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Retardation effects in the interaction of charged particle beams with bounded condensed media

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Abstract. The interaction of a relativistic electron beam, travelling parallel to the surface of a semi-infinite medium, is analysed. The specific energy loss of the beam and the probability of excitation of surface and bulk modes in the medium are calculated. Beams both external and internal with the medium are considered, and the predictions are compared with well known non-retarded limits. A detailed analysis of the expressions derived is provided for electron beams interacting with specific dielectric materials. The effects of retardation are seen to be large, particularly for beam energy losses in regions where the real part of the dielectric constant is large. The retarded excitation probability is typically about 10–30% larger than the non-retarded result, for electron energies greater than about 100 keV. The image force is calculated for an external beam, and the radiative de-excitation of the medium is also analysed.

1. Introduction

The interaction of electron beams with thin films has been studied extensively by means of the dielectric theory (Ritchie 1957, Ritchie and Eldridge 1962, Otto 1967, Kröger 1968, 1970), including the retardation of the electromagnetic field due to the finite velocity of light. The direction of the beam was taken to be perpendicular to the film by Ritchie and Eldridge (1962) and an arbitrary direction was considered by Otto (1967). In transmission experiments (von Festenberg 1969) both bulk and surface plasmons are seen, but the thinner the film is, the more the surface modes should dominate. Electron energy losses also occur as a result of the Cherenkov radiation generated in the dielectric medium (Kröger 1968, 1970). For electrons impinging at grazing incidence or which interact weakly with the medium by passing above it the excitation of surface modes becomes important (Echenique *et al* 1981, Garcia-Molina *et al* 1985).

Only recently have experiments been performed (Marks 1982, Cowley 1982a, b, Wheatley *et al* 1983), where the impact parameter of the beam (or beam–surface distance) is maintained constant, and the beam probes the surface plasmon field of the irradiated material apart from, possibly, the excitation modes of the bulk.

Some capabilities of the modern analytical scanning transmission electron microscope (STEM) have been discussed by Pennycook (1981). This gives energy-loss spectra from precisely defined regions near surfaces or other known internal defects such as precipitates or dislocations. One may thus obtain information from the loss spectra (Pennycook 1981), or from cathodoluminescence (Pennycook *et al* 1980) about localised electron states associated with the defects—such states can be in the energy gap.

It has been suggested by Cowley (1982a, b) that relativistic effects may be important in the interpretation of recent STEM experiments, where electrons with an energy of about 100 keV interact with small crystallites. It is our purpose here to estimate when retardation effects are expected to contribute to the electron-energy-loss spectrum (EELS) of high-energy electrons.

We shall calculate the excitation probability and specific energy loss, including retardation, of electron beams travelling parallel to, both inside and outside, the plane surface of a semi-infinite solid. The non-retarded limit will be retrieved, in particular. Predictions will be made for specific dielectric media characterised by a frequency-dependent complex dielectric function.

The expression derived below for a relativistic beam external to a semi-infinite solid is equivalent to that given by Otto (1967) without a detailed derivation. He did not consider the case of a beam travelling through the material and parallel with its surface.

The conditions under which radiative emission from the excited medium may take place will also be investigated.

The question of significant deflection due to image or other forces (e.g. electrostatic charging of the target) arose in Cowley's experiments (1982a, b). Some discussions given by Howie (1983) and by Echenique and Pendry (1975) will be extended to study the effect of retardation on the force experienced by the beam due to the presence of the crystal.

2. Energy loss and excitation probability

We wish to consider the cases of a charged beam travelling parallel to the planar surface of a semi-infinite medium, both external and internal to the condensed medium. Although one of the semi-spaces will usually be the vacuum, it is convenient to consider the general case of two adjacent materials separated by a planar boundary and characterised by the dielectric functions $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$ as shown in figure 1.

