\sum_l(2l+1)(e^{2i\delta_l}-1)P_l(\cos\theta), \quad (3)$$

where δ_l's are the phase-shifts (at the Fermi level) and the P_l's are the Legendre polynomials. From eq. (3) the standard calculation yields:

$$|f|^2=\frac{1}{4k_F^2}\sum_l\sum_n(2l+1)(2n+1)B(\delta_l,\delta_n)\times P_l(\cos\theta)P_n(\cos\theta), \quad (4)$$

where

$$B(\delta_l,\delta_n)=1-\cos(2\delta_l)-\cos(2\delta_n)+\cos[2(\delta_l-\delta_n)].$$

In the dielectric formalism [1,6], which corresponds to a first Born approximation, one can write for the scattering amplitude:

$$f(k)=\frac{2Z_i}{k^2\epsilon(k)}, \quad (5)$$

where $\epsilon(k)$ is the static dielectric function [1] and Z_i $(i=1,2)$ is the charge of a bare constituent of the dicluster [6]. In first Born approximation the scattering amplitude is a pure real function.

By using the well-known expansion [8]

$$\frac{\sin(kR)}{kR}=\sum_m(2m+1)j_m^2(k_FR)P_m(\cos\theta), \quad (6)$$

where the j_m's are spherical Bessel functions of the first kind, together with eqs. (1)–(4) the stopping for the dicluster can be calculated.

3. Results for proton dicluster

The remaining problem, as is usual in the scattering approach, is the determination of the scattering potential which yields to the phase-shift values. In the dielectric formalism (static linear screening) the potential is immediately given.

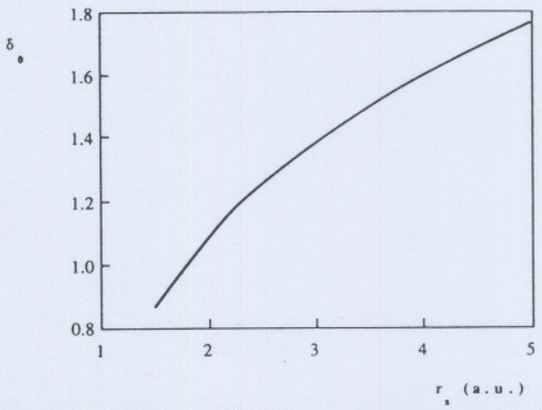

Fig. 1. Leading phase-shift (δ_0) as a function of the density parameter r_s.

The screening *nonlinearities* are well described [4] in the framework of density functional theory (DFT), in a self-consistent way. Let us briefly summarize some results of this theory. In the case of protons, at metallic densities ($k_F\cong 1$), the leading ($l=0$) phase-shift gives a good approximation for the scattering amplitude. In fig. 1 we have plotted this leading phase-shift (δ_0) as a function of the density parameter $r_s=(9\pi/4)^{1/3}/k_F$. In the density range of interest δ_0 is not small and it is not far from the unitary limit ($\delta_0=\pi/2$), which simply comes from the Friedel sum rule [9]. This rule is a self-consistency condition on the screened scattering potential [10]. Furthermore, the static charge density profiles are close to hydrogenic 1s wave functions [11].

Motivated by *these* physical statements and to get easily surveyed analytical results, we use an extra assumption:

(v) only the dominating phase-shift ($l=0$) gives an essential contribution ("one-phase-shift" approximation).

Using eqs. (1), (2) and (6) we write in our approximation [see (i)–(v)] the stopping power for a slow "homoatomic" (screened proton) dicluster as:

$$\frac{dE}{dR}=\left[n_0(vk_F)\frac{4\pi}{k_F^2}\sin^2\delta_0\right][2+G(k_FR)], \quad (7)$$

where

$$G(k_FR)=\sum_{m=0}^{1}(2m+1)j_m^2(k_FR)\times\int_0^\pi d\theta\,\sin\theta(1-\cos\theta)P_m(\cos\theta). \quad (8)$$

The so-called vicinage function (g) measures the spatial

configuration effect, and it is given by ($g \equiv G/2$; and $x = k_F R$):

$$g(x) = j_0^2(x) - j_1^2(x). \tag{9}$$

The first term of eq. (7) is the stopping power for a slow proton. In the $x \to \infty$ limit the stopping power for the dicluster is twice this quantity, because of the asymptotic form

$$\lim_{x \to \infty} g(x) = \frac{-\cos(2x)}{x^2}.$$

It is easy to show that the second term in the right-hand side of eq. (9) originates from the cos θ factor of eq. (1); and $\max\{m\} = 1$ in eq. (8) is due to assumption (v). The first three zeros of $g(x)$ are at x-values of 2.043, 3.812 and 5.396 respectively. The first minimum is about at $x = 2.745$, for which $g(2.745) \simeq -0.130$.

To show the validity of the "one-phase-shift" approximation, in fig. 2 we have plotted the stopping power for a slow individual proton as a function of r_s. Curve (a) is the result of the "one-phase-shift" ($l = 0$) approximation (first term of eq. (7)), while curve (b) is obtained by taking into account phase-shifts of higher l, in the general expression [2]:

$$n_0(vk_F)\frac{4\pi}{k_F^2}\sum_l (1+1)\sin^2(\delta_l - \delta_{l+1}),$$

within the same density functional approach [4]. The comparison between the two curves shows that within the uncertainty of the measurements (10–15%) the "one-phase-shift" approximation is adequate, at metallic densities of the electron gas.

