Constrained local plasma density approximation for the calculation of the stopping power for slow ions in solids

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A constrained local plasma density approximation is presented for the calculation of the stopping power of solids for slow ions. Using the density-functional theory with the local density approximation for exchange-correlation in the scattering potential and the constrained local plasma density approximation for averaging over effective solid-state electron densities, the stopping power for slow hydrogen and helium projectiles in different solids are calculated. The obtained results of the stopping powers for slow H and He projectiles, and the stopping power ratio \( R = \frac{\langle -dE/dx \rangle_{\text{He}}}{\langle -dE/dx \rangle_{\text{H}}} \) in some solids are in agreement with recent experimental predictions. [S0163-1829(98)02330-3]

I. INTRODUCTION

The problem of energy losses suffered by charged particles moving in matter is of continuous interest in physics. When the projectile velocity is much greater than the average velocity of valence electrons in targets, a good description of energy losses of ions can be obtained by using the linear response theory, in which the screened potential is treated to the lowest order, and the obtained energy loss is proportional to the square of the ion charge. However, at low velocities, the importance of screening nonlinearities was demonstrated by using a scattering theory approach to the stopping power and density-functional theory (DFT). The scattering theory naturally results in the \( Z_1 \) oscillation (\( Z_1 \) is the ion charge) of the transport cross section and, for screened potentials of DFT, provides close agreement with experimental stopping powers, especially for the case of channeling ions.

When an ion traverses through a solid in random incidence conditions, it encounters the space-varying density of electrons. Therefore, a detailed stopping-power calculation should involve averaging over the realistic distribution of the electron density encountered by the projectile in its trajectory through the solid.

Recently, the local plasma density approximation (LPDA) (Ref. 5,6) was applied to nonlinear calculations of stopping powers of solids for slow ions. In Ref. 7 the authors used a Yukawa-type screened potential to calculate the scattering cross section, and the relevant screening parameter in the screened potential was adjusted in a consistent way by satisfying the Friedel sum rule. Using the LPDA, their nonlinear calculation replicated well the experimental \( Z_1 \) oscillations of the stopping power.

In Ref. 8 the LPDA was combined with self-consistent density-functional theory, and the obtained stopping powers of realistic solids for slow H and He projectiles are in agreement with experimental predictions. For slow H and He projectiles in the solids with \( r_s \leq 2 \), the contribution of core electrons to the stopping power was found to be relatively small as compared with that of valence electrons.

The original LPDA is defined in terms of the total electron density. In this paper, we present a constrained LPDA for calculations of the stopping power of solids for slow ions \( (v \approx v_F) \), where \( v \) is the projectile velocity and \( v_F \) is the Fermi velocity of an electron gas, in which an effective electron density from the core of target atom is taken into account in the usual, volume-averaging procedure (see Sec. II C). The purpose of this work is to investigate the contribution of weakly bound core electrons to the stopping power of the solids with \( r_s \) covering a wide range, \( 2 \leq r_s \leq 5.5 \), using a concept of LPDA for the nonlinear density-functional calculation of stopping power. Hartree atomic units, i.e., \( \hbar = m = e = 1 \), will be used throughout this paper.

II. FORMALISM

A. Nonlinear stopping power of an electron gas

Considering an ion of charge \( Z_1 \) moving slowly in a homogeneous electron gas with density \( n_0 \), the energy loss of the ion traversing a path length \( dx \) can be written as

\[
\frac{dE}{dx} = n_0 v_F^2 \sigma_a(v_F),
\]

where \( v_F = (3\pi^2 n_0)^{1/3} \), and \( \sigma_a \) is the momentum-transfer (transport) cross section. For a spherically symmetric scattering potential \( \sigma_a(v_F) \) can be expressed as
TABLE I. Parameters used in the calculation.

<table>
<thead>
<tr>
<th>Solid</th>
<th>Z_a</th>
<th>r_s</th>
<th>N_a</th>
<th>c_i</th>
<th>R</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mg</td>
<td>12</td>
<td>2.70</td>
<td>6.38 \times 10^{-3}</td>
<td>3.93 \times 10^{-3}</td>
<td>2.61</td>
</tr>
<tr>
<td>Al</td>
<td>13</td>
<td>2.12</td>
<td>8.93 \times 10^{-3}</td>
<td>7.14 \times 10^{-3}</td>
<td>2.29</td>
</tr>
<tr>
<td>Ca</td>
<td>20</td>
<td>3.06</td>
<td>3.43 \times 10^{-3}</td>
<td>6.95 \times 10^{-4}</td>
<td>2.73</td>
</tr>
<tr>
<td>Rb</td>
<td>37</td>
<td>5.45</td>
<td>1.60 \times 10^{-3}</td>
<td>8.10 \times 10^{-4}</td>
<td>5.10</td>
</tr>
<tr>
<td>Sr</td>
<td>38</td>
<td>3.32</td>
<td>2.65 \times 10^{-3}</td>
<td>1.08 \times 10^{-3}</td>
<td>3.19</td>
</tr>
</tbody>
</table>

