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ENERGY LOSS OF EXCITED SLOW IONS IN ELECTRON GAS

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The electronic energy loss and the straggling of the energy loss of the degenerate electron gas for excited H*-, He*-, He**-, and Li*-like ions were calculated. The results were compared with the corresponding characteristics for ions kept in the ground state. The linear response theory was used. The ion was described by the Hartree–Fock–Slater formalism and the medium by the dielectric function. The stopping and straggling effective charges Z_{ef} for the energy loss were analysed and they were found to differ from each other and to depend on the one-electron radius r_s, on the ion atomic number Z_i, and on the number of electrons N_i carried by the ion.

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1. Introduction

When an ion approaches and crosses the surface of a solid and then penetrates the bulk it exchanges electrons with the solid forming intermediate electronic configurations. It experiences complicated excitation–deexcitation transitions. For a short time it remains excited. The configuration alters the electronic energy loss and the straggling of the energy loss for the ion beam. This is particularly important at low ion velocity v. The basic papers in this field [1, 2] were related to the analysis of the stopping and straggling of an atomic nucleus as an ion and thus neglecting the effect of its electronic configuration. The results of calculation for structured projectiles at high [3–6] and low [7–9] ion velocities, and also experimental results for low velocity heavy ions [10] were published. To the author's knowledge, neither theoretical nor experimental data on the energy loss experienced by low velocity excited ions are available in the literature.

The most characteristic output of theory and experiment at low ion velocity v is proportionality of the energy loss to v and its straggling to v^2. It origins from the proportionality of the response function to the energy transfer from the low velocity ion to the medium, $\epsilon^{-1}(k,\omega) \propto \omega$. The target and the ion features, hidden

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in the proportionality factor, are dependent on the model used for calculations. The free electron gas (at $T = 0$ K) characterised by the one-electron radius r_s ($n = 3/4\pi r_s^3 a_0^3$ is the electron gas density) is considered as a target. The ion of interest consists of an atomic nucleus of atomic number Z_i, moving slowly with the velocity v, and carrying $N_i \leq 4$ electrons.

In this paper m, e, a_0, and v_0 are the electron rest mass, the elementary charge, the Bohr radius, and the Bohr velocity, respectively. Atomic units are used throughout, unless otherwise indicated.

2. Calculation procedure

Within the random phase approximation (RPA) the probability for the transfer of the energy ω and the momentum k from an ion to a degenerate free electron gas is described by the equilibrium dielectric function $\epsilon(u,z) = 1 + (\chi^2/z^2)[f_1(u,z) + \mathrm{i}f_2(u,z)]$, where the parameters are defined as $z = k/(2k_F)$, $u = \omega/(kv_F)$, $\chi^2 = r_s/(\pi\alpha)$. The Fermi wave vector is $k_F = \alpha/(a_0 r_s)$ and $\alpha = (9\pi/4)^{1/3}$. The electronic energy loss per unit path length x, $\mathrm{d}E/\mathrm{d}x$, and the straggling of the energy loss parameter Ω^2 (per electron) are given by [1]

$$\frac{\mathrm{d}E}{\mathrm{d}x} = \frac{4}{3\pi}\frac{v}{v_0}Z_1^2, \qquad \frac{\Omega^2}{x} = \frac{9}{r_s^3}\left(\frac{v}{v_0}\right)^2 Z_2^2, \tag{1}$$

where the Z_m^2 functions are defined as

$$Z_m^2 = \int_0^1 \mathrm{d}z z^{m+2} \frac{Z^2(z)}{[z^2 + \chi^2 f_1(0,z)]^2}. \tag{2}$$

For $f_1(0,z)$, the expressions derived for the Fermi momentum distribution [1] can be used

$$f_1(0,z) = \frac{1}{2} + \frac{1-z^2}{4z}\ln\frac{z+1}{z-1}. \tag{3}$$

The form factor $Z^2(z)$ is the Fourier transform of the spatial electron distribution on the ion [6] being a sum of the screening component Z_s^2 reducing distant collisions, and the anti-screening component Z_a^2, strengthening close collisions

$$Z^2(z) = Z_s^2(z) + Z_a^2(z) = [Z_i^2 - \rho(z)N_i]^2 + N_i[1 - \{\rho(z)\}^2], \tag{4}$$

where $\rho(z)$ is the one-electron form factor.

The conduction electrons of a solid screen the quasi-static electric potential of a slow ion due to dielectric response. Provided the speed of the ion is lower than the Fermi velocity $v_F = v_0\alpha/r_s$, this screening can be approximately described in terms of the screening function $\exp(-rk_{TF})$, where the Thomas–Fermi wave number k_{TF} is related to the Fermi wave number as $k_{TF}^2 = 4k_F/(\pi a_0)$.