We shall make use of the (classical) dielectric approach to calculate the excitation probability and the energy loss of the relativistic charge. We solve Maxwell's equations in terms of the Hertz vector $\mathbf{\Pi}$. The electromagnetic fields are given by Stratton (1941)

$$\boldsymbol{E} = \nabla(\nabla\mathbf{\Pi}) + (\varepsilon\omega^2/c^2)\mathbf{\Pi} \tag{1}$$

$$\boldsymbol{H} = -(\mathrm{i}\omega\varepsilon/c)\nabla \times \mathbf{\Pi} \tag{2}$$

where the spectral analysis used is

$$\mathbf{\Pi}(\boldsymbol{r}, t) = \int_{-\infty}^{\infty} \frac{\mathrm{d}\omega}{2\pi} \exp(-\mathrm{i}\omega t)\mathbf{\Pi}(\boldsymbol{r}, \omega) \tag{3}$$

and similarly for the other quantities. Here c denotes the velocity of light. The wave equation for the Hertz potential follows from Maxwell's equations (Stratton 1941):

$$\left(\nabla^2 + \frac{\varepsilon\omega^2}{c^2}\right)\mathbf{\Pi} = (4\pi/\mathrm{i}\omega\varepsilon)\boldsymbol{J} \tag{4}$$

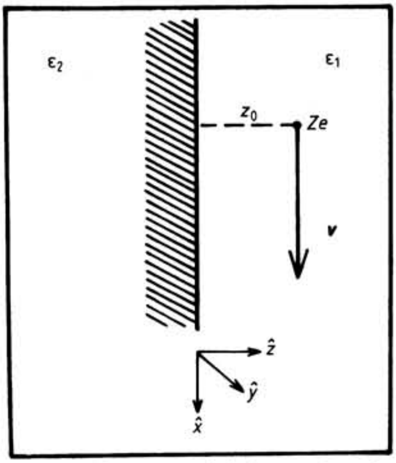

Figure 1. Geometry of the interaction of a charged particle Ze, moving with velocity v, at a distance z_0 from the interface between media 1 and 2, characterised by the complex dielectric functions $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$.

where $\boldsymbol{J}$ is the current density and the laplacian operator is supposed to act on each cartesian component of $\boldsymbol{\Pi}$.

The charged beam which moves along the x axis with (constant) velocity v and at a fixed distance z_0 from the surface, is defined by

$$\boldsymbol{J}(\boldsymbol{r}, t) = Zev\hat{x}\delta(x - vt)\delta(y)\delta(z - z_0). \tag{5}$$

where $\hat{x}$ is a unit vector along the x direction.

There is translational invariance parallel to the plane surface (the xy plane) and we introduce Fourier transforms

$$\boldsymbol{\Pi}(\boldsymbol{r}, \omega) = \int \frac{\mathrm{d}^2 k}{(2\pi)^2} \exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{\rho})\boldsymbol{\Pi}(\boldsymbol{k}, \omega, z) \tag{6}$$

where $\boldsymbol{\rho} = (x, y, 0)$ and $\boldsymbol{k} = (k_x, k_y, 0)$. Then Fourier transforming equation (5) according to equations (3) and (6) gives:

$$\boldsymbol{J}(\boldsymbol{k}, \omega, z) = 2\pi Zev\hat{x}\delta(\omega - k_x v)\delta(z - z_0). \tag{7}$$

From symmetry, one may assume that $\boldsymbol{\Pi}$ has components only in the $\hat{x}$ and $\hat{z}$ directions, $\boldsymbol{\Pi} = (\Pi_x, 0, \Pi_z)$. The equations one needs to solve, for a beam external to the solid, are from (4) and (7),

$$\begin{aligned}
&(\mathrm{d}^2/\mathrm{d}z^2 - \nu_2^2)\Pi_x^- = 0 \\
&\left(\frac{\mathrm{d}^2}{\mathrm{d}z^2} - \nu_1^2\right)\Pi_x^+ = \frac{8\pi^2 Zev}{\mathrm{i}\omega\varepsilon_1}\delta(\omega - k_x v)\delta(z - z_0) \\
&(\mathrm{d}^2/\mathrm{d}z^2 - \nu_2^2)\Pi_z^- = 0 \\
&(\mathrm{d}^2/\mathrm{d}z^2 - \nu_1^2)\Pi_z^+ = 0
\end{aligned} \tag{8}$$

where $\boldsymbol{\Pi}^{\pm}$ represents the Hertz vector for $z \gtrless 0$, and

$$\begin{aligned}
\nu_1 &= (k^2 - \varepsilon_1\omega^2/c^2)^{1/2} \\
\nu_2 &= (k^2 - \varepsilon_2\omega^2/c^2)^{1/2}.
\end{aligned} \tag{9}$$

