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## ENERGY LOSS IN SPHERES BY PENETRATING ELECTRONS

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A classical dielectric theory is developed to provide an expression for the energy loss of fast electrons passing through a metallic sphere at a fixed impact parameter, in the case of spheres coated by a thin oxide layer. The expressions for energy loss are valid for any dielectric function  $\epsilon(\omega)$ . A high number of terms of the multipole expansion is required in order to take into account the effects of the oxide layer. Results are compared with experimental measurements of energy loss obtained by STEM techniques.

### 1. Introduction

The energy loss of fast electrons interacting with solids has been a subject of permanent interest over the years. When a fast electron approaches a polarizable body it creates excitations which in turn interact with the incident particle. The resulting interaction has a conservative component, coming from virtual excitations on the medium, and a dissipative component, arising from real excitations.

The case of a charged particle interacting with a spherical polarizable body has been of continuing interest because of its many applications in both

fundamental and applied physics. The recent development of the scanning transmission electron microscope (STEM) has allowed probing of small-sized bodies with a 0.2 nm beam of fast ( $\sim 100$  keV) electrons [1–5]. This instrument yields electron energy loss spectra from selected local regions of the sample. Fujimoto and Komaki [6] used the free-electron model for spherical targets to analyze electron loss spectra for a broad beam geometry. This model has been applied to the well-focused-beam case by Schmeits [7] and Kohl [8], but only for dipole ( $l=1$ ) and quadrupole ( $l=2$ ) excitations. More recently, the contribution of higher  $l$  modes has been identified [9]. The assumption of treating the fast electron as a point classical particle on focused beam configurations has been shown [10,11] to be generally valid in many situations. Barberán and Bausells [12] have analyzed the effects of the finite beam size in the energy loss probability of the first modes of the sphere. Using a more general dielectric formalism, Batson [1,2,13,14] has determined the resonance frequencies of small spheres, including the effects of surface coatings. Ferrell and Echenique [15] have presented a new form of the dielectric model including all multipoles, valid for electron trajectories outside a spherical particle. This model has been recently generalized to include the presence of a surface coating [16]. Very recently a new general self-energy approach to the problem of the interaction of fast electrons with solid surfaces has been presented [17] and has been applied to the problem of the energy loss of electrons with penetrating trajectories in metallic spheres.

In this paper we generalize the classical dielectric model of refs. [15,16] to the case of penetrating trajectories in spheres with surface coatings. Our formalism is valid for any dielectric response function  $\epsilon(\omega)$  of both the sphere and the coating and includes many ( $l, m$ ) multipoles. Our calculations are compared with existing experimental results.

## 2. Classical formalism

The total energy loss experienced by a charged particle  $q$  moving with constant velocity  $v$ , in a medium can be obtained for any dielectric response of the medium  $\epsilon(\omega)$  in the frame of the classical dielectric theory. Energy loss is given by

$$E_l = -q \int (\partial \phi^{\text{in}} / \partial z) dz, \quad (1)$$

where  $\phi^{\text{in}}$  is the scalar potential induced by the charge  $q$  evaluated at the charge position. This potential is obtained from the Poisson equation with the external charge density given by:

$$\rho(r, t) = q \delta(\mathbf{R} - \mathbf{s}) \delta(z - vt), \quad (2)$$

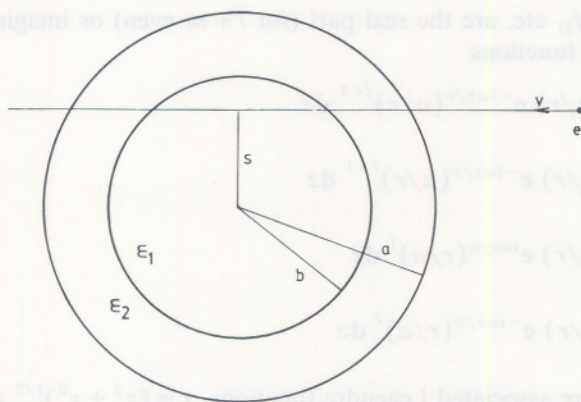


Fig. 1. Charged particle  $q$  moving at impact parameter  $s$  near two concentric dielectric spheres of radii  $a$  and  $b$ , and dielectric responses  $\epsilon_1$  and  $\epsilon_2$ .