For ions carrying $N_i \leq 4$ electrons we apply the Hartree–Fock–Slater (HFS) description. The total self-consistent Hamiltonian is given by

$$H = -\frac{1}{2}\sum_j \Delta_j + \sum_j V_{ne}(r_j) + \frac{1}{2}\sum_{j\neq k} V_{ee}(|r_j - r_k|). \tag{5}$$

V_{ee} and V_{ne} are the exponentially screened interactions between two electrons and between an electron and a nucleus, respectively. Physically, this phenomenological Hamiltonian gives an account of an approximate, long wavelength screening exerted by the electron gas instead of the response provided by the full dielectric function $\epsilon(k,\omega)$.

Approximate eigenfuctions of the Hamiltonian Eq. (5), in the form of HFS determinants, are built from $1s$ and $2s$ one-electron trial functions

$$\psi_{1s}(r) = \pi^{-1/2}(\lambda)^{-3/2}\exp(-r/\lambda),$$

$$\psi_{2s}(r) = \pi^{-1/2}(2\lambda)^{-3/2}(1 - r/2\lambda)\exp(-r/2\lambda). \tag{6}$$

We call $\lambda = a_0/Z$ the size parameter of the electron distribution. This parameter is modified when the ion enters a solid. We determine $Z = Z_{\min}$ from the variational condition of minimum for the expectation values of the total Hamiltonian $\overline{H}$. For different $1s2s$ configurations of excited (Ion*) ions and ions in the ground state (Ion) we have

$$\overline{H}(\mathrm{H} \equiv 1s^1) = E_{1s},$$

$$\overline{H}(\mathrm{H}^* \equiv 1s^0 2s^1) = E_{2s},$$

$$\overline{H}(\mathrm{He} \equiv 1s^2) = 2E_{1s} + V_{1s1s},$$

$$\overline{H}(\mathrm{He}^* \equiv 1s^1 2s^1) = E_{1s} + E_{2s} + V_{1s2s} - A_{1s2s},$$

$$\overline{H}(\mathrm{He}^{**} \equiv 1s^0 2s^2) = 2E_{2s} + V_{2s2s}, \tag{7}$$

$$\overline{H}(\mathrm{Li} \equiv 1s^2 2s^1) = 2E_{1s} + E_{2s} + V_{1s1s} + 2V_{1s2s} - A_{1s2s},$$

$$\overline{H}(\mathrm{Li}^* \equiv 1s^1 2s^2) = E_{1s} + 2E_{2s} + V_{2s2s} + 2V_{1s2s} - A_{1s2s},$$

$$\overline{H}(\mathrm{Be} \equiv 1s^2 2s^2) = 2E_{1s} + 2E_{2s} + V_{1s1s} + V_{2s2s} + 4V_{1s2s} - 2A_{1s2s}.$$

The energy (E), the direct Coulomb (V), and the exchange (A) integrals were calculated analytically and are given in Appendix.

Taking a Fourier transform of the electron density built from the eigen functions of Eqs. (6) we have

$$N_i\rho(z) = N_{1s}\rho_{1s}(2\phi) + N_{2s}\rho_{2s}(2\phi)$$

$$= N_{1s}\frac{1}{(1+\phi^2)^2} + N_{2s}\frac{1 - 3(2\phi)^2 + 2(2\phi)^4}{[1+(2\phi)^2]^4}, \tag{8}$$

where $\phi = k_F\lambda z$, N_a is the number of electrons in a state.

3. Results and discussion

When an ion with a few electrons is in the vacuum (only atomic screening is effective) then $k_{TF} = 0$, and then the $\overline{H}$'s of Eq. (7) reach minima (calculated as $\partial\overline{H}/\partial Z$) at the values $Z_{\min}$ presented in Table for excited ions and these in

TABLE

$Z_{\min}$ and $\overline{H}$ for H- to Be-like ions in vacuum ($k_{TF} = 0$).

ion-like	N_i	$Z_{\min} - Z_i$	$\overline{H}/Z_{\min}^2$
H	1	0	−1/2
H*	1	0	−1/8
He	2	−0.3125	−1
He*	2	−0.15034	−5/8
He**	2	−0.30078	−1/4
Li	3	−0.45458	−9/8
Li*	3	−0.36546	−3/4
Be	4	−0.6284	−5/4

the ground state. In this model the energy required for excitation or for electron detachment can be calculated as a difference of adequate $\overline{H}$'s. For instance, the calculated total energy for He atom, $\overline{H}(\mathrm{He}(1s^2 2s^0)) = -77.45$ eV is a bit worse than the reference value of −77.83 eV [11]. The excitation $\mathrm{He}(1s^2 2s^0) \rightarrow \mathrm{He}(1s^1 2s^1)$ demands 19.29 eV, and detachment of electron $\mathrm{He}(1s^2 2s^0) \rightarrow \mathrm{He}(1s^1 2s^0) + e^-$ demands 23.06 eV, whereas the He atomic energy level taken from a photoelectron measurement is 24.58 eV [12].