*a* Values of \( r_s \) for all solids are from Ref. 15

*b* Quoted from Ref. 14

\[
\sigma_n(v_F) = \frac{4 \pi}{v_F^2} \sum_{l=0}^{\infty} (l+1) \sin^2 \left[ \delta_l(v_F) - \delta_{l+1}(v_F) \right],
\]

(2)

where \( \delta_l(v_F) \) is the \( l \)th partial-wave phase shift at the Fermi level. The scattering potential is determined self-consistently by using the DFT with a local density approximation (LDA) for the exchange-correlation potential.\(^1\)

B. Electron density in solids

The electron density in solids differs from that of a free atom due to the overlap of electron wave functions of neighbouring atoms. The LCAO calculations have shown\(^2\) that the valence-shell electrons appear to be delocalized. The density of the such electrons is nearly constant throughout most of the solid volume, so the valence electrons may be regarded as a uniform electron gas. On the other hand, the core electron wave functions are similar to those of a free atom. We make use of a simple model proposed by Gertner et al.\(^5\) which allows for the abovementioned features. The electron density of an atom in a solid is assumed to be constant \( n_0 \) in the outer region \((r \gg R)\) and slightly changed in the inner region \((r \ll R)\) compared with the electron density \( n^A(r) \) of the free atom. The total electron density has the following form:\(^6\)

\[
n(r) = \begin{cases} 
n^A(r) + c_i & \text{if } R \ll r \ll R_0, \\
n_0 & \text{if } R_0 < r < R.
\end{cases}
\]

(3)

where \( c_i \) is a constant density correction and \( R_0 \) is the radius of an atom in solid, which is determined from \( \frac{2}{3} \pi R_0^3 = N \) \((N\) is the number of atoms per unit volume in the target quoted from Ref. 14). The density \( n_0 \) of valence electrons is derived from the experimental measurement of the plasma frequency \( \omega_p = 4 \pi n_0 \)\(^1/2\), and the relevant \( r_1 \) is listed in Table I. The constants \( c_i \) and \( R \) are fixed by the following coupled equations:

\[
4 \pi \int_0^R r^2 n^A(r) dr + \frac{4}{3} \pi R^3 c_i + \frac{4}{3} \pi (R_0^3 - R^3) n_0 = Z_2,
\]

(4)

\[
n_0 = c_i + n^A(R),
\]

(5)

where \( Z_2 \) is the atomic number for the target. The electron density of a free atom \( n^A(r) \) is quoted from Ref. 16. By solving Eqs. (4) and (5), the values of constants \( c_i \) and \( R \) for some solids under study are obtained and listed in Table I.

C. A constrained local plasma density approximation

As we know, the energy gaps from deep shells (strongly bound by target nucleus) in the core of target atom to the lowest unoccupied level are quite large, and therefore the electrons in those deep shells are difficult to be excited by a slow ion \((v \ll v_F)\). As an approximation, we freeze the excitation of deep core shells of target atom, and introduce an effective electron density \( n_{\text{eff}}(r) \) which is defined as

\[
n_{\text{eff}}(r) = \begin{cases} 
\sum_{n \epsilon_{n,l}^A, \epsilon_{n,l}^i} n_{n,l}^i(r) & \text{if } R_0 < r < R, \\
n_0 & \text{if } R \ll r < R_0, 
\end{cases}
\]

(6)

where \( n_{n,l}^i(r) \) is the electron density of the \((n,l)\) shell of a free atom \((n\) and \(l\) are principal quantum number and orbital angular momentum number, respectively), and related to the total electron density \( n^A(r) \) of the free atom by

\[
n^A(r) = \sum_{n \epsilon_{n,l}^A, \epsilon_{n,l}^i} n_{n,l}^i(r),
\]

(7)

\( \epsilon_{n,l}^i \) is the one-electron energy in the \((n,l)\) shell and the subscripts \( n_0 \) denote the lowest excitable shell.

For simplicity, we choose the effective core shells in the following way. If in the ground state the principal quantum number of valence electrons is \( n \), the shells (in the core of target atom) with the principal quantum number equal to \((n - 1)\) are included (un) in the calculation of the stopping power for slow H and He projectiles, and all other core shells with smaller principal quantum number are excluded. For the metals under study, the relevant shells, which are included in the stopping-power calculation, are listed in Table II. Then, the effective electron density \( n_{\text{eff}}(r) \) can be easily obtained from Eq. (6).