where  $z$  has been taken in the same direction as the particle velocity and  $\mathbf{R}$  is the transversal component of the  $\mathbf{r}$  vector. The induced potential is:

$$\phi^{\text{in}}(\mathbf{r}, t) = \frac{1}{2\pi} \int W(\mathbf{r}, \mathbf{r}', \omega) e^{-i\omega(t-t')} \rho(\mathbf{r}', t') d\omega dt' d\mathbf{r}' \quad (3)$$

$W(\mathbf{r}, \mathbf{r}', \omega)$  is the Fourier  $\omega$ -component of the potential created by an unit charge at rest in point  $\mathbf{r}'$ , at point  $\mathbf{r}$ .

For the case of a charged particle interacting with two concentric dielectric spheres, of radii  $a$  and  $b$  and dielectric response  $\epsilon_1(\omega)$  and  $\epsilon_2(\omega)$  (fig. 1), the induced potential  $\phi^{\text{in}}$  can be easily obtained from standard electrodynamics by means of a multipole expansion of this potential. In such a way the energy loss can be written as:

$$E_l = \int_0^\infty \omega P_s(\omega) d\omega, \quad (4)$$

we use atomic units throughout. The energy-loss probability  $P_s(\omega)$  is given by:

$$\begin{aligned} P_s(\omega) = & \frac{q^2}{a} \frac{1}{(v\pi)^2} \sum_l \sum_{m=0}^l (2l+1)(2-\delta_{m0})(l-m)!/(l+m)! \\ & \times \{ \alpha_l(\omega) f_{1l}^2 + 2\gamma_l(\omega) f_{1l} f_{2l} + 2\beta_l(\omega) f_{1l} f_{3l} + 2\delta_l(\omega) f_{1l} f_{4l} \\ & + 2\eta_l(\omega) f_{2l}^2 - 2\xi_l(\omega) f_{2l} f_{3l} + 2\mu_l(\omega) f_{2l} f_{4l} \\ & + \rho^{2l+1} [\theta_l(\omega) f_{3l}^2 + 2\nu_l(\omega) f_{3l} f_{4l} + \xi_l(\omega) f_{4l}^2] \}, \quad (5) \end{aligned}$$

where  $\delta_{m0}$  is the Kronecker delta. Functions  $\alpha_l(\omega)$ ,  $\beta_l(\omega)$  and so on are the imaginary part of the response functions given in the appendix. The remaining

function  $f_{1l}$ ,  $f_{2l}$  etc. are the real part (for  $l + m$  even) or imaginary part (for  $l + m$  odd) of functions

$$\begin{aligned} f_{1l} &= \int_{r_a}^{\infty} P_{lm}(z/r) e^{-i\omega z/v} (a/r)^{l+1} dz \\ f_{2l} &= \int_{r_b}^{r_a} P_{lm}(z/r) e^{-i\omega z/v} (a/r)^{l+1} dz \\ f_{3l} &= \int_{r_b}^{r_a} P_{lm}(z/r) e^{i\omega z/v} (r/a)^l dz \\ f_{4l} &= \int_{r_a}^0 P_{lm}(z/r) e^{-i\omega z/v} (r/a)^l dz \end{aligned} \quad (6)$$

Here  $P_{lm}$  are associated Legendre functions,  $r = (z^2 + s^2)^{1/2}$  and  $r_a$  and  $r_b$  are the distances between the  $z = 0$  plane and the intersection of the particle trajectory with the spheres.

The loss due to bulk terms has not been included in eq. (5). To calculate the total energy loss one has to add in eq. (5) the well known expression for the bulk energy loss [19].

### 3. Results and discussion

The preceding formalism has been applied to the case of 100 keV electrons incident on an aluminium sphere of radius 200 au coated by an  $\text{Al}_2\text{O}_3$  layer. We use experimental values of the  $\epsilon(\omega)$  response functions for both materials [20]. In order to get good convergence in our calculation more than forty  $l$  multipole terms were generally required. In fig. 2 we show the energy loss probability  $P_s(\omega)$  as a function of the energy  $\omega$  for three values of the impact parameter  $s$ . The thickness of the oxide layer is 40 au. The peaks at 7.2 eV correspond to the surface aluminium modes and their positions are independent of the impact parameter. The peak height has a maximum value when the incident trajectory crosses near the Al- $\text{Al}_2\text{O}_3$  interface. In fig. 2a the peak at 15.1 eV correspond to the bulk Al mode.

