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Nonlinear Effects in Interactions of Swift Ions with Solids

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1. Introduction

The passage of a swift charged particle through a solid gives rise to a wake of induced electron density behind the particle. Figure 1 shows how the wake would look to an observer in the solid, behind the projectile. It is calculated for a proton penetrating an electron gas having the density of the valence electrons in gold, assuming linear response of the medium.

The induced potential associated with the wake is responsible for the energy loss of the particle, and for many effects that have captured recent interest. These include, among others, vicinage effects on swift ion clusters, emission of electrons from bombarded solids, forces on swift ions near a surface, and energy shifts in electronic states of channeled ions. Furthermore, the wake has a determining influence on the spatial distribution, and character, of energy deposition in the medium. (For reviews, see references [1],[2],[3]). Previous theoretical studies of these phenomena (with the one exception mentioned below [4]) have employed a linear wake, i.e., one that is proportional to the charge of the projectile, eZ. However, in most experiments that measure these effects, the conditions are such that the wake must include higher-order terms in Z. The purpose of this study is to analyze the *nonlinear* wake, to understand how the linear results must be revised.

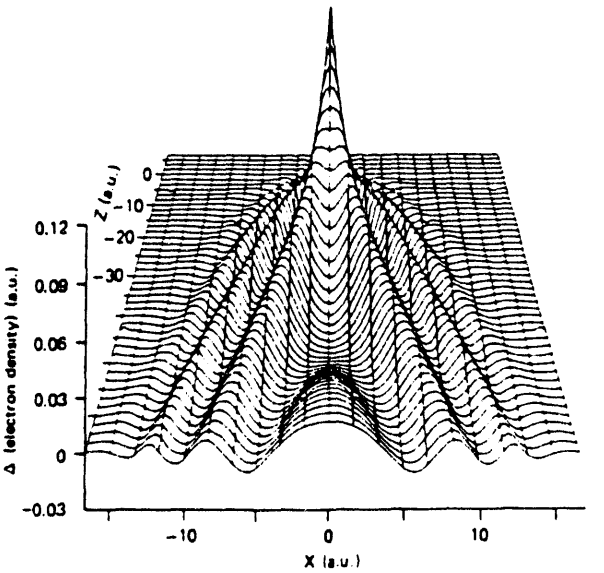

Fig. 1. The wake of induced density for a proton in gold, treated as an electron gas and assuming linear response.

The stopping power is given by the gradient of the induced potential at the position of the ion, multiplied by the charge of the latter. Therefore, the linear wake gives a stopping power proportional to Z^2, while the first nonlinear term in the wake produces a Z^3 term. The latter is commonly called the Barkas [5] correction. Its value was first estimated by Ashley, Ritchie, and Brandt [6], from a classical treatment of distant collisions of a charged particle with a harmonically bound electron. Their result was subsequently confirmed in a quantum mechanical treatment of the harmonic oscillator by Hill and Merzbacher [7].

The first treatment of a nonlinear wake was performed by Faibis *et al.* [4]. These workers calculated the Coulomb wake, i.e., the induced density (and potential) due directly to Coulomb scattering of independent, noninteracting electrons from a charged projectile.

Sung and Ritchie [8] applied many-body perturbation theory to the Fermi electron gas, using a random phase approximation (RPA). There resulted expressions for the second-order wake and for the Barkas correction. Numerical evaluation was discussed, but computations were not carried out.

Recently, Esbensen and Sigmund [9],[10] have extended Lindhard's self-consistent, linear treatment of the electron gas [11] to second order in an external disturbance, deriving formulae for the nonlinear wake and the Barkas effect for several model systems. The results for the Fermi gas are identical to those of Sung and Ritchie [8]. Computations were presented for both the nonlinear wake potential and the Barkas correction in an electron gas. However, because of the complexity of the formulae, it was necessary to ignore Fermi motion of the electrons, thus specializing the numerical results to a static electron gas.

Only within the last year were calculations finally carried out, by Pitarke *et al.* [12], [13] for the Barkas correction in the RPA for a Fermi gas. The basic formulae used are equivalent to those of references [8] and [9]. Numerical evaluations of the second-order wake in this theory have not yet been achieved.

2. Hydrodynamical Model

The application of hydrodynamics should prove fruitful in this problem, for various reasons. First, it is simpler than the RPA, and should therefore enable one to carry out calculations that—as mentioned above—have not been possible in the RPA. Second, a close connection exists between hydrodynamics and density functional theory, as discussed below. Finally, new insights might emerge.

