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Anomalous electron energy loss in small spheres

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Abstract. In analogy to the case of optical absorption it is shown that inclusion of non-local surface effects results in a very large contribution to the energy loss of fast electrons scattered by a sphere. The scattering is found to be predominantly in the forward direction and is an order of magnitude larger than that calculated classically.

1. Introduction

Within the last two years experiments by Schmidt-Ott and Siegman (1980) and theory by Penn and Rendell (1981, 1982) and by Apell and Ljungbert (1982a, 1983) have been concerned with the anomalously large optical absorption and photoyields exhibited by small metal spheres. The theories demonstrate that the anomolous absorption is directly related to the surface photoeffect which produces enhanced excitation of electron–hole pairs in the spheres. It is of interest to investigate the possibility that a similar enhancement can occur in the energy loss of fast electrons through small spheres. Because the dominant electron scattering is found to be in the forward direction there is a strong analogy with photon absorption where only small momentum transfers to the sphere can occur. We therefore begin by reviewing the role of the surface in photoabsorption and then show how the relevant theory can be applied to the case of electron energy loss by generalising the method developed by Apell and Ljungbert (1982a) to treat photoabsorption.

The understanding of the interaction between light and a metal surface has increased considerably during recent years (Kliewer 1980, Feibelman 1982, Mukhopadhyay and Lundqvist 1978). The presence of a surface has a profound influence on the behaviour of the total electromagnetic field induced in the solid. First of all it breaks the translational invariance of the solid and therefore provides a region where the induced field can have a large spatial variation compared with the wavelength of the incident radiation. Secondly the surface is the region in between which the dielectric properties go from their vacuum to their bulk values. The treatment of these spatial aspects constitutes what is known as non-local optics as compared with the standard local treatment of a sharp vacuum–metal interface in classical optics. The first aspect of non-locality has been treated extensively within the semiclassical infinite-barrier model (Kliewer 1980, Mukhopadhyay and Lundqvist 1978, Apell 1978, 1981a) which has a sharp vacuum–

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metal interface as in the case of the classical treatment but the metal is described with a dielectric function containing information about single-particle excitations, thus being superior to for instance the Drude dielectric function used in the classical treatment which only incorporates the dispersionless bulk plasmon. The main effect of including electron–hole pair excitations in the semiclassical infinite-barrier model is to allow the classical singular surface charge to relax spatially into the metal. The qualitative aspects of this model are rather well understood. However, the model fails in its quantitative predictions (Kliewer 1980, Feibelman 1982, Apell 1982) basically because it does not contain the second aspect of non-locality: the extension of the interface into the vacuum region due to the spill-out of the metal electrons. In our view this is essential to a quantitative treatment of non-local optics.

The effects on the electromagnetic field of this continuous transition in the dielectric properties have been illustrated, primarily numerically, by Feibelman (1975). Even though it is a formidable task to gain additional insight into this aspect without having to resort to heavy numerical calculations it is actually possible, in a restricted sense, to accomplish this for a realistic metal surface using a procedure developed by Apell (1981b) based on ideas presented by Flores and Garcia-Moliner (1972)—where the two Maxwell equations which require continuity of the field components parallel to the surface ($E_\parallel$ and $H_\parallel$) are integrated over the surface region in a pillbox manner. We ask what the changes in the Fresnel reflection coefficients are when there is no longer a sharp interface between vacuum and metal. For incident light with the field vector parallel to the surface (s-polarised) there is found to be no significant change since the field is already continuous at a sharp interface. However, for p-polarised light which has a discontinuous field component perpendicular to the surface in the classical treatment we find a non-negligible correction to the Fresnel laws. Since the non-local treatment allows for the surface charge to relax spatially the correction to the Fresnel equations enters naturally as the first moment of the induced charge density, denoted by $d_\perp$ (Feibelman 1976). This is *the* length of non-local optics and is typically of the order of ångströms. With reasonable estimates for $d_\perp$ a good account has been achieved for various physical situations pertinent to surface physics such as the photoyield from aluminium (surface photoelectric effect) (Apell 1982), damping of vibrationally excited CO on copper (Apell 1983) as well as the dispersion and damping of the surface plasmon on Al (Ahlqvist and Apell 1982). The quantitative comparison of $d_\perp$ with the experimental findings reveals several aspects of importance for a proper treatment of the non-local response of a metal (surface). It is necessary to use a profile rather than a sharp interface, at least for energies below the bulk plasmon frequency, even if electron–hole pair excitations are included in the latter (Kliewer 1980, Apell 1982). Not surprisingly a *self-consistent* potential is superior to other types such as the sharp interface treated by Kliewer (1980); the difference can amount to an order of magnitude in the predicted absorption. This is related to the fact that the sharp interface imposes a severe restriction on the induced density in that it cannot relax outside of the sharp barrier.