Fig. 2b has been obtained for an impact parameter  $s = 210$  au and the incident trajectory passes through the  $\text{Al}_2\text{O}_3$  layer. For  $\omega$  values above 10 eV the energy loss probability coincides qualitatively with the probability obtained for an electron incident axially over an  $\text{Al}_2\text{O}_3$  sphere of similar size. The peak at 24.4 eV is the bulk  $\text{Al}_2\text{O}_3$  mode. This mode should be responsible of the fact that in fig. 2a the loss probability in this energy region decreases more slowly than in an oxide-free Al sphere. [18]

In fig. 2c the electron trajectory is outside the spheres. Our results then reproduce those of ref. [16]. In our approximation only surface modes contribute to the energy loss probability. Therefore the losses above 10 eV can be

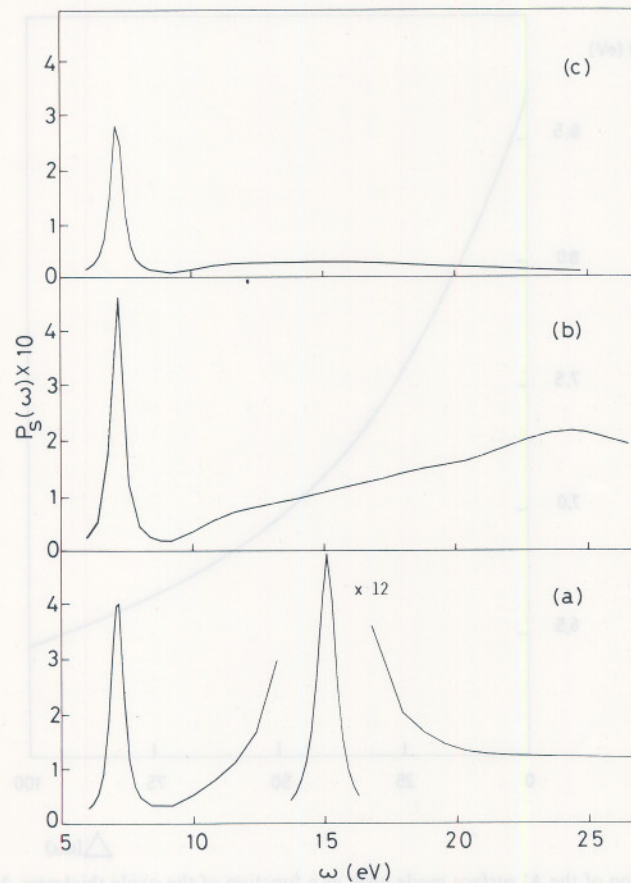


Fig. 2. Energy loss probability as a function of the energy  $\omega$  for an electron incident over an Al sphere of radius 200 au with an  $\text{Al}_2\text{O}$  layer of thickness 40 au. (a) Axial trajectory. (b) Impact parameter  $s = 210$  au. (c) Impact parameter  $s = 250$  au.

interpreted as due to surface modes of the oxide. From our calculations the maximum loss appears in the neighbourhood of 15.5 eV. This surface losses presumably contribute the loss probability in the energy region of 10–20 eV of fig. 2b.

The results of figs. 2a and 2c are in good qualitative agreement with the experimental results of Batson and Treacy [21]. The fact that the Al surface mode peak position doesn't depend on the impact parameter was also observed in the same work. Also, the qualitative behaviour of the Al surface mode peak height in ref. [21] agrees with our results.

