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2010 J. Phys.: Condens. Matter 22 015602

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# The screening and stopping coefficients of slow light ions

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Received 20 August 2009, in final form 6 November 2009

Published 2 December 2009

Online at [stacks.iop.org/JPhysCM/22/015602](http://stacks.iop.org/JPhysCM/22/015602)

## Abstract

We present a theoretical approach to study the screening charge density  $n_s(r)$  and the respective stopping coefficient  $Q$  for hydrogen and helium at the low velocity limit. An electron gas, with electronic density  $n_e$ , is used to represent the conduction or valence electrons of the target material. Solving numerically the Schrödinger radial equation, for a given potential  $V(r)$ , the phase shifts  $\delta_l$  and the corresponding stopping coefficient  $Q$  are calculated as a function of  $n_e$ . The cusp condition and the Friedel sum rule are imposed on the charge density  $n(r) = n_s(r) + n_e$  at the origin and to the phase shifts, respectively. The results are compared with density functional calculations and with available experimental results.

The interaction of external charged particles with the free electrons of a metal is a problem of relevance for numerous studies dealing with impurities in solids, radiation effects and materials analysis. In the last few decades, several studies have been made using the method of density functional theory (DFT), producing accurate results that serve as a point of reference for related studies. Still, there is a need for simplified methods that could be used in different alternative studies (like several processes of interaction of slow ions with solids, molecular dynamics codes, ion implantation, range distributions, radiation effects and others) and could provide fast and reliable results. A simplified method of calculation has been proposed by Ferrell and Ritchie for slow He ions in a free-electron gas [1], using a parameterized interaction potential which was adjusted to satisfy the Friedel sum rule. This method was further applied in [2, 3] to protons and heavier ions, respectively. The method was also extended by Apagyí and Nagy [4] to take into account the additional restriction imposed by the Kato cusp condition. This approach yields fairly good values of the stopping coefficient  $Q$  in comparison with those obtained using the DFT formalism [5] (with differences of about 5–10%). However, the differences in the values of the induced density on the impurity  $n_s(0)$  with those obtained by more exact calculations [6–8] are very large.

One remarkable aspect of the DFT is that the quantum mechanical problem of determining the equilibrium state of a many-body system may be solved if the ground state density of the electrons is found. In this work we study a method to calculate the most representative terms that characterize the interaction of an external charge with a free-electron gas

(FEG), taking as the input variable a given screening density function  $n_s(r)$ , together with the corresponding screening potential  $V_s(r)$ . The appropriate form of  $V_s(r)$  is searched on the basis of physical criteria and taking into account two main conditions imposed by the Friedel sum rule [9] and the Kato cusp condition [10]. The objective of this study is to provide a simplified screening potential  $V_s(r)$  and associated screening density  $n_s(r)$  which could reproduce with accuracy the relevant values of the stopping coefficient  $Q$ , screening density at the origin  $n_s(0)$  and the value of the main phase shift  $\delta_0$ , in close agreement with the DFT values [5]. Additionally, we also compare the results with experimental results for the stopping coefficient  $Q$ . The valence and conduction electrons of the material are represented as an electron gas characterized by its mean electronic density  $n_e$  and, alternatively, as a nonuniform gas with density  $n_e(r)$ .

The calculation method applied here is the following. The screening density around an impurity ion is represented in terms of analytical functions  $f_i(b_i r)$  in the form

$$n_s(r) \equiv \sum_i^n a_i f_i(b_i r), \quad (1)$$

with appropriate coefficients  $a_i$ , where  $\sum_i a_i = Z$  and with  $\int_0^\infty f_i(b_i r) 4\pi r^2 dr = 1$ . In the following, atomic units will be used.

As a first condition, we require that the total density, given by  $n(r) \equiv n_s(r) + n_e$ , shall satisfy the Kato cusp relation:

$$\frac{n'(0)}{n(0)} = -2Z \quad (2)$$

which provides a mathematical constraint on the density function  $n_s(r)$ .

The generated electrostatic potential  $V_s(r)$  is found by solving

$$V_s(r) = \frac{1}{r} \int_0^r n_s(r') 4\pi r'^2 dr' + \int_r^\infty \frac{n_s(r')}{r'} 4\pi r'^2 dr' \quad (3)$$

and the total scattering potential is

$$V(r) = -\frac{Z}{r} + V_s(r). \quad (4)$$

Then, we consider the scattering of free electrons on this potential and numerically solve the corresponding Schrödinger equation. In this way we can determine the values of the phase shifts  $\delta_l(v_e)$ , where  $v_e$  is the electron speed.