The non-local treatment as presented by Apell (1981b) has later been applied to photon absorption in a metal sphere (Apell and Ljungbert 1982a). A general framework was developed there in which earlier non-local theories were shown to be in essence different ways of calculating the centre of gravity of the induced charge (denoted d_r for a sphere). The reason for studying a sphere for non-local effects was that in this case light can couple to the surface plasmon resonance which is not possible at a planar surface because of a mismatch in momentum. This coupling enables one to obtain a larger field strength in the medium and thus the electron–hole pair production is expected

to increase (Penn and Rendell 1981, 1982). The non-local theory developed for the sphere has further been successfully applied to explain the observed red shift of the surface plasmon resonance compared with the Mie prediction, Apell and Ljungbert (1982b) whereas earlier theories based on sharp surfaces by necessity gave a blue shift.

In view of those findings we felt the need to extend the treatment, so far limited to incident photons, to describe the interaction between probing electrons and spherical particles. It may be that electron scattering from spherical particles can provide a surface-sensitive tool because previous studies have been based on theories which did not contain electron–hole pair excitations (Fujimoto and Komaki 1968).

The present theory involves screening the fast electron by the *bulk* dielectric function and including the effects of the sphere surface as a correction factor to the classical formulation. This is crucial because of the difficulties in calculating a momentum- and frequency-dependent dielectric function for a sphere when the surface is taken into account. It should be noted that the semiclassical infinite-barrier model also makes use of the bulk dielectric function but as mentioned above the model gives an absorption coefficient that is at least an order of magnitude too small for frequencies below the bulk plasmon frequency.

The only previous calculations of the energy loss of fast electrons incident on a sphere were carried out by Fujimoto and Komaki (1968) in the hydrodynamic approximation and by Lushnikov and Simonov (1975) who obtained similar results. We will show that the inclusion of non-local surface effects increases the rate of energy loss by over an order of magnitude. In § 2 we present the theory for the energy loss, and numerical results are presented and compared with those of Fujimoto and Komaki (1968).

2. Theory of the electron energy loss

In this section we generalise the work of Fujimoto and Komaki (1968) to include the excitations of electron–hole pairs near the surface of the sphere. A fast electron moving with velocity v in the z direction moves in a path given by $\boldsymbol{r}_0 = (\boldsymbol{\rho}_0, vt)$ where the centre of sphere is at the origin. Consequently $|\boldsymbol{\rho}_0|$ is the distance of closest approach between the electron and sphere. The charge density of the electron is

$$\boldsymbol{\rho}_0(\boldsymbol{r}, t) = e\delta(\boldsymbol{\rho} - \boldsymbol{\rho}_0)\,\delta(z - vt) \tag{1a}$$

and its Fourier transform is

$$\rho_0(\boldsymbol{q}, \omega) = (2\pi e/v)\exp(-\mathrm{i}\boldsymbol{q}_\parallel \cdot \boldsymbol{\rho}_0)\,\delta(q_z + \omega/v). \tag{1b}$$

The charge creates a potential

$$\varphi^{(i)}(\boldsymbol{r}, \omega) = \sum_q (4\pi/q^2)\exp(\mathrm{i}\boldsymbol{q} \cdot \boldsymbol{r})\,\rho^{(i)}(q, \omega) \qquad i = \text{in or ex} \tag{2}$$

where outside the sphere $\rho^{(\mathrm{ex})}(q, \omega) = \rho_0(q, \omega)$ and inside the sphere the charge is screened so that $\rho^{(\mathrm{in})}(q, \omega) = \rho_0(q, \omega)/\varepsilon(q, \omega)$ where $\varepsilon(q, \omega)$ is the *bulk* momentum-dependent dielectric function. Thus surface effects are not included in $\rho^{(\mathrm{in})}$. The potential due to the charge is given by using (1) and (2);