The solutions to equations (8) which are not divergent at infinity are:

$$\Pi_x^- = C\exp(\nu_2 z) \tag{10a}$$

$$\Pi_x^+ = -(4\pi^2 Zev/\mathrm{i}\omega\varepsilon_1\nu_1)\delta(\omega - k_x v)\exp(-\nu_1|z - z_0|) + A\exp(-\nu_1 z) \tag{10b}$$

$$\Pi_z^- = D\exp(\nu_2 z) \tag{10c}$$

$$\Pi_z^+ = B\exp(-\nu_1 z). \tag{10d}$$

Both ν_1 and ν_2 are understood to have a positive real part in all subsequent manipulations, in order that the expressions in equations (10) be bounded for large z.

The four constants appearing in the solutions must be evaluated from the continuity of the tangential (i.e. the x and y) components of the fields $\boldsymbol{E}$ and $\boldsymbol{H}$ at the boundary. According to equations (1), (2), (3) and (6) the corresponding conditions for the Hertz potential require

$$\begin{aligned} &\varepsilon_1\Pi_x^+ = \varepsilon_2\Pi_x^- \\ &\mathrm{i}k_x\Pi_x^+ + \partial\Pi_z^+/\partial z = \mathrm{i}k_x\Pi_x^- + \partial\Pi_z^-/\partial z \\ &\varepsilon_1\Pi_z^+ = \varepsilon_2\Pi_z^- \\ &\varepsilon_1\partial\Pi_x^+/\partial z = \varepsilon_2\partial\Pi_x^-/\partial z \end{aligned} \tag{11}$$

evaluated at $z = 0$. For our purposes only the coefficients A and B will be required, and these are given by

$$A = [(\nu_1 - \nu_2)/(\nu_1 + \nu_2)]\Lambda \tag{12a}$$

$$B = 2\mathrm{i}k_x\nu_1\Lambda[(\varepsilon_2 - \varepsilon_1)/(\nu_1 + \nu_2)(\nu_2\varepsilon_1 + \nu_1\varepsilon_2)] \tag{12b}$$

where

$$\Lambda = -(4\pi^2 Zev/\mathrm{i}\omega\varepsilon_1\nu_1)\exp(-\nu_1 z_0)\delta(\omega - k_x v). \tag{12c}$$

The retarding force at the particle in the $(-x)$ direction is equal to its energy loss per unit path length. This is

$$\begin{aligned} -\frac{\mathrm{d}W}{\mathrm{d}x} &= -ZeE_x(vt, 0, z_0, t) \\ &= -\frac{Ze}{(2\pi)^3}\int_{-\infty}^{\infty}\mathrm{d}k_x\int_{-\infty}^{\infty}\mathrm{d}k_y\int_{-\infty}^{\infty}\mathrm{d}\omega\exp[\mathrm{i}(k_x v - \omega)t]E_x(\boldsymbol{k}, \omega, z_0). \end{aligned} \tag{13}$$

It is also useful to consider the probability of excitation of a frequency ω per unit path and per unit frequency, $\mathrm{d}^2P/\mathrm{d}x\,\mathrm{d}\omega$, defined through (Ritchie 1957):

$$\frac{\mathrm{d}W}{\mathrm{d}x} = \int_0^{\infty}\frac{\mathrm{d}^2P}{\mathrm{d}x\,\mathrm{d}\omega}\hbar\omega\,\mathrm{d}\omega. \tag{14}$$

We now consider the cases of external and internal beams separately.