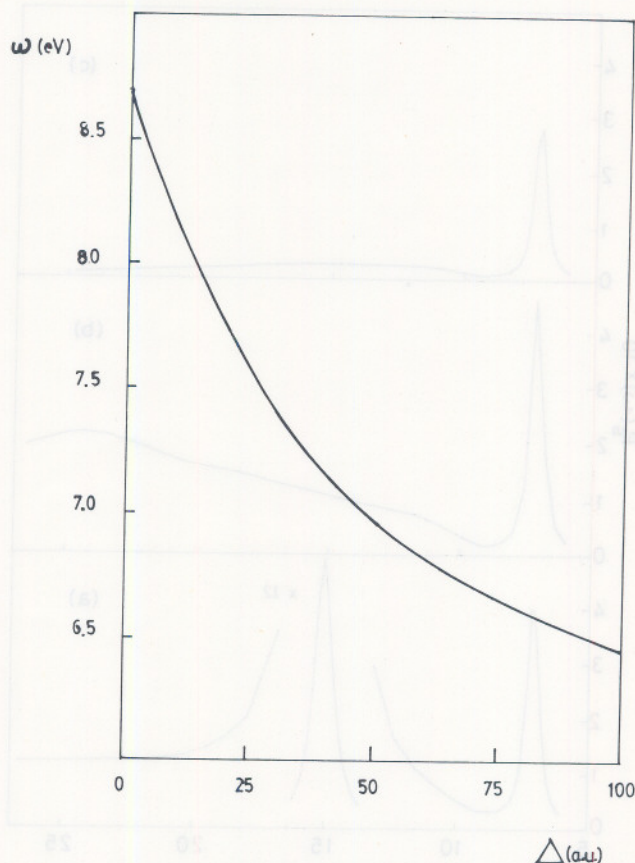


Fig. 3. Position of the Al surface mode peak as a function of the oxide thickness  $\Delta = a - b$ .

In fig. 3 we show the dependence of the Al surface mode peak position on the thickness of the oxide layer. The radius of the Al sphere is 200 au and the electron trajectories are axial. For small oxide thickness the peak position tends to the bare Al result  $\omega_p/\sqrt{3} = 8.7$  eV for  $l = 1$  and for big oxide thickness it approaches the result for an Al sphere surrounded by an infinite  $\text{Al}_2\text{O}_3$  medium, which is 5.2 eV for  $\epsilon_{\text{ox}} = 3.73$ .

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## Appendix

$$\alpha_l(\omega) = \frac{4\pi}{2l+1} \frac{l}{l+1} \frac{1}{\Delta} \left[ (1 - \epsilon_3) \left( 1 + \frac{l+1}{l} \epsilon_2 \right) - \rho^{2l+1} (1 - \epsilon_2) \left( 1 + \frac{l+1}{l} \epsilon_3 \right) \right].$$

$$\gamma_l(\omega) = \frac{4\pi}{l+1} \frac{(1 - \epsilon_3)}{\Delta}.$$

$$\beta_l(\omega) = -\frac{4\pi}{l+1} \rho^{2l+1} \frac{1}{\Delta} \left( 1 + \frac{l+1}{l} \epsilon_3 \right).$$

$$\delta_l(\omega) = -\frac{4\pi}{l+1} \frac{2l+1}{l} \rho^{2l+1} \frac{\epsilon_3}{\Delta}.$$

$$\theta_l(\omega) = \frac{4\pi}{2l+1} \frac{(1 - \epsilon_2)(1 + \epsilon_3(l+1)/l)}{\epsilon_2 \Delta}.$$

$$\xi_l(\omega) = \frac{4\pi}{2l+1} \frac{l+1}{l} \frac{1}{\Delta \epsilon_1}$$

$$\times \{ (1 - \epsilon_2) [1 + \epsilon_3 l / (l+1)] - \rho^{2l+1} (1 - \epsilon_3) [1 + \epsilon_2 l / (l+1)] \}.$$

$$\eta_l = \frac{4\pi}{2l+1} \frac{(1 - \epsilon_3) [1 + \epsilon_2 l / (l+1)]}{\Delta \epsilon_2}.$$

$$\zeta_l = -\frac{4\pi}{2l+1} \frac{(1 - \epsilon_2)(1 - \epsilon_3)}{\epsilon_2 \Delta}.$$

$$\mu_l = -\frac{4\pi}{l} \rho^{2l+1} \frac{1 + \epsilon_2 l / (l+1)}{\epsilon_1 \Delta}.$$

$$\nu_l = \frac{4\pi}{l} \frac{(1 - \epsilon_2)}{\epsilon_1 \Delta}.$$

$$\Delta = (1 - \epsilon_2)(1 - \epsilon_3) - [1 + \epsilon_3(l+1)/l][1 + \epsilon_2 l / (l+1)] \rho^{2l+1}.$$

$$\epsilon_3 = \frac{\epsilon_2}{\epsilon_1}, \quad \rho = a/b.$$

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