The system of interest is a fluid of electrons on a uniform background of positive charge that neutralizes the total charge. This fluid flows steadily past a point charge eZ fixed at the origin. Isentropic, irrotational flow is assumed. The mass density and the velocity are described by continuous functions of position, $\rho(\vec{r})$ and $\vec{v}(\vec{r}) = -(\nabla\psi)$ respectively, where ψ is the velocity potential. The classical Lagrangian density, $\mathcal{L}$, is given by the expression [14]

$$\mathcal{L} = \rho \frac{\partial \psi}{\partial t} - \frac{1}{2} \rho (\nabla \psi)^2 - \rho U - \rho \mathcal{E} \tag{1}$$

where $U(\vec{r})$ and $\mathcal{E}[\rho] = \mathcal{E}(\rho, \nabla\rho)$ are respectively the potential energy and the internal energy per unit mass.

The potential energy per unit mass U is given by the product of the charge-to-mass ratio $-e/m$ and the electrical potential. It consists of two terms, one from the external potential (due directly to the projectile charge) and one from the induced potential in the fluid itself, as follows

$$U(\vec{r}) = U_{ext}(\vec{r}) + \left(\frac{e}{m}\right)^2 \int \frac{\rho(\vec{r}') - \rho_0}{|\vec{r} - \vec{r}'|} d^3r', \tag{2}$$

where ρ_0, the density at large distances, is subtracted from ρ to account for the background of positive charge.

The Euler-Lagrange equations of $\mathcal{L}$ are obtained by requiring that

$$\delta \int_{t_1}^{t_2} dt \int \mathcal{L}(n, \nabla n, \psi, \nabla \psi) d^3r = 0 \tag{3}$$

for fixed $n(\vec{r})$ and $\psi(\vec{r})$ at times t_1 and t_2, where the inner integral extends over all space. There result two equations: the continuity equation,

$$\frac{\partial n}{\partial t} - \nabla \cdot (\rho \nabla \psi) = 0, \tag{4}$$

and the Bernoulli equation,

$$\frac{\partial \psi}{\partial t} = \frac{1}{2} (\nabla \psi)^2 + U_{ext}(\vec{r}) + \left(\frac{e}{m}\right)^2 \int \frac{\rho(\vec{r}') - \rho_0}{|\vec{r} - \vec{r}'|} d^3r' + \frac{\delta(\rho \mathcal{E})}{\delta \rho} - \mu, \tag{5}$$

where μ is the chemical potential, a constant. In the present case, steady flow past a stationary point particle of charge Ze, both $\partial n/\partial t$ and $\partial \psi/\partial t$ vanish identically, and U_{ext} is given by $-(e/m)(Ze/r)$.

Up to this point, the theory is entirely classical. Furthermore, no property of the electron except the charge-to-mass ratio has entered the equations. Is it reasonable to apply such a theory to a (quantum mechanical) electron gas? The answer is: Yes, at least in principle. The above two Euler-Lagrange equations of the hydrodynamical model can be derived from time-dependent density-functional theory, by considering an adiabatic

perturbation of the ground state of the system [15]. In the context of density functional theory, $\mathscr{E}[n]$ is the electronic energy (excluding the potential energy U) per unit mass, as a functional of the (exact) single-particle density $n(\vec{r})$. The description of the fluid as a quantum-mechanical gas of electrons must evidently be embodied in $\mathscr{E}[n]$. Although the exact functional is not known in advance, many useful results have been achieved by suitable approximations for $\mathscr{E}[n]$.

The first application of hydrodynamics to a system of electrons was Bloch's derivation [16] of a formula for the stopping power of many-electron atoms. Close collisions were treated the same as in Bethe-Bloch theory, while hydrodynamics was assumed to apply to large-impact-parameter collisions. The Thomas-Fermi energy functional, equal to the kinetic-energy per unit mass of a degenerate Fermi gas,

$$\mathscr{E}_{TF}(n) = \frac{3\hbar^2}{10m^2}(3\pi^2)^{2/3}n^{2/3}, \tag{6}$$

was assumed, where $n(\vec{r}) = \rho(\vec{r})/m$ is the number density. Bloch worked with the linearized equations, obtained from Eqs. (4), (5), and (6) by expanding n and ψ about their initial values, and retaining only the terms that are linear in the disturbance. Since then, hydrodynamical models (using various forms of the internal-energy functional) have been applied numerous times to atoms and to the inhomogeneous electron gas, mainly in studies of collective modes [17], [18].