$$\varphi^{(i)}(\boldsymbol{r}, \omega) = \int \frac{\mathrm{d}^2 q_\parallel}{(2\pi)^2} F^{(i)}(\boldsymbol{q}_\parallel, \omega)\exp[\mathrm{i}\boldsymbol{q}_\parallel \cdot (\boldsymbol{\rho} - \boldsymbol{\rho}_0)]\exp(-\mathrm{i}\omega z/v) \tag{3a}$$

where

$$F^{(i)}(\boldsymbol{q}_\parallel, \omega, \boldsymbol{r}; \boldsymbol{\rho}_0) = \frac{4\pi e}{v} \frac{1}{Q^2 \varepsilon^{(i)}(Q, \omega)} \tag{3b}$$

$$\boldsymbol{Q} = (\boldsymbol{q}_\parallel, -\omega/v) \tag{3c}$$

$$\varepsilon^{(i)}(q, \omega) = \varepsilon(q, \omega) \qquad i = \text{in} \tag{3d}$$
$$= 1 \qquad i = \text{ex.}$$

Equation (3*a*) can also be written as

$$\varphi^{(i)}(\boldsymbol{r}, \omega) \equiv \sum_{lm} \varphi^{(i)}_{lm}(\boldsymbol{r}, \omega) Y_{lm}(\hat{\Omega}_r) \tag{4a}$$

where

$$\varphi^{(i)}_{lm}(r, \omega) = 4\pi i^l \int \frac{\mathrm{d}^2 q_\parallel}{(2\pi)^2} F^{(i)}(q_\parallel, \omega) \exp(-i\boldsymbol{q}_\parallel \cdot \boldsymbol{\rho}_0) j_l(Qr) Y^*_{lm}(\hat{\Omega}_Q). \tag{4b}$$

In addition to the 'direct' potential created by the fast electron the potential due to the surface response of the sphere must be considered. Near the sphere surface we do not know the potential; it can only be obtained if we know the dielectric response of the sphere including the surface response. We assume that there is a region outside of which the effects of the surface are damped out and the potential takes on an asymptotic form. In the case of a plane surface Feibelman (1975, 1982) has found that the asymptotic region begins roughly 10 Å from the surface and we assume that the case of a sphere is not significantly different. In this region the potential must have the form

$$\varphi^<(\boldsymbol{r}, \omega) = \varphi^{(\text{in})}(\boldsymbol{r}, \omega) + \sum_{lm} \beta_{lm} \left(\frac{r}{R}\right)^l Y_{lm}(\hat{\Omega}_r) \qquad r < r_1 \tag{5a}$$

$$\varphi^>(\boldsymbol{r}, \omega) = \varphi^{(\text{ex})}(\boldsymbol{r}, \omega) + \sum_{lm} \gamma_{lm} \left(\frac{R}{r}\right)^{l+1} Y_{lm}(\hat{\Omega}_r) \qquad r > r_2 \tag{5b}$$

where $r_1 < R < r_2$ and R is the sphere radius.

These potentials satisfy $\nabla^2 \varphi^> = -4\pi\rho_0^{(\text{ex})}$ and $\nabla^2 \varphi^< = -4\pi\rho_0^{(\text{in})}$ and the terms involving the βs and γs exhibit the correct behaviour at $r = 0$ and $r = \infty$ respectively. While the above potentials are physically correct only for $r > r_2$ or $r_1 > r$ they can of course be continued into the regions $r_2 > r > R$ and $R > r > r_1$ and it will be convenient to do so. Define $\varphi(\boldsymbol{r}, \omega)$ to be the potential given by (5*b*) but with the condition $r > r_2$ replaced by $r > R$ and define $\tilde{\varphi}^<(\boldsymbol{r}, \omega)$ to be the potential given by (5*a*) but with the condition $r < r_1$ replaced by $r < R$. Denote the *true* potential in the region $r_1 < r < R$ by $\varphi^<(\boldsymbol{r}, \omega)$ and the *true* potential for $r_2 > r > R$ by $\varphi^>(\boldsymbol{r}, \omega)$ where $\varphi^{\gtrless}$ are explicitly given by equation (5) for $r > r_2$ and $r < r_1$. Thus φ and $\tilde{\varphi}$ differ only in the region $r_1 < r < r_2$ and $\tilde{\varphi}$ has the form of the potential one uses in the classical treatment. If

$$\varphi(\boldsymbol{r}) = \sum_{lm} \varphi_{lm}(r) Y_{lm}(\hat{\Omega}_r)$$

then

$$E_r(\boldsymbol{r}) = -\frac{\partial \varphi(\boldsymbol{r})}{\partial r} = \sum_{lm} \left(-\frac{\partial \varphi_{lm}(r)}{\partial r}\right) Y_{lm}(\hat{\Omega}_r) \equiv \sum_{lm} E_r^{(lm)}(r) Y_{lm}(\hat{\Omega}_r) . \tag{6}$$