The second relevant condition is provided by the Friedel sum rule, namely

$$Z = \frac{2}{\pi} \sum_{l=0}^{\infty} (2l+1) \delta_l(v_F), \quad (5)$$

which imposes a mathematical condition on the values of the phase shifts at the Fermi velocity  $v_F$ .

Using the phase shift values so obtained we calculate the transport cross section  $\sigma_{tr}$  by

$$\sigma_{tr} = \frac{4\pi}{v_F^2} \sum_{l=0}^{\infty} (l+1) \sin^2[\delta_l(v_F) - \delta_{l+1}(v_F)], \quad (6)$$

and finally we determine the stopping coefficient  $Q$  using the relation [1, 11, 12]

$$Q \equiv -\frac{1}{v} \frac{dE}{dx} = n_e m v_F \sigma_{tr}. \quad (7)$$

In a simplified approach, the problem of an  $H^+$  ion in jellium can be treated in a straightforward way assuming that the screening density has an exponential dependence on the distance  $r$ , namely  $n_s(r) = (1/8\pi)b^3 e^{-br}$  [2] (that is,  $n = 1$ ,  $a_1 = 1$  and  $b_1 = b$  in equation (1)). The potential generated by this screening charge (equation (3)) is then

$$V_s(r) = \frac{1}{r} - \frac{1}{r} e^{-br} \left(1 + \frac{1}{2} br\right). \quad (8)$$

The potential given by equation (8) is similar to that of the 1s hydrogenic state but the screening parameter  $b$  is now a function of the electronic density (to satisfy the Friedel sum rule). Notice that the approach in this case contains a single variational parameter  $b$  and so the calculation ends up when the value chosen for  $b$  leads to phase shift values  $\delta_l(v_F)$  that satisfy equation (5). There is no room in this simplified approach for taking into account the Kato cusp condition which deals with the values of the screening density near the impurity ion.

In order to extend and improve the agreement with more refined theoretical (DFT) values, we extend the analysis to the case where the density function in equation (1) contains three terms. The first two terms  $f_{1,2}$  in equation (1) are screening charges of the same kind as the 1s and 2s hydrogenic states,

with parameters  $b_1$  and  $b_2$ , respectively. Hence, the density function takes the form

$$n_s(r) = a_1 \frac{b_1^3}{8\pi} e^{-b_1 r} + a_2 \frac{b_2^3}{32\pi} (2 - b_2 r)^2 e^{-b_2 r} + a_3 f_3(r). \quad (9)$$

The third function  $f_3$  in this equation was chosen after extensive numerical study with different trial functions. From this analysis we found that very good results could be obtained using one of the simplest trial forms:

$$f_3(r) \equiv \frac{b_3^5}{96\pi} r^2 e^{-b_3 r}. \quad (10)$$

The potential  $V_s$ , calculated using equation (3), becomes now

$$V_s(r) = \frac{1}{r} \left( Z - \sum_i^3 a_i p_i(r) e^{-b_i r} \right), \quad (11)$$

where

$$p_1(r) = 1 + \frac{b_1}{2} r, \quad (12)$$

$$p_2(r) = \frac{(b_2 r)^3}{8} + \frac{(b_2 r)^2}{4} + \frac{3b_2 r}{4} + 1 \quad (13)$$

and

$$p_3(r) = \frac{(b_3 r)^3}{24} + \frac{(b_3 r)^2}{4} + \frac{3b_3 r}{4} + 1. \quad (14)$$

Hence, the potential  $V(r)$  in equation (4) becomes

$$V(r) = -\frac{1}{r} \sum_i^3 a_i p_i(r) e^{-b_i r}. \quad (15)$$

It should be noted that the self-consistent potential used in the DFT formulation contains the exchange and correlation term, which we do not consider specifically here. Hence, the functions used here, equations (9) and (15), may be regarded as effective densities and potentials that serve to approximate the DFT results.

Using the cusp condition of equation (2) for  $n(r)$  we find a relation between coefficients:

$$a_1 = \frac{(Z - a_3)(Z - b_2)b_2^3 + 8Z\pi n_e}{(Z - b_2)b_2^3 - (Z - \frac{b_1}{2})b_1^3}, \quad (16)$$

where  $n_e$  is the normal density of the jellium in the absence of the impurity ion.