In the calculations [19] reported here, the Thomas-Fermi-Weizsäcker (TFW) functional is employed:

$$\mathscr{E}_{TFW}(n, \nabla n) = \frac{3\hbar^2}{10m^2}(3\pi^2)^{2/3}n^{2/3} + \frac{\hbar^2}{8m^2}\frac{(\nabla n)^2}{n^2}, \tag{7}$$

in which the first term is the kinetic energy (per unit mass) of a degenerate Fermi gas, while the second one is the gradient correction proposed by von Weizsäcker [20]. This functional when used in atomic structure calculations has been shown to produce an electron density that satisfies the proper cusp condition at the nucleus (r = 0) and has the correct asymptotic form as r goes to infinity [21]. Similarly, it leads to a static response function having the correct values at wave number $k = 0$ and (asymptotically) as $k \to \infty$ [22].

3. Calculations and Discussion

We have made a first set of calculations of the nonlinear wake and of the Barkas correction to the stopping power using this model [19]. The electron density and the velocity

potential are formally expanded in powers of Z, and the Euler-Lagrange equations for $\mathcal{L}$ are solved numerically for both the 1st- and 2nd-order parts of the density and the velocity potential. The Z^2 and Z^3 terms in the stopping power are then computed.

First, consider the linear-response stopping power. We find [19] that energy-loss functions $\mathrm{Im}(-1/\varepsilon[\omega,k])$ in the TFW hydrodynamical model and the RPA essentially agree at both $k/k_F \ll 1$ and $k/k_F \gg 1$, where k_F is the Fermi wave number. Furthermore, $\mathrm{Im}(-1/\varepsilon[\omega,k])$ satisfies Bethe's generalized oscillator-strength sum rule for all k. These conditions suffice to guarantee [23] that the stopping power (in linear response) approaches that of the RPA with increasing velocity, above the Fermi velocity. This is borne out in Figure 2, where the two theories are compared for an electron gas of $r_s = 2.07$ a.u. [24] (Fermi velocity is 0.9 a.u.). Note from the figure that for velocity $v<0.9$ a.u., the hydrodynamical model predicts zero stopping power. This is because low-energy electron-hole excitations, which are responsible for low-velocity electronic stopping, are not contained in this model.

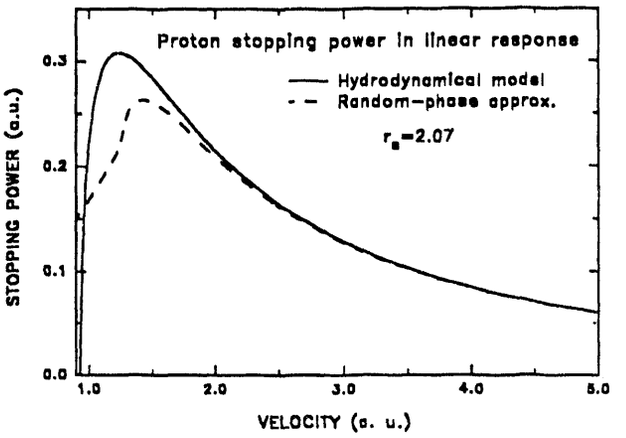

Fig. 2. Comparison of stopping powers from the TFW hydrodynamical model and the RPA, considering only linear response.

Figure 3 shows stopping powers of an electron gas with $r_s = 2$ a.u. in the hydrodynamical model, for protons and antiprotons. The dashed curve is the linear result. The Barkas correction is seen to be substantial, even for $Z = \pm 1$.