It is useful to define a quantity

$$\Delta E_r^{(lm)}(r) = (E_r^{(lm)}(r) - \tilde{E}_r^{(lm)}(r))/(\tilde{E}_r^{(lm)}(R^+) - \tilde{E}_r^{(lm)}(R^-)) \tag{7}$$

where $R^{\pm} = R \pm \delta$ as $\delta \to 0$ and where $E_r^{(lm)}(r)$ is the true field derived from $\varphi^{\gtrless}$ and $\bar{E}^{(lm)}(r)$ is the field defined from $\bar{\varphi}$ defined above. We define d_r which is fundamental to the present approach

$$d_r^{(lm)} \equiv \int_{r_1}^{r_2} \mathrm{d}r\, \Delta E_r^{(lm)}(r). \tag{8}$$

It follows immediately (see Appendix 1) that

$$d_r^{(lm)} = (\bar{\varphi}_{lm}^{<}(R) - \bar{\varphi}_{lm}^{>}(R))\left(\frac{\partial}{\partial r}\bar{\varphi}_{lm}^{<}(R) - \frac{\partial}{\partial r}\bar{\varphi}_{lm}^{>}(R)\right)^{-1}. \tag{9}$$

From the definition of $\bar{\varphi}$ and use of equation (5) one has

$$\beta_{lm}\left(1 - \frac{l}{R}d_r^{(lm)}\right) - \gamma_{lm}\left(1 + (l+1)\frac{d_r^{(lm)}}{R}\right) = \left(1 - d_r^{(lm)}\frac{\partial}{\partial r}\right)(\varphi_{lm}^{(\mathrm{ex})}(R) - \varphi_{lm}^{(\mathrm{in})}(R)). \tag{10}$$

In order to determine the coefficients β and γ we use $\nabla \cdot \boldsymbol{D} = 0$, i.e., the radial component of $\boldsymbol{D}$ must be continuous. We will assume that $\boldsymbol{D}$ can be approximated adequately by $\boldsymbol{D} = \varepsilon \bar{\boldsymbol{E}}$ where $\bar{\boldsymbol{E}} = -\nabla\bar{\varphi}$. This is a reasonable assumption because $\boldsymbol{D}$ is a continuous function in the classical treatment and the use of $\bar{\varphi}$ is equivalent to the classical approximation. From equation (5) we have $\boldsymbol{D} = -\nabla\varphi_D$ where

$$\varphi_D(\boldsymbol{r}, \omega) = \varphi^{(\mathrm{ex})}(\boldsymbol{r}, \omega) + \varepsilon \sum_{lm} \beta_{lm}\left(\frac{r}{R}\right)^{l} Y_{lm}(\hat{\Omega}_r) \qquad r < R \tag{11a}$$

$$= \varphi^{(\mathrm{ex})}(\boldsymbol{r}, \omega) + \sum_{lm} \gamma_{lm}\left(\frac{R}{r}\right)^{l+1} Y_{lm}(\hat{\Omega}_r) \qquad r > R \tag{11b}$$

where

$$\varepsilon = \varepsilon(0, \omega). \tag{11c}$$

Note that this expression for φ_D yields $\nabla \cdot \boldsymbol{D} = -4\pi\rho_0$ as required. Continuity of D_r now gives

$$\beta_{lm}\varepsilon l + \gamma_{lm}(l+1) = 0. \tag{12}$$

Equations (10) and (12) yield

$$\beta_{lm} = -(l+1)\Gamma_{lm}/\Delta_{lm} \tag{13a}$$

$$\gamma_{lm} = \varepsilon l\Gamma_{lm}/\Delta_{lm} \tag{13b}$$

where

$$\Delta_{lm} = \varepsilon l + (l+1)[1 + lR^{-1}d_r^{(lm)}(\varepsilon - 1)] \tag{13c}$$

$$\Gamma_{lm} = \left(d_r^{(lm)}\frac{\partial}{\partial r} - 1\right)(\varphi_{lm}^{(\mathrm{ex})}(R) - \varphi_{lm}^{(\mathrm{in})}(R)). \tag{13d}$$