The general behaviour of the vicinage function $g(k_F R)$ in our approximation [see points (i)–(v)] is shown in fig. 3. The curves are calculated for $2 \le r_s \le 5$ as a function of the separation distance R. The results

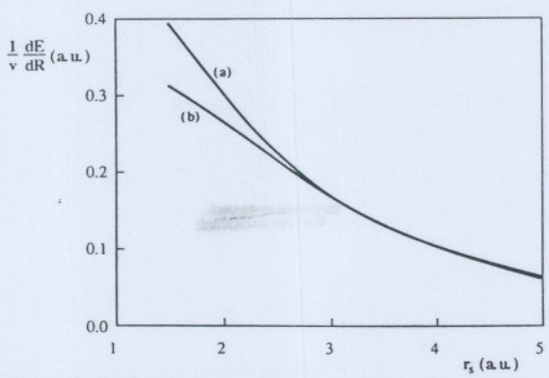

Fig. 2. Individual proton stopping power as a function of r_s. Curve (a) is the result of the one-phase-shift approximation. Curve (b) is obtained taking into account phase-shifts of higher order l.

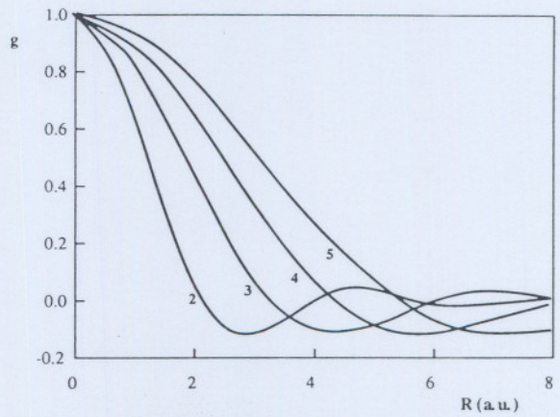

Fig. 3. The vicinage function g as a function of the separation distance between the cluster constituents R. The corresponding values of the density parameter r_s are indicated on the curves.

show that, for low velocities, interference effects in the dicluster stopping power may be important. Particularly we have obtained good agreement with the experimental data [12] for slow H_2^+ stopping in aluminium ($r_s = 2$). For our estimates we have used the value $R_0 = 2.44$ for the separation distance [5,12]; $g(k_F R_0) \cong -0.11$. For $v = 0.7$ (12.5 keV per nucleon) the measured value for the ratio $(1 + g)$ of the stopping powers of the dicluster and two separate protons is about 0.87 (see fig. 2 of ref. [12]).

4. Summary and comments

In this short paper we have developed a simple model to treat slow-dicluster stopping in a homogeneous electron gas. Within the applied assumptions (i)–iv) eqs. (1), (2) and (6) give an exact description for the examined quantity (dE/dR). To obtain simple analytical results for proton diclusters we have used the dominating "one-phase-shift" (additional) approximation. Our vicinage function (g) reproduces in a proper way the available experimental data.

Because the typical potential ranges ($\cong 1$ a.u.) are comparable to the mean separation distance ($R \cong 2.5$ a.u.), from a theoretical point of view the multiple-scattering effects (off-the-energy-shell matrix elements) need a careful examination.

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References

[1] J. Lindhard, K. Dan. Vidensk. Selsk. Mat. Fys. Medd. 28 (1954) no. 8.

[2] A. Blandin, A. Nourtier and D.W. Hone, J. Phys. (Paris) 37 (1976) 369;
P. Sigmund, Phys. Rev. A26 (1982) 2497;
K. Schönhammer, Phys. Rev. B37 (1988) 7735;
I. Nagy, A. Arnau and P.M. Echenique, Phys. Rev. B38 (1988) 9191.

[3] T.L. Ferrell and R.H. Ritchie, Phys. Rev. B16 (1977) 115.

[4] P.M. Echenique, R.M. Nieminen and R.H. Ritchie, Solid State Commun. 37 (1981) 779;
P.M. Echenique, R.M. Nieminen, J.C. Ashley and R.H. Ritchie, Phys. Rev. A33 (1986) 897;
P.M. Echenique, Nucl. Instr. and Meth. B27 (1987) 256;
P.M. Echenique, F. Flores and R.H. Ritchie, to be published in Solid State Physics Series, no. 53, eds. H. Ehrenreich and D. Turnbull.

[5] G. Basbas and R.H. Ritchie, Phys. Rev. A25 (1982) 1943;
J.C. Ashley and P.M. Echenique, Phys. Rev. B35 (1987) 8701, and references therein.

[6] N.R. Arista, Phys. Rev. B18 (1978) 1.

[7] N.F. Mott and H.S.W. Massey, The Theory of Atomic Collisions (Clarendon, Oxford, 1965) vol. I, pp. 194–209.

[8] L.I. Schiff, Quantum Mechanics (McGraw, Singapore, 1985) p. 119 and p. 329.

[9] J. Friedel, Philos. Mag. 43 (1952) 153;
J.S. Langer and V. Ambegaokar, Phys. Rev. 121 (1961) 1090.

[10] C. Kittel, Quantum Theory of Solids (Wiley, New York, 1987) p. 343.

[11] C.O. Almbladh, U. von Barth, Z.D. Popovic and M.J. Stott, Phys. Rev. B14 (1976) 2250.

[12] J.C. Eckardt, G. Lantschner, N.R. Arista and R.A. Baragiola, J. Phys. C11 (1978) L851.