In Figure 4, the Barkas corrections from four calculations are compared for $r_s = 2$ [25]. The short-dashed curve is the Ashley-Ritchie-Brandt formula [6], with Jackson and McCarthy's value [26] for the minimum impact parameter. The oscillator frequency is set equal to the plasmon frequency of the electron gas, and the result is multiplied by 2 as suggested by Lindhard

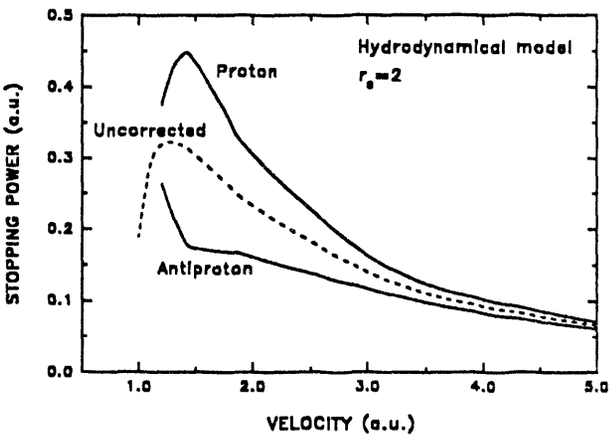

Fig. 3. Proton and antiproton stopping in an electron gas in the TFW hydrodynamical model, including the Barkas correction.

[10]. The long-dashed curve is taken from the static-gas calculation of Esbensen and Sigmund [9]. Note that this calculation and twice the Ashley-Ritchie-Brandt one agree over most of the velocity range covered in the figure. The medium-dashed curve is the RPA result of Pitarke and coworkers [13]. The above theories agree for velocities greater than 2 a.u. Finally, the solid curves are the present TFW hydrodynamical values. (The Z^2 term is shown in order to indicate the relative importance of the Barkas correction.) In the neighborhood of its maximum, the present result resembles the RPA one.

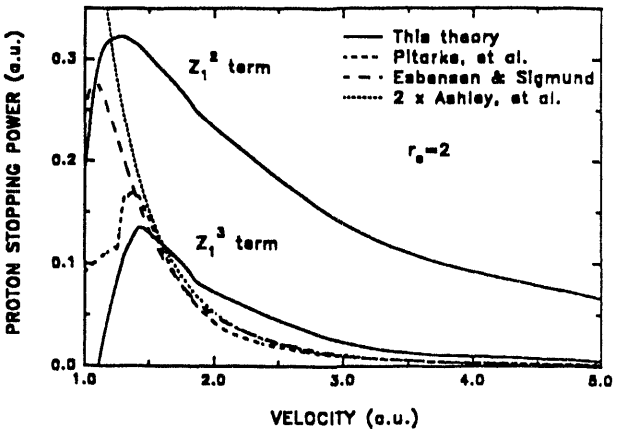

Fig. 4. Comparison of Barkas corrections to proton stopping power from various theories, as functions of velocity. See text.

A comparison of the four theories is shown in Figure 5, for $r_s = 2$ [25], at velocities of 2-5 a.u. The present results clearly lie above the others in this velocity range, whereas the latter appear to approach a common curve. Also shown (circles) are the experimental values of the Barkas correction for protons in Si , calculated as half the difference between the stopping powers of protons and antiprotons [27]. (Counting only the four valence electrons, Si has $r_s = 2.01$). Note that the hydrodynamical results are consistent with the experiment. However, this comparison with experiment is questionable, since it neglects the inner-shell electrons. When the RPA result is averaged over the local electron density $n(\vec{r})$ in Si, using a local plasmon approximation, it increases substantially and comes into agreement with these experimental values [13].

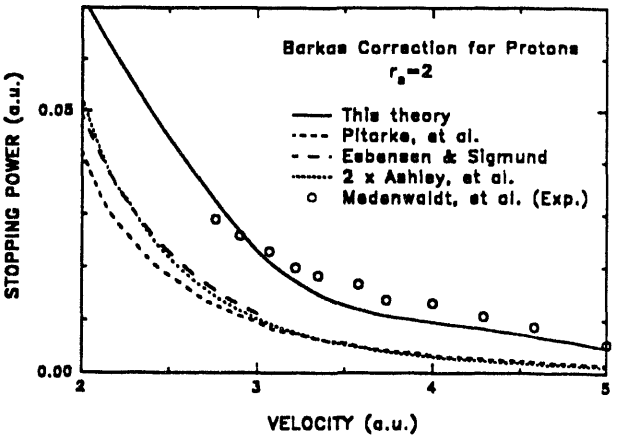

Fig. 5. Comparison of Barkas corrections to proton stopping power from various theories, as functions of velocity. The circles are experimental results for Si. Tee text.

We have seen that the RPA and TFW hydrodynamics both give the correct stopping power for the electron gas in the high-velocity, nonrelativistic limit. Thus it is surprising that

the Z^3 effects in the two theories differ so much in the same limit. It is not obvious which is the better result. This question is under investigation.

Acknowledgments

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