The numerical studies covered initially a wide range of parameters, but after many tests we found that the region of values with  $b_2 = b_1$  produced very good general agreement with the theoretical DFT results. The values of the terms  $a_i$  are constrained by the condition  $\sum_i a_i = Z$  and by the Kato condition of equation (16), so that only one of the  $a_i$  may be considered as a free parameter. Hence, we considered a fine mesh of trial values of  $b_3$  and  $a_3$  around the optimal region and adjusted the value of  $b_1$  until (a) the Friedel sum rule is satisfied and (b) the values of  $n_s(0)$ ,  $\delta_0(v_F)$  and  $\delta_1(v_F)$  are in close agreement with the results of DFT.

The values of the parameters obtained from this adjustment are contained in tables 1 and 2, for H and He, respectively. The electron gas density  $n_e$  is here represented

**Table 1.** Result for hydrogen.

$r_s$	$b_1 = b_2$	$b_3$	$a_1$	$a_3$	$n_s(0)$	$n_s(0)^{\text{DFT}}$	$\delta_0$	$\delta_0^{\text{DFT}}$	$\delta_1$	$\delta_1^{\text{DFT}}$	$Q$	$Q^{\text{DFT}}$
5.5	1.064	0.801	0.790	-6.032	0.337	0.336	1.811	1.812	-0.080	-0.097	0.047	0.049
5	1.071	0.822	0.857	-6.012	0.343	0.343	1.753	1.756	-0.061	-0.074	0.059	0.059
4	1.074	0.836	0.863	-6.288	0.360	0.360	1.598	1.599	-0.009	-0.017	0.098	0.098
3	1.072	0.826	0.744	-7.044	0.395	0.395	1.329	1.369	0.080	0.053	0.163	0.163
2	2.076	5.437	1.358	-0.388	0.494	0.495	1.049	1.049	0.138	0.124	0.255	0.258
1.5	2.230	5.371	1.466	-0.460	0.644	0.629	0.857	0.852	0.162	0.150	0.310	0.310

**Table 2.** Result for helium.

$r_s$	$b_1 = b_2$	$b_3$	$a_1$	$a_3$	$n_s(0)$	$n_s(0)^{\text{DFT}}$	$\delta_0$	$\delta_0^{\text{DFT}}$	$\delta_1$	$\delta_1^{\text{DFT}}$	$Q$	$Q^{\text{DFT}}$
5.5	2.378	2.351	2.108	-4.644	3.554	3.556	2.778	2.787	0.114	0.120	0.012	0.012
5	2.583	2.865	2.466	-3.473	3.560	3.562	2.724	2.735	0.129	0.133	0.019	0.019
4	2.703	3.605	2.840	-2.556	3.580	3.581	2.597	2.592	0.171	0.168	0.047	0.047
3	2.854	4.475	2.333	-1.920	3.627	3.627	2.391	2.365	0.228	0.217	0.135	0.135
2	3.076	6.905	2.258	-1.275	3.791	3.781	2.082	1.982	0.295	0.279	0.427	0.427
1.5	3.086	4.634	2.352	-1.453	4.038	4.032	1.634	1.689	0.350	0.309	0.754	0.754

by the usual  $r_s$  parameter, being  $(4\pi/3)r_s^3 n_e = 1$ . We also include in these tables the values of the friction coefficient  $Q$  and of the screening charge density on the impurity  $n_s(0)$ , and compare these values with those obtained from the DFT calculations<sup>1</sup>. As may be observed in these tables, the results show excellent agreement with those obtained from the DFT, both for hydrogen and helium, and for the range of  $r_s$  values of interest for metallic elements. We note that the negative values of  $a_3$  indicate the need for an *antiscreening* term (to compensate for some overscreening produced by the two other terms in equation (9)).

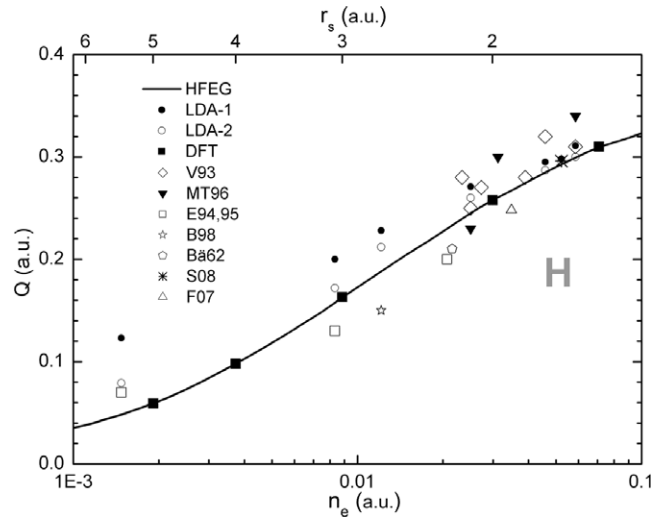
Before comparing the values of the stopping coefficient with experimental results, one should give some consideration to the fact that the model of a uniform electron gas applied before may not be quite realistic to simulate the conditions in solid targets. Previous authors [13, 14] have considered the use of the local density approximation (LDA), which is based on the ansatz that a nonuniform electron distribution may be modeled using the results obtained for a uniform electron gas followed by an appropriate average with the real electron density inside the solid [15, 16]. Following this line of approach we have performed two types of atomic averaging. The first one considers the full average of the stopping coefficient on the atomic cell in the solid, given by