The effects of the sphere surface are in $d_r^{(lm)}$. If $d_r^{(lm)} = 0$ one obtains the classical solution. It is shown in Appendix 2 that $d_r^{(lm)}$ is directly related to the surface charge density induced by the fast electron

$$d_r^{(lm)} = R\int_{r_1}^{r_2} \mathrm{d}r\, r(R - r)\,\delta\rho^{(lm)}\left(\int_{r_1}^{r_2} \mathrm{d}r\, r^2\,\delta\rho^{(lm)}\right)^{-1} \tag{14a}$$

where

$$\delta\rho = \sum_{lm} \delta\rho^{(lm)} Y_{l,m}(\hat{\Omega}_r) . \tag{14b}$$

Here $\delta\rho$ is the difference between the charge induced by the fast electron and the charge induced if the sphere were described by the bulk dielectric function. The evaluation of $d_r^{(lm)}(\omega)$ will be discussed later in the paper. Because $\delta\rho$ represents a charge induced in the sphere it will have the symmetry of the sphere and can be expressed as

$$\delta\rho(\boldsymbol{r}) = \sum_{lm} \rho_l(r) Y_{lm}(\hat{\Omega}_r)$$

so that d_r is a function of l but not m. This point is discussed in Appendix 2. Consequently Δ_{lm} of equation (13*c*) is also a function of l but not of m.

The work done by the fast electron with trajectory $\boldsymbol{r} = (\boldsymbol{\rho}_0, vt)$ is shown by Fujimoto and Komaki (1968) to be

$$W(\boldsymbol{\rho}_0) = e \int_{-\infty}^{\infty} \mathrm{d}t\, v \Delta E_z(\boldsymbol{r}_0, t) \tag{15}$$

where $\Delta E_z(\boldsymbol{r}_0, t)$ is the z component of electric field *induced* by the particle, i.e., $\Delta \boldsymbol{E}$ is the difference between the field in the presence of the sphere and the field in the absence of the sphere. Using $\Delta E_z = -\partial \Delta\varphi/\partial z$ and writing $\Delta\varphi(r, t)$ in terms of its Fourier transform yields

$$W(\boldsymbol{\rho}_0) = \mathrm{i}\frac{e}{2\pi v} \int \frac{\mathrm{d}^2 q_\parallel}{(2\pi)^2} \int_{-\infty}^{\infty} \mathrm{d}\omega\, \omega \exp(\mathrm{i}\boldsymbol{q}_\parallel \cdot \boldsymbol{\rho}_0) \Delta\varphi(Q, \omega; \rho_0) \tag{16}$$

where Q is given by (3*c*). We require

$$W = \int \mathrm{d}^2\rho_0 \,\mathrm{Re}\{W(\boldsymbol{\rho}_0)\} \equiv \int \mathrm{d}^2 q_\parallel \int_0^{\infty} \mathrm{d}\omega\, \hbar\omega P(\boldsymbol{q}_\parallel, \omega) \tag{17}$$

where $P(\boldsymbol{q}_\parallel, \omega)$ is the probability the fast electron loses energy ω and momentum $\boldsymbol{q}_\parallel$ in the scattering from the sphere. Use of (16) in (17) yields

$$P(q_\parallel, \omega) = -\mathrm{Im}\left(\frac{2e}{\hbar v} \frac{1}{(2\pi)^3} \int \mathrm{d}^2\rho_0 \exp(\mathrm{i}\boldsymbol{q}_\parallel \cdot \boldsymbol{\rho}_0) \Delta\varphi(Q, \omega)\right) \tag{18}$$

where $\Delta\varphi$ is the potential created by the fast electron minus the potential it creates in vacuum.