In matter these parameters are modified due to the interaction with electron gas, but an ion exists as long as its total energy remains negative. In this case $Z_{\min}$ was calculated by taking a numerical minimum of the appropriate $\overline{H}$ from Eq. (7), so $Z_{\min}$ depends on Z_i, N_i, and additionally on r_s. Subsequently, $Z_{\min}$ was used in Eqs. (8, 4). As r_s decreases, for a given Z_i, $Z_{\min}$ increases causing a decrease in the total projectile energy, which means a stronger screening of the ion nucleus interaction by the medium. This screening is also much more important at low Z_i. For instance, in the electron gas of $r_s = 1$, $\overline{H}(\mathrm{He}(1s^2 2s^0)) = -2.48$ eV. For excitation 1.47 eV is required and for electron detachment 2.23 eV. For dense electron gas, $r_s \rightarrow 0$, the minima of energy are reached at $Z_{\min} = 0$ in consequence of the model with exponential screening. For a given Z_i a minimum of the total energy is reached at $N_i = Z_i$, whereas minima at other N_i's are local and reached by deformation of the charge distribution shown in $Z_{\min}$. For instance, we can have a metastable configuration of a proton even with four electrons. From Table the minimization of $\overline{H}$ for $\mathrm{Be}(1s^2 2s^2)$ in the case of $Z_i = 1$ gives a minimum equal to −4.7 eV for $Z_{\min} = 0.3716$.

As an example of behaviour of an ion in electron gas, the dependence of $Z_{\min}$ and $\overline{H}$ upon Z_i and r_s for excited $\mathrm{Li}^*(1s^1 2s^2)$ atom and for the same atom in the ground state $\mathrm{Li}(1s^2 2s^1)$ are drawn in Fig. 1. It should be noted that the denser the gas is, the smaller $Z_{\min}$, and the flatter the electron distribution becomes. Below $r_s = 0.6$ neither Li* nor Li can create a bound state in electron gas. Moreover, $Z_{\min}$ is much smaller for an exited ion than for an ion in the ground state.

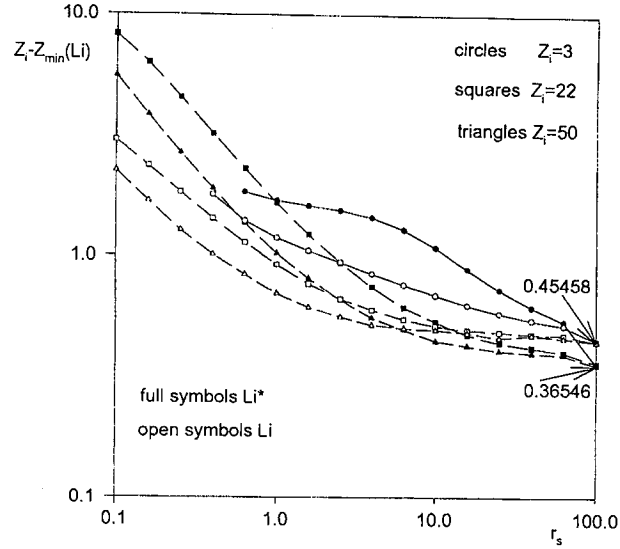

Fig. 1. $Z_{\min}$ calculated numerically as a function of (r_s, Z_i) for Li-ion electronic configuration. For $r_s = 100$ the asymptotic values are shown. In the whole (r_s, Z_i) region $Z_{\min}(\mathrm{Li}^*) < Z_{\min}(\mathrm{Li})$. Li atoms cannot be stable below $r_s = 0.8$.