$$Q_{\text{av1}} = \frac{1}{V_0} \int_0^{r_{\text{max}}} Q[r] 4\pi r^2 dr, \quad (17)$$

where  $r_{\text{max}}$  is the radius of one atomic cell in the solid and  $V_0 = 4\pi r_{\text{max}}^3/3$  is its corresponding atomic volume. The quantity  $Q[r]$  here corresponds to the value of the stopping coefficient calculated as in equation (7) for the local value of the electron density in the cell  $n_e(r)$ . To perform this calculation we have used previous calculations of electron densities from [17, 18].

One of the shortcomings of the previous approach is the fact that it includes in the integration all the atomic electrons within a free-electron gas approximation, without

<sup>1</sup> E Zaremba (Queen's University, Canada) supplied the computational code for the DFT calculations showed in the tables and in the graphs.



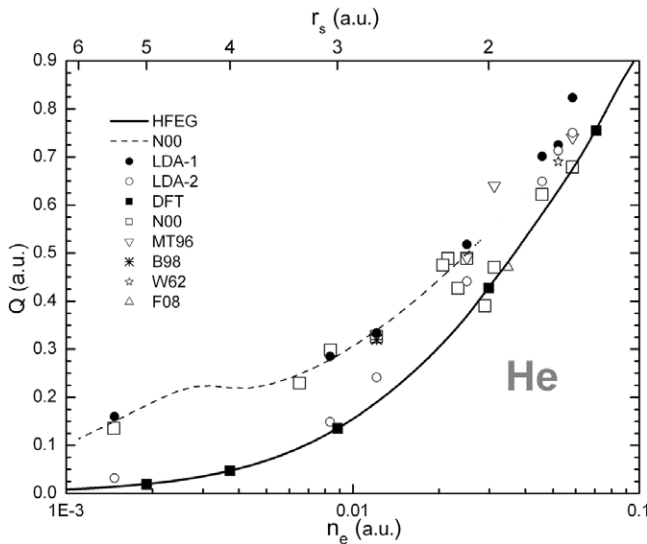
**Figure 1.** The coefficient  $Q$ , for protons, as a function of the density of the electron gas  $n_e$  (with  $r_s$  in the upper scale). The solid line shows the present theoretical result for a homogeneous FEG system. The calculations of  $Q_{\text{av1}}$  and  $Q_{\text{av2}}$  are indicated by solid (LDA-1) and open (LDA-2) circles, respectively. The filled squares show the DFT values<sup>1</sup>. The other symbols show the experimental values obtained at low velocities.

consideration of the important binding effects that will tend to cancel the contribution of inner shells to the stopping in the case of slow ions. To quantify this effect in a simplified but effective way, we restrict the integration to the outer region of the electronic cloud, in the form

$$Q_{\text{av2}} = \frac{1}{V_c} \int_{r_c}^{r_{\text{max}}} Q[r] 4\pi r^2 dr, \quad (18)$$

where  $r_c$  is numerically determined by the condition that the number of outer electrons (in the range  $r_c < r < r_{\text{max}}$ ) agrees with the number of valence or conduction electrons per atom in the solid.

The comparison of these calculations with experimental results of the stopping coefficients is illustrated in figures 1 and 2, for H and He, respectively. The calculated stopping



**Figure 2.** The coefficient  $Q$ , for He ions, as a function of the density of the electron gas  $n_e$  (with  $r_s$  in the upper scale). The solid line shows the present theoretical result for a homogeneous FEG system. The calculations of  $Q_{av1}$  and  $Q_{av2}$  are indicated by solid (LDA-1) and open (LDA-2) circles, respectively. The filled squares show the DFT values<sup>1</sup>. The other symbols show the experimental values obtained at low velocities. The dashed line shows the theoretical result of [26].

