**From: REFLECTION HIGH ENERGY ELECTRON DIFFRACTION
AND REFLECTION ELECTRON IMAGING OF SURFACES**
Edited by P. K. Larsen and P. J. Dobson
(Plenum Publishing Corporation, 1988)

EXCITATION OF DIELECTRIC SPHERES BY ELECTRON BEAMS

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Recent new developments and applications of the scanning transmission microscope (STEM) have stimulated renewed interest in the interaction of high-energy electron beams with small particles and surfaces[1-7]. Usually the classical theory of energy loss has been employed to analyze the experimental energy-loss spectra for planar, spherical or cylindrical geometries[8-14]. A new general approach, based on the self-energy formalism of the many body problem has been presented recently[15]. In this approach, the mean potential energy Σ_0 of the incoming electron, in a state defined by a wave function $\psi_0(r)$ and energy E_0 is written as the average of an effective local potential $V_{\text{eff}}(r)$, representing the complex interaction of the incoming electron with the many body target

$$\Sigma_0 = \int \psi_0^*(\vec{r}) V_{\text{eff}}(r) \psi_0(\vec{r}) d\vec{r}. \quad (1)$$

We use atomic units ($e^2 = \hbar = m$) throughout this paper. The real part of Σ_0 gives us the lowering of the energy of the particle due to virtual excitations of the medium, and the imaginary part is directly related to the probability of energy loss due to creation of real excitations. The effective potential is written as a spatial integral of the non local self-energy[16], $\Sigma(r, r', E_0)$, which in turn can be expressed in terms of the Green's function $G(r, r', \omega + E_0)$ and the causal screened interaction $W(r, r', \omega)$ [16]. The Green's function is given as a sum over a complete set $\Psi_f(r)$ of final states with energy E_f . After some algebra, we get

$$\Sigma_0 = \frac{1}{\pi} \Sigma_f \int \frac{\psi_f^*(\vec{r}) \psi_0(\vec{r}) \psi_f(\vec{r}) \psi_0^*(\vec{r})}{\omega + \Delta E + i\delta} \times \text{Im}[W^R(\vec{r}, \vec{r}', \omega)] \times d\omega d\vec{r} d\vec{r}' \quad (2)$$

where $\Delta E = E_0 - E_f$, δ is an infinitesimally small positive constant and W^R stands for retarded response function. A quantity of practical utility in electron microscopy is the probability of losing energy ω , P_ω , which is directly related to the energy loss rate γ , experienced by the particle

$$\gamma = v \int P_\omega d\omega \quad (3)$$

where v is the velocity of the incident electron. The energy loss rate, γ , is in turn, given in terms of the imaginary part of the incident electron self-energy

$$\gamma = -2 \operatorname{Im} \Sigma^0 \quad (4)$$

Now we use Equation (2) for the case of an electron of velocity v incident upon a dielectric sphere, radius a , characterized by a dielectric response function $\epsilon(\omega)$. The screened interaction is obtained from the Poisson equation[15,17].

To proceed further, we need to specify the initial state Ψ_0 . If we have a broad-beam geometry, i.e. if the width of the incident electron beam is much greater than the size of the sphere, we use plane waves to describe the incident electron states. The expression one obtains[15] is valid for any dielectric response function $\epsilon(\omega)$. When the further approximation of the free electron gas model is made, it reproduces the results of Fujimoto and Komaki[18] (when recoil is completely neglected) and the results of Barberán and Bausells[19] (when only the forward scattering recoil is neglected). In STEM experiments we look for the probability of energy loss for a well-focused beam incident upon a sphere of radius a at a fixed impact parameter b . A physical description can be obtained, if we neglect beam size effects, by taking, in Equation (2) a δ function in the transverse direction and plane waves in the direction of motion to describe the electron[20]. When the incident electron moves outside the sphere, the probability of energy loss obtained from Equation (2) agrees with the result first obtained by the use of classical electrodynamics by Ferrell and Echenique[10], and is given in terms of the usual angular momentum components ℓ, m by

$$P_{\omega}(b) = \frac{4}{\pi v^2 a^2} \sum_{\ell=0}^{\infty} \sum_{m=0}^{\ell} \frac{(2 - \delta_{0m})}{(\ell+m)! (\ell-m)!} \times \left[\frac{\omega a}{v} \right]^{2\ell} \times K_m^2 \left[\frac{\omega b}{v} \right] \times \operatorname{Im} \left[\frac{(\epsilon(\omega) - 1)a^3}{\epsilon(\omega) + (\ell + 1/\ell)} \right]. \quad (5)$$

Here δ_{0m} is the Kronecker delta, and K_m is the modified Bessel function of order m . In Equation (5) $b > a$. For $\ell = 1$, the dipole contribution goes over into the usual result; in this case, the energy loss is proportional to the square of the ω Fourier component of the external field, if such a field is taken to be constant over the volume of the sphere[17]. The usual result for a harmonically bound charge replacing the sphere is then recovered when one takes a unit volume containing a single charge replacing the sphere and if $\epsilon(\omega)$ is taken to be descriptive of such a situation.