2.1. External beam

A relativistic charged particle beam skims along, in the vacuum, (i.e. $\varepsilon_1 = 1$) next to a semi-infinite solid which is characterised by a dielectric function $\varepsilon_2 = \varepsilon(\omega)$. In calculating the specific energy loss, equation (13), one neglects the inhomogeneous term (the one containing the $\delta(\omega - k_x v)$ in equation (10*b*)) in order to subtract from the total field the part that is related with the beam moving in vacuum.

From equations (1), (10) and (13), after some algebra, one obtains

$$-\frac{\mathrm{d}W}{\mathrm{d}x} = \frac{2(Ze)^2}{\pi v^2}\int_0^\infty \mathrm{d}k_y \int_0^\infty \mathrm{d}\omega\, \omega \frac{\exp(-2\nu_0 z_0)}{\nu_0}\{\mathrm{Im}\,\lambda_e(\omega, k)\} \tag{15}$$

where the so-called retarded loss function is given by

$$\lambda_e(\omega, k) = \frac{1}{\nu_0 + \nu}\left(\frac{2\nu_0^2(\varepsilon - 1)}{\varepsilon\nu_0 + \nu} - (1 - \beta^2)(\nu_0 - \nu)\right) \tag{16}$$

and

$$k^2 = k_y^2 + \omega^2/v^2 \tag{17}$$

has to be inserted into equation (9), which becomes

$$\begin{aligned} \nu &= [k_y^2 + (\omega^2/v^2)(1 - \varepsilon\beta^2)]^{1/2} \\ \nu_0 &= [k_y^2 + (\omega^2/v^2)(1 - \beta^2)]^{1/2} \end{aligned} \tag{18}$$

and $\beta = v/c$.

The probability of excitation follows from equations (14) and (15)

$$\frac{\mathrm{d}^2 P}{\mathrm{d}x\,\mathrm{d}\omega} = \frac{2(Ze)^2}{\pi\hbar v^2}\int_0^\infty \mathrm{d}k_y \frac{\exp(-2\nu_0 z_0)}{\nu_0}\mathrm{Im}\{\lambda_e(\omega, k)\}. \tag{19}$$

Without explicit derivation, Otto (1967) provided a slightly different, but equivalent, form for the retarded loss function in equation (16). Otto also analysed the retarded dispersion relation of a surface plasmon at the boundary of the bulk material, and considered the case of a slightly damped free-electron gas.

One may check the limit $c \to \infty$, corresponding to a non-relativistic beam. Then $\nu \to \nu_0 \to k$, and the classical excitation probability becomes

$$\frac{\mathrm{d}^2 P}{\mathrm{d}x\,\mathrm{d}\omega} = \frac{2(Ze)^2}{\pi\hbar v^2} K_0(2\omega z_0/v)\,\mathrm{Im}\left(\frac{\varepsilon - 1}{\varepsilon + 1}\right) \tag{20}$$

which agrees with the results found neglecting retardation (Howie 1983). $K_0(z)$ is a modified Bessel function (Abramowitz and Stegun 1972) of zeroth order.

One should note,that the non-retarded and the retarded loss functions are very similar in magnitude for a wide range of values of the real and the imaginary parts of the dielectric function, and of k_y and v. (A more detailed discussion follows in § 4).

It is also of interest to evaluate the transverse force, $F_\perp$, experienced by the beam due to the presence of the surface. We neglect the contribution to the force due to the magnetic field, since we are not interested in extremely high velocities. The electric field in the z direction is required, and one obtains an attractive transverse force given by

$$F_\perp = -\frac{2(Ze)^2}{\pi v}\int_0^\infty \mathrm{d}k_y \int_0^\infty \mathrm{d}\omega \exp(-2\nu_0 z_0)\,\mathrm{Re}\left(\frac{\varepsilon\nu_0 - \nu}{\varepsilon\nu_0 + \nu}\right) \tag{21a}$$

which is also found following the procedure given by Kröger (1970). The non-retarded limit was derived by Takimoto (1966) and Howie (1983):

$$F_\perp = -\frac{2(Ze)^2}{\pi v^2}\int_0^\infty \mathrm{d}\omega\, \omega K_1\left(\frac{2\omega z_0}{v}\right)\mathrm{Re}\left(\frac{\varepsilon - 1}{\varepsilon + 1}\right) \tag{21b}$$