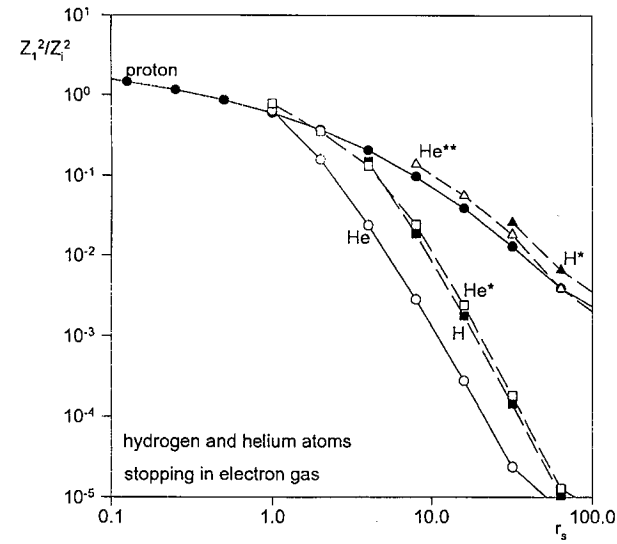

Fig. 2. Reduced energy loss function Z_1^2/Z_i^2 for H and He atoms. To get energy loss in eV/Å multiply by $21.82 Z_i^2 v$.

In Fig. 2 we compared the reduced energy loss Z_1^2/Z_i^2 for hydrogen and helium atoms in electron gas as functions of r_s. We get the energy loss per unit path length (in eV/Å) by multiplying the vertical scale by $21.82 Z_i^2 v$. Furthermore, the atoms cannot exist in solids where $r_s < 1$. In the region of metallic density H

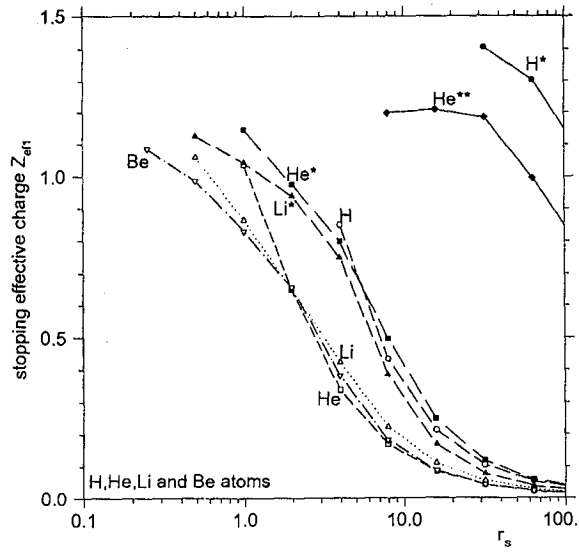

Fig. 3. The stopping effective charge for H to Be atoms (including excited).

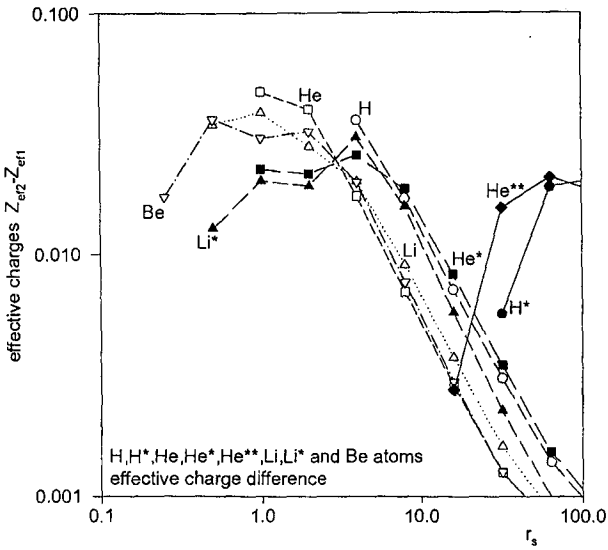

Fig. 4. The straggling effective charge Z_{ef2} calculated for H to Be atoms is generally larger than the stopping effective charge Z_{ef1}. For $r_s > 2$ this difference is larger for excited ions than for ions in the ground state.

and He* lose energy in accordance with proton, whereas for dilute gas their lossof energy decreases by several orders of magnitude when compared to proton. It is interesting to note that H* and He** lose energy like proton in the r_s region in which they can exist. The energy loss straggling reveals the same behaviour.