values are represented here as a function of the FEG density  $n_e$  (lower scale) and the equivalent  $r_s$  parameter (upper scale). The solid lines show the values of  $Q$  for a homogeneous free-electron gas (HFEG) of density  $n_e$  obtained with the present theoretical approach, equation (7). In these figures we show the experimental results for  $H^+$  [19–25] in Rb, Ca, Mg, Pb, Sn, Bi, Al, Sb, Zn, Cu, Ag, C and Au and for  $He^+$  [26, 20, 27, 24] in Rb, Sr, Ca, Mg, Pb, Sn, Bi, Al, Ge, Zn, Ag, C and Au in increasing order of FEG density. We also show here the experimental stopping value for channeling in Au from [28, 29].

To represent the experimental results on the FEG density scale here, we have considered for each solid its experimental plasmon frequency  $\omega_p$  and assigned the corresponding position on this plot using the effective density that corresponds to the plasmon frequency measurements [30]<sup>2</sup>, namely  $n = \omega_p^2/4\pi$ . In the case of channeling we use the mean value of the density in the center of the channel. The calculations of  $Q_{av}$  according to equations (17) and (18) for Rb, Ca, Mg, Al, C, Ag and Au are indicated by solid and open circles, respectively.

The results obtained from the present approach (solid lines) coincide almost exactly with those of the DFT obtained from<sup>1</sup>. We note that, according to [5], the value for  $Z = 1$ ,  $r_s = 2$ , was not obtained in a self-consistent way but was interpolated from other  $r_s$ . For this reason the DFT values included in tables 1 and 2 are those obtained from Zaremba's code<sup>1</sup>. It should be mentioned also that the results obtained with this program for all other  $r_s$  values were in close agreement with those in [5]. As may be observed, the lines corresponding to this calculation (and also to the DFT values)

<sup>2</sup> In the case of Ag and Au the  $r_s$  values were calculated by an average in the same way as for  $Q_{av2}$ .

describe the general behavior of the experimental data in the region of medium and high densities, where the lines goes through the cloud of points. For hydrogen, the calculated stopping coefficient  $Q$  is in relatively good agreement with the experiments, while for helium the numerical agreement is poor and only the general trend of the data is reproduced. For the range of densities larger than  $\sim 0.03$  (a.u.) the calculated curve follows the experimental tendency and both are in fair agreement, although the individual deviations of the experimental points are significant. For densities lower than  $\sim 0.03$  (a.u.) the calculated values are too small compared with the experiments, and we find a large disagreement at low densities, particularly in the case of Rb (with  $r_s = 5.45$  and  $n_e = 0.00147$ ). A more specific quantitative study of differences between DFT and experiments for H and He in various metals was made in [20].

To estimate the possible effects produced by the density inhomogeneities in real solids we made use of the local density approximation (LDA) and performed the calculations indicated by equations (17) (unrestricted, LDA-1) and (18) (restricted, LDA-2). The corresponding results are indicated by solid and open circles in these figures. The quality of the improvement obtained by this method is, however, not the same in all cases; for instance, in the case of Rb we find a very good agreement using LDA-2 for hydrogen, but a better agreement using LDA-1 for helium. In addition to these calculations we have included in figure 2 by a dashed line, for the range of high  $r_s$ , the calculation of [26] using a phase shift sum rule for the amplitude of the dipolar backflow current. The agreement with the experimental values of that [26] in this case is good. However, we found that if the same approach is used for hydrogen the results show a large disagreement with the experiments. Therefore, we consider that there is a more basic problem still unsolved in the description of the interaction between external impurities and an FEG in the range of large  $r_s$ . This indicates also a point of interest for further investigation concerning the description of inhomogeneous electron gas interactions in high- $r_s$  metals.

The comparison with experiments is not satisfactory in quantitative terms, although the general behavior is qualitatively represented. We point out the need for additional experimental and theoretical research of the observed differences, particularly in the case of metals with high  $r_s$  values, where the discrepancy between the previously mentioned sum rules should be investigated. In summary, we have studied a simple formulation of the screening of light ions in a free-electron gas, using a hydrogenic type of density function with adjustable parameters. The parameters are adjusted to satisfy the Friedel sum rule and the Kato cusp condition. We find a convenient framework that describes with excellent accuracy the values of the density at the origin, the stopping coefficient and the main phase shift values, as compared with density functional calculations.

## Acknowledgments

We are very grateful to E Zaremba (Queen's University, Canada) for providing a computer program to perform the



DFT calculations. This work was partially supported by ANPCYT/Argentina (project PICT 0903/07).

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