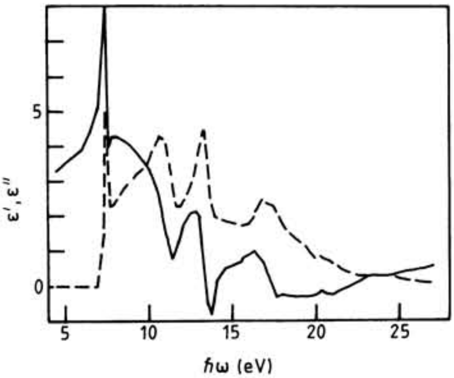

Figure 2. The complex dielectric function of MgO, from Roessler and Walker (1967). Full curve, $\varepsilon' = \mathrm{Re}\,\varepsilon$; broken curve, $\varepsilon'' = \mathrm{Im}\,\varepsilon$.

where K_1 is a modified Bessel function of first order (Abramowitz and Stegun 1972). Realistic numerical calculations of the effect of the attractive image force on the beam trajectory for a given material require detailed knowledge of the dielectric function $\varepsilon(\omega)$. An analysis of the integrands in equations (21*a*) and (21*b*) indicates that the transverse force is always larger when retardation effects are included. For instance, for a 100 keV electron beam interacting at a distance of 5 Å with a MgO surface, by using the data in figure 2, the ratio of the transverse forces is about 1.04.

2.2. *Internal beam*

When the charged particle beam travels through the condensed medium, one sets $\varepsilon_1 = \varepsilon(\omega)$, $\varepsilon_2 = 1$, and one has to consider both terms in equation (10*b*) in order to find the specific energy loss. The result is

$$-\frac{\mathrm{d}W}{\mathrm{d}x} = \frac{2(Ze)^2}{\pi v^2}\int_0^\infty \mathrm{d}k_y \int_0^\infty \mathrm{d}\omega\, \omega\, \mathrm{Im}\{\lambda_i(\omega, k)\} \tag{22}$$

where now the loss function is

$$\lambda_i(\omega, k) = \frac{1}{\nu_0 + \nu}\left(\frac{2\nu^2(1-\varepsilon)}{\varepsilon\nu_0 + \nu} - (1-\varepsilon\beta^2)(\nu - \nu_0)\right)\frac{\exp(-2\nu z_0)}{\nu\varepsilon} - \frac{1-\varepsilon\beta^2}{\nu\varepsilon}. \tag{23}$$

Both interface terms and bulk terms arise in the loss function. The term in equation (23) which is independent of z_0, corresponds to the excitation of bulk modes in an infinite medium (Landau and Lifshitz 1982) and does not appear in the case of an external beam when spatial dispersion in the medium is neglected.

The non-retarded limit is again a useful reference. For $c \to \infty$, equation (22) becomes

$$-\frac{\mathrm{d}W}{\mathrm{d}x} = \frac{2(Ze)^2}{\pi v^2}\int_0^\infty \mathrm{d}k_y \int_0^\infty \mathrm{d}\omega \frac{\omega}{k}\left[\mathrm{Im}\left(\frac{2}{1+\varepsilon} - \frac{1}{\varepsilon}\right)\exp(-2kz_0) + \mathrm{Im}(1/\varepsilon)\right] \tag{24}$$

or, in terms of the excitation probability for a classical beam,

$$\frac{\mathrm{d}^2P}{\mathrm{d}x\,\mathrm{d}\omega} = \frac{2(Ze)^2}{\pi\hbar v^2}\left[\mathrm{Im}(-1/\varepsilon)[\ln(k_c v/\omega) - K_0(2\omega z_0/v)] + \mathrm{Im}\left(\frac{\varepsilon-1}{\varepsilon+1}\right)K_0(2\omega z_0/v)\right]. \tag{25}$$

Here k_c, a cut-off wavenumber, has to be introduced in order to prevent the divergence of the integral for $k_y \to \infty$. This cut-off must be taken such that $k_c \gg \omega/v$, and the excitation probability is positive-definite.