By using the concept of LPDA\(^5\), we extend the theoretical framework for a homogeneous electron gas to the inhomogeneous electron gas specified by \( n_{\text{eff}}(r) \). Finally, we obtain the average stopping power

\[
- \frac{dE}{dx} = N \int_0^{R_0} 4 \pi r^2 dr \left[ - \frac{dE}{dx} [n_{\text{eff}}(r)] \right],
\]

(8)

where \(- (dE/dx)[n_{\text{eff}}(r)] \) is calculated by using Eq. (1) for the electron density equal to \( n_{\text{eff}}(r) \). Since now the stopping power is calculated for the effective electron density \( n_{\text{eff}}(r) \) defined in Eq. (6) instead of the total electron density, we call this calculation the constrained local plasma density approximation (CLPDA).


TABLE II. Lowest shell \((n_0,l_0)\) and all relevant shells for effective electron density \( n_{\text{eff}}(r) \).

<table>
<thead>
<tr>
<th>Solid</th>
<th>( n_0 )</th>
<th>( l_0 )</th>
<th>( n_{\text{eff}}(r) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mg</td>
<td>12</td>
<td>2s</td>
<td>[2s^2,2p^6,3s^2]</td>
</tr>
<tr>
<td>Al</td>
<td>13</td>
<td>2s</td>
<td>[2s^2,2p^6,3s^2]</td>
</tr>
<tr>
<td>Ca</td>
<td>20</td>
<td>3s</td>
<td>[3s^2,3p^6,4s^2]</td>
</tr>
<tr>
<td>Rb</td>
<td>37</td>
<td>4s</td>
<td>[4s^2,4p^6,5s^2]</td>
</tr>
<tr>
<td>Sr</td>
<td>38</td>
<td>4s</td>
<td>[4s^2,4p^6,5s^2]</td>
</tr>
</tbody>
</table>
III. RESULTS AND DISCUSSIONS

Using the CLPDA described in Sec. II, the nonlinear calculation of the stopping powers for slow H and He projectiles \((v \ll v_f)\) in Al, Mg, Ca, Sr, and Rb has been performed, and the obtained results are shown (by scattered crosses) in Figs. 1 and 2. The nonlinear stopping power of a homogeneous electron gas for slow H and He projectiles are exhibited in Figs. 1 and 2, respectively, as a solid curve. The corresponding experimental results \(^9–11\) are also displayed in those two figures by scattered solid triangles. From Figs. 1 and 2, one can find that our calculation results of stopping powers for He in the metals under study using the CLPDA are in good agreement with the experimental data, \(^9–11\) while those for H are slightly higher than the experimental predictions. \(^9–11\)

The stopping-power ratio \(R\), which is defined as

\[
R = \frac{\left[-\frac{dE}{dx}\right]_{\text{He}}}{4\left[-\frac{dE}{dx}\right]_{\text{H}}}
\]

for the metals under study, is also calculated by using the CLPDA and shown in Fig. 3 as scattered crosses. For comparison, the experimental predictions \(^9\) for the stopping-power ratio in those metals are also given in Fig. 3 by solid triangles. Correspondingly, in Fig. 3 the calculation result for a homogeneous electron gas is exhibited as a solid curve.

It is of interest to compare the volume-averaged stopping power of a solid obtained using the CLPDA with that obtained by using the original LPDA (OLPDA). For convenience, let us define the relative difference \(\eta\) between the results of those two calculations

\[
\eta = \frac{(-dE/dx)_{\text{OLPDA}} - (-dE/dx)_{\text{CLPDA}}}{(-dE/dx)_{\text{CLPDA}}},
\]

where \((-dE/dx)_{\text{OLPDA}}\) and \((-dE/dx)_{\text{CLPDA}}\) are the volume-averaged stopping powers obtained using the OLPDA and CLPDA, respectively. For H and He projectiles moving slowly in Mg and Al, the relative difference of the OLPDA and CLPDA calculations \(\eta\) is found to be less than 0.1%. For H in Ca, Sr, and Rb the values of the relative difference \(\eta\) are 0.3, 0.8, and 1.0 %, respectively. For He in Ca, Sr, and Rb the values of \(\eta\) increases slightly to 1.3, 4.0, and 5.4 %, respectively.

In summary, in this paper we presented a constrained local plasma density approximation for the calculation of the stopping power for slow ions \((v \ll v_f)\) in solids. Using the density-functional theory with the local density approximation for exchange-correlation in the scattering potential and the constrained local plasma density approximation for
volume-averaging, the stopping powers for hydrogen and helium projectiles moving at low velocities \((v \approx v_f)\) in Mg, Al, Ca, Rb, and Sr have been calculated, and the obtained results of the stopping powers as well as the calculated stopping-power ratio \(R\) are in agreement with experimental predictions.9–11 A comparison of the original LPDA and constrained LPDA calculations shows that the contribution of very strongly bound core electrons of a solid atom to the stopping power for slow H and He projectiles is very small.

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9 On leave from Fudan University, Shanghai, P. R. China.
11 D. Semrad (private communication).