In general, many (ℓ, m) terms are necessary to describe adequately the interaction process. The dipole ($\ell=1$) approximation is only valid either when the electron is far away from the sphere, or for very small spheres, which is not the case in most experimental situations. To illustrate this point, we first consider a free electron response $\epsilon(\omega)$ and calculate the energy-loss probability as a function of impact parameter for a 50-keV electron incident upon a sphere of radius 100 Å. In Fig. 1, we show the bulk losses, the dipole mode contribution and the total contribution of the first twenty modes. The dipole contribution, however, does not show the probability enhancement near the surface, which also can be seen in the experimental results[21]. In Fig. 2 we show, for a grazing incident electron of energy 50 keV the relative weight of the contribution to the total energy loss of the ℓ^{th} mode, in the free-electron gas model, for a one electron radius of $r_s = 2$, for different sphere radii. As it is clear from the figure for large enough radii, the dipole contribution is negligible. Also we show, on the right-hand side the relative contributions of the first ten ℓ modes as function of the sphere radius. In Fig. 3, we show the probability of losing energy ω for a 50 keV electron at grazing incidence

Fig. 1. Energy-loss probability [calculated in the free electron gas model, $r_s = 2$] as a function of impact parameter b for a 50 keV electron incident upon an aluminium sphere of radius 100 Å. Solid line, bulk mode; dashed line, total contribution of the first 20 surface modes ($\times 6$); dashed-dotted line, dipole ($\ell = 1$) surface mode ($\times 6$).

Fig. 2. Relative weight R_ℓ of the ℓ^{th} mode contribution to the total energy loss [calculated in the free-electron gas model for $r_s = 2$ as a function of the mode number for an electron of energy 50 keV at grazing incidence ($b=a$) for different sphere radii]. Also, the contribution to the loss of the first ten ℓ modes is shown as a function of the radius of the sphere.

Fig. 3. Probability P_{ω} losing energy ω for a 50 keV electron moving at grazing incidence on an aluminium sphere of radius $a = 10$ nm... dipole mode $\ell = 1$; -- $\ell = 1 + \ell = 2$; -.- contribution of the first ten ℓ modes; — contribution up to $\ell = 100$ included.

on a sphere of radius $a = 10$ nm. We show the contributions of the dipole mode, the first two ℓ 's, and ten ℓ 's, and the total probability. For small radii and low energy losses, the dipole contribution dominates; while even at as a small radius as 10 nm, it is inadequate to describe the high energy losses. We have used experimental optical data for $\epsilon(\omega)$ in these calculations[22].

CONCLUSIONS

We have presented a general formula to calculate the probability-loss function in electron microscopy, and in RHEED spectroscopy, and apply it to some specific cases. Our analysis is valid for any ω -dependent dielectric response function $\epsilon(\omega)$. We have shown that, for most cases of practical interest in electron microscopy, or even more in RHEED, many multipoles (ℓ, m) should be included to describe adequately the experimental data over the relevant energy range. The free electron model for spherical targets, originally used by Fujimoto and Komaki[18] in broad-beam geometry, has been applied to the focused-beam case by Schmeits[23] and Kohl[24] but only for the case of dipole or quadrupole excitations. The case of a sphere coated with an oxide layer can be analyzed easily within our formalism. Calculations using the classical theory of energy loss valid for any $\epsilon(\omega)$ [11,12, 25] or within the free electron gas model[26] have been presented recently. No account of beam profile has been given, but beam size effects can be introduced in a natural way into our formalism. Ritchie and Howie have proved that if all inelastically scattered electrons are collected, the quantum mechanical excitation probability is obtained by convoluting the classical, impact parameter dependent result over the distribution of impact parameter determined by the probe intensity function[27]. Spatial dispersion and relativistic effects have been discussed, within the specular reflection model of Ritchie and Marusak[28] by Echenique[29]. Retardation effects for the case of planar geometries have been analyzed by Garcia-Molina et al.[30]

Acknowledgement

It is a pleasure to thank Professors A. Howie and R.H. Ritchie for many stimulating conversations and Iberduero S.A. for help and support. Support for the Spanish CAYCIT, Eusko Jaurlaritza and Gipuzkoako Foru Aldundia is gratefully acknowledged.

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