3. Radiative emission

The energy losses suffered by a relativistic particle passing through, or close to, a dielectric medium can be partly due to electronic excitations, e.g. bulk or surface plasmons, and partly due to radiative excitations, e.g. Cherenkov radiation, which occurs when $\varepsilon'\beta^2 \geqslant 1$. We now briefly consider whether the radiation can be detected, particularly in the case when the beam is outside the dielectric medium.

In MgO, for instance, a material to be analysed in the following section, the published dielectric data, figure 2 (Roessler and Walker 1967), indicate that the Cherenkov condition holds for photon energies between 0 and 10 eV for a 100 keV electron beam and for photon energies between 6 and 8 eV for an 80 keV electron beam. It is only in the region below the band gap energy, $E_g = 7.5$ eV, however where $\varepsilon'' = 0$ that we can definitely identify all the losses given by equations (15) or (22) as radiative losses.

To be detected, the Cherenkov photons must emerge from the dielectric medium and not be confined by total internal reflection. Reference to figure 3, depicting the situation in a small cube, shows that photons will emerge from the lower face of the cube provided that the semi-angle $\theta = \sec^{-1} \beta(\varepsilon')^{1/2}$ does not exceed the critical angle $\theta_c = \operatorname{cosec}^{-1}(\varepsilon')^{1/2}$. Combining this result with the Cherenkov condition we obtain

$$0 \leqslant \varepsilon' - c^2/v^2 \leqslant 1. \qquad (26)$$

For 100 keV electrons this condition is satisfied for photon energies between 4 and 9 eV. For 80 keV electrons the allowed photon energies lie between 6 and 7 eV. In the case of a small cube, the length of the side L may possibly set an upper limit $\lambda \simeq 2L$ to the wavelengths of photons which can be excited in the medium. This corresponds to a lower

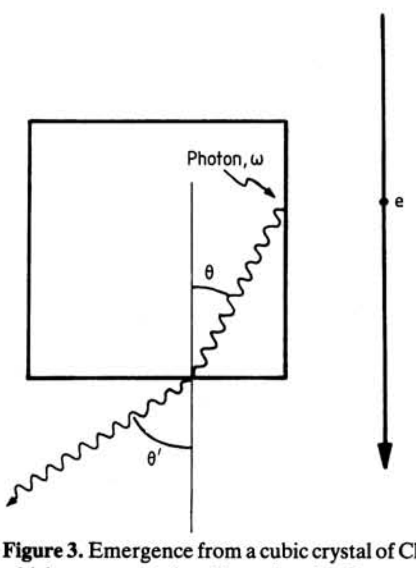

Figure 3. Emergence from a cubic crystal of Cherenkov radiation created by a charged beam which passes nearby. Note that MgO crystallites of cubic shape were employed in the experiments of Cowley (1982a, b) and Marks (1982). For more comments see text.

limit in the photon spectrum of $\hbar\omega = \hbar\pi v/L$. For a cube of size $L = 1000$ Å this lower limit is 3.4 eV in the case of 100 keV electrons and 2.9 eV in the case of 80 keV electrons. Using equation (15) for the case of a 100 keV electron beam aligned parallel to and 20 Å outside the face of an MgO cube of side $L = 1000$ Å, we can then estimate that one Cherenkov photon will emerge from the lower surface for about 250 electrons passing. With the typical beam currents available this could correspond to a photon yield of over 10^6 per second.

In many cases these Cherenkov photons will be superimposed on a potentially much higher background of radiation arising from the decay of electronic excitations. Direct radiative decay of surface and bulk plasmons is not forbidden in small particles but would give photons with energies in the range $E_g = 7.5 \text{ eV} \leqslant \hbar\omega \leqslant 24$ eV in the case of MgO. Plasmon decay via electron–hole pairs could however ultimately produce recombination radiation photons with energies of E_g, or even less if there are defect states in the gap.

It therefore appears that it may well be possible to detect Cherenkov photons generated in small MgO particles by relativistic electron beams which pass near the crystal. Indeed the problem of recombination radiation may be less serious with such external beams than with electron beams passing through the particle. For an unambiguous identification of Cherenkov photons it will, however, be necessary to employ clean defect-free material and to confirm that the signal disappears at beam energies below the Cherenkov threshold.