Usually, for the energy loss analysis effective charge is defined [6, 9], relating the stopping and straggling produced by a given ion to the same quantities by the

ion atomic nucleus. For ions with N_i electrons, the effective charge for the energy loss Z_{ef1} and for the straggling of energy loss Z_{ef2} is $Z_{efm} = \sqrt{Z_m^2(N_i)/Z_m^2(0)}$, where Z_m^2 is given by Eq. (2). For a point charge $Z_{efm} = 1$, but for an ion in solid it depends on Z_i, N_i, and r_s. In Fig. 3 the effective charge for stopping Z_{ef1} for all the atoms with ($N_i \leq 4$) is displayed. We can distinguish three groups: atoms in the ground state losing the energy slowly, atoms singly excited (and proton), and atoms doubly excited (and excited hydrogen). These groups correspond to the size of projectiles. Particularly, in the case of He atoms, at nearly metallic density $r_s = 8$, the stopping effective charges differ from each other by the order of magnitude. Also, it should be noted that for all the atoms there are regions of r_s within which $Z_{ef1} > 1$, i.e. the regions where atoms lose energy to the medium more effectively than their nuclei. This is due to the inclusion of the anti-screening of atomic electrons in the model given by Eq. (4).

An interesting result, with no experimental reference, is shown in Fig. 4. The straggling effective charge turns out to be several percent larger than the stopping effective charge, $Z_{ef2} > Z_{ef1}$. This difference decreases rapidly as $r_s \to \infty$, and also as $Z_i \gg 1$. Again the excited atoms (and hydrogen) are shifted towards dilute electron gas when compared to the atoms in the ground state.

4. Conclusions

The results of the calculation of the electronic energy loss and the straggling of the energy loss in free electron gas for low velocity excited ions were reported. The calculations were carried within the linear response theory. The electron gas was described by the Lindhard dielectric function. For H- to Be-like ions the Hartree–Fock–Slater description was used. A screening parameter of the wave function, Z_{min}, was calculated from a variational principle and shown to depend on r_s, Z_i, N. In consequence, a region in the (r_s, Z_i) plane can be found in which an ion of a given $1s2s$ electronic configuration is stable. It was shown that the effective charges for the energy loss (and also for straggling of it) for ions kept excited during the passage through matter and these in the ground state may differ by an order of magnitude. Particularly promising are He atoms in three states of excitation. It was found that $Z_{ef2} > Z_{ef1}$ and the difference is the largest for dense gas and light ions. For dense electron gas, this difference was shown to be smaller for ions being excited than for ions in the ground state. For dilute electron gas the differences are larger for excited ions.

Appendix

The energy (E), the Coulomb (V), and the exchange (A) integrals

$$E_a = \int \mathrm{d}^3 r \psi_a^*(r) \left(-\frac{1}{2}\Delta + \frac{Z_i}{r} \exp(-r k_{TF}) \right) \psi_a(r),$$

$$V_{ab} = \int \mathrm{d}^3 r \int \mathrm{d}^3 r' |\psi_a(r)|^2 |r - r'|^{-1} \exp(-|r - r'| k_{TF}) |\psi_b(r')|^2,$$

$$A_{ab} = \int \mathrm{d}^3 r \int \mathrm{d}^3 r' \psi_a^*(r) \psi_b^*(r') |r - r'|^{-1} \exp(-|r - r'| k_{TF}) \psi_a(r') \psi_b(r)$$

were calculated as ($\gamma = k_{\mathrm{TF}}\lambda$ and $\gamma' = 2\gamma/3$)

$$E_{1s} = \frac{Z^2}{2} - \frac{Z_{\mathrm{i}}Z}{(1+\gamma/2)^2}, \tag{9}$$

$$E_{2s} = \frac{Z^2}{8} - \frac{Z_{\mathrm{i}}Z}{4(1+\gamma)^2}\left[2 - \frac{4}{(1+\gamma)} + \frac{3}{(1+\gamma)^4}\right], \tag{10}$$

$$V_{1s1s} = Z\frac{5+4\gamma+\gamma^2}{8(1+\gamma/2)^4}, \tag{11}$$

$$V_{2s2s} = Z\frac{77+104\gamma+227\gamma^2+144\gamma^3+123\gamma^4+40\gamma^5+5\gamma^6}{512(1+\gamma)^8}, \tag{12}$$

$$V_{1s2s} = Z\frac{4}{81}\frac{17+4\gamma+21\gamma^2+8\gamma^3+\gamma^4}{(1+\gamma)^4(2+\gamma)^2}, \tag{13}$$

$$A_{1s2s} = Z\frac{16}{729}\frac{3-25\gamma'^2+150\gamma'^4-256\gamma'^5+150\gamma'^6-25\gamma'^8+3\gamma'^{10}}{3(1-\gamma'^2)^6} \tag{14}$$

in Hartree units.

For $\gamma' = 1$ the exchange integral A is finite $A_{1s2s} = Z(16/729)(5/12)$. In the high electron density limit, as $r_{\mathrm{s}} \to 0$, the energies $E_{1s} = 4E_{2s} = Z^2/2$ and the Coulomb and exchange integrals vanish.

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