4. Analysis

Detailed analysis of the expressions derived previously is only possible when the dielectric function appropriate to a given condensed medium is specified. Here we shall refer to the interaction of electron beams with MgO cubes, a system for which recent experimental data on electron energy losses exist (Marks 1982, Cowley 1982a, b). The complex dielectric function of MgO has been measured experimentally (Roessler and Walker 1967), and calculated theoretically (Fong *et al* 1968). The experimental curves for $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ are shown in figure 2. In the experiments by Marks (1982) and Cowley (1982a, b), electron energies of about 100 keV were employed. The excitation function was measured for various beam positions with respect to the surface of the wedge, within distances up to 100 Å on either side of the surface. The condensed media consisted of small crystallites of cubic symmetry.

Figure 4 shows the ratio between the non-retarded and the retarded excitation probabilities for 100 keV beams interacting externally to the medium and at different impact parameters with respect to its surface. It is seen that the retarded probability is always larger than the non-retarded prediction, except for beams close to the surface and except in frequency ranges where ε' and ε'' are both small ($\leqslant 1$). From figures 2 and 4 it follows that the larger the values of $\varepsilon'(\omega)$, the larger the retarded probability is in comparison with the non-retarded predictions. The same result holds for large impact parameters, obviously, since the classical expression neglects retardation effects and these will be appreciable at large beam–surface distances. In addition, a lot of structure appears in figure 4 below the surface plasmon energy ($\hbar\omega \sim 16$ eV), and little structure is seen above the bulk plasmon energy ($\hbar\omega \sim 22$ eV). Similar results were found when a simple dielectric function of the Drude type is taken for the calculations. In all cases, the effect of varying the distance z_0 from the beam to the target is larger than a variation in the beam velocity.

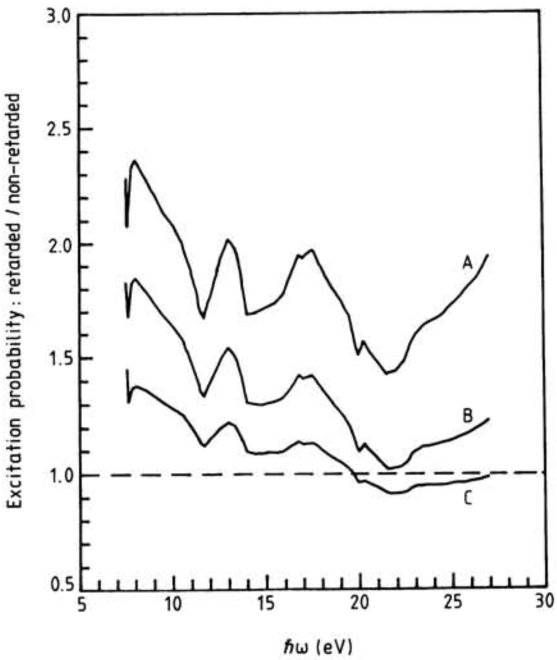

Figure 4. Ratio between the excitation probability for a relativistic beam, equation (20), and a non-relativistic beam, equation (19), travelling externally to a given solid surface for a 100 keV electron beam. The distances from the beam to the surface for the various curves were: A, 100 Å; B, 40 Å; C, 5 Å. The dielectric material is MgO.

Figure 5 shows the retarded excitation probability for a 80 keV beam and for a 1000 keV beam interacting with MgO at a distance $z_0 = 20$ Å above the surface of the cube (a typical value in the experiments reported by Marks (1982) and Cowley (1982a, b)). The excitation probability decreases with increasing beam velocity according to equation (19), and the overall structure tends to smooth out.

A similar comparison between the retarded and non-retarded results for a beam internal to the medium, equations (22) and (24), could be conducted. Since the presence of the cut-off wavenumber k_c in equation (25) introduces an element of ambiguity and the predictions are sensitive to the choice of k_c, we shall not proceed along these lines. A detailed comparison of theoretical and experimental loss spectra is not attempted. Most of the recent data for excitation by non-penetrating beams refers to MgO, a material for which the dielectric constant is not too large. For this material the most pronounced effects of retardation appear below about 10 eV, where the loss spectrum is rather weak so that the zero of the intensity scale must be known rather accurately in order to do proper comparisons with theory. It is worthwhile to point out, here, the basic facts. In order to obtain an appreciable retardation effect one requires $\varepsilon' > c^2/v^2$ and ε'' to be small. These conditions tend to hold in the region $0 < \hbar\omega < E_g$, where E_g is the average energy gap of the semiconductor or insulator. However, the following approximate relation holds, $\varepsilon'(0) = 1 + (\hbar\omega_p/E_g)^2$, where $\omega_p = (4\pi ne^2/m)^{1/2}$ is the plasma frequency and n is the valence electron density. Thus, a large value of ε' means a small gap and therefore the retardation effect, though large, occurs at very small losses requiring great precision in the spectra and careful subtraction of the zero-loss peak

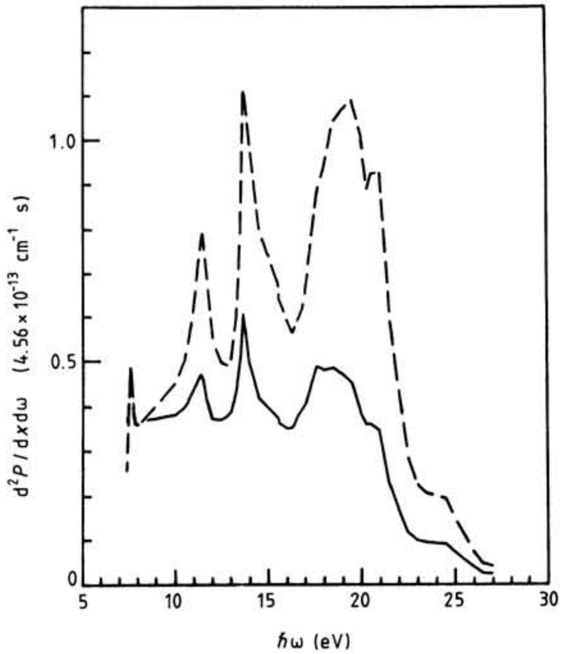

Figure 5. Excitation probability for a relativistic beam travelling externally to a solid and at a distance of 20 Å, from equation (20). The energies of the beams were: broken curve, 80 keV; full curve, 1000 keV. In the experiment reported by Marks (1982) the beam energy is 80 keV. The dielectric material is MgO.

(von Festenberg 1969). Alternatively as in MgO, the gap is relatively big but ε' is then not so large and the whole effect is not so easy to detect. In conclusion, relativistic effects in the interaction of charged particles at fixed impact parameter with respect to the surface of a condensed medium, may be appreciable even for the energies of current interest in STEM machines, about 100 keV, and use of non-relativistic expressions for the excitation probability or specific energy loss may not be sufficient in order to understand the experimental data.

In particular, we note that the strongest effects occur in insulators and semiconductors at energies below the band gap where $\mathrm{Im}(\varepsilon)$ is extremely small but $\mathrm{Re}(\varepsilon)$ can be large. Therefore, in the energy-loss experiments in this region the retarded theory must be used, otherwise data could be erroneously interpreted to suggest the presence of electron states in the gap. Note also that the relativistic effect will give a background contribution coming from all parts of the surrounding crystal against which the localised defect state contribution has to be detected and analysed. On the other hand, let us stress that although the ratios plotted in figure 4 show that the effect of retardation is a large one, the experimental detection may be difficult for the reasons discussed in §§ 3 and 4.

The analysis developed in this paper has been applied to the data on loss spectra generated by Wheatley *et al* (1983). It turns out that the relativistic theory does not explain the large difference between internal and external beam loss spectra that was observed (Wheatley *et al* 1983) in the case of Al_2O_3. It is now reported (Wheatley, private communication) that the large peak at 15 eV is due to the conversion of Al_2O_3 into Al under the very high current density in the electron beam in the STEM.

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