From equation (5) we have

$$\Delta\varphi^{<}(\boldsymbol{r}) = \sum_{lm} \left[\Delta\varphi_{lm}^{(\mathrm{in})}(r) + \beta_{lm}\left(\frac{r}{R}\right)^l\right] Y_{lm}(\hat{\Omega}_r) \tag{19a}$$

$$\Delta\varphi^{>}(\boldsymbol{r}) = \sum_{lm} \gamma_{lm}\left(\frac{R}{r}\right)^{l+1} Y_{lm}(\hat{\Omega}_r) \tag{19b}$$

where

$$\Delta\varphi_{lm}^{(\mathrm{in})}(r) = (4\pi)^2 \frac{e}{v} \mathrm{i}^l \int \frac{\mathrm{d}^2 q_\parallel}{(2\pi)} \frac{j_l(Qr)}{Q^2} Y_{lm}^*(\hat{\Omega}_Q) \left(\frac{1}{\varepsilon(Q, \omega)} - 1\right) \exp(-\mathrm{i}\boldsymbol{q}_\parallel \cdot \boldsymbol{\rho}_0). \tag{19c}$$

The quantities β_{lm} and γ_{lm} are given by equations (13*a*) and (13*b*) respectively and we note that equation (13*d*) can be written as

$$\Gamma_{lm} = \left(1 - d_r^{(l)} \frac{\partial}{\partial r}\right) \Delta \varphi_{lm}^{(\mathrm{in})}(R). \tag{20}$$

It is shown in Appendix 3 that use of equations (19) and (18) yields

$$P(\boldsymbol{q}_\parallel, \omega) = \frac{4}{\pi} \frac{e^2}{\hbar v^2} \left[\frac{1}{3} \frac{R^3}{Q^2} \mathrm{Im}\left[\frac{1}{\varepsilon(Q, \omega)} - 3 \sum_l (2l+1) (j_l'(QR))^2 \frac{d_r^{(l)}}{R} \left(\frac{1}{\varepsilon(Q, \omega)} - 1 \right) \right] \right.$$
$$- \frac{R}{Q^4} \sum_l l(l+1)(2l+1)$$
$$\left. \times \mathrm{Im}\left\{ \left[\left(1 - d_r^{(l)} \frac{\partial}{\partial r}\right) j_l(QR) \right]^2 \left(\frac{1}{\varepsilon(Q, \omega)} \right) \frac{(1-\varepsilon)}{\Delta_l} \right\} \right]. \tag{21}$$

In the classical case $d_r^{(l)} = 0$ in equation (21) while in the work of Fujimoto and Komaki (1968) $P = 0$ where P is a measure of the plasmon dispersion. It is readily verified that their results agree with equation (21) in this limit.

Following Apell and Ljungbert (1982b) we assume R is large so that $d_r^{(l)}$ may be replaced by $d_\perp$, the response of a flat surface discussed by Feibelman (1976) and Apell (1981b). This approximation follows from the fact that $d_r^{(l)}$ is a function of $l/(Rk_F)$ where k_F is of the order of the inverse Fermi screening length and only small ls are important in equation (21) because the factors Q^{-2} and Q^{-4} favour forward-scattering, i.e., $QR \ll 1$. Because we use $d_r^{(l)} \sim d_\perp$ we limit ourselves to spheres such that $R \geqslant 20$ Å. The first sum over l in equation (21) can now be carried out using

$$\sum_l (2l+1) (j_l'(QR))^2 = \tfrac{1}{3} \tag{22}$$

to obtain

$$P(\boldsymbol{q}_\parallel, \omega) \simeq \frac{4}{\pi} \frac{e^2}{\hbar v^2} \left(\frac{1}{3} \frac{R^3}{Q^2} \mathrm{Im} \frac{1}{\tilde{\varepsilon}(Q, \omega)} \right) - \frac{R}{Q^4} \sum_l l(l+1)(2l+1)$$
$$\times \mathrm{Im}\left\{ \left(\frac{1}{\varepsilon(Q, \omega)} - 1 \right) \frac{1-\varepsilon}{\Delta_l} \left[\left(1 - d_\perp \frac{\partial}{\partial r} j(QR)\right) \right]^2 \right\} \tag{23a}$$

where

$$\tilde{\varepsilon}(Q, \omega)^{-1} = \varepsilon(Q, \omega)^{-1} - (d_\perp/R)(\varepsilon(Q, \omega)^{-1} - 1) \tag{23b}$$

which is a direct generalisation of the effective dielectric function found by Apell and Ljungbert (1982a) where $\varepsilon(Q, \omega)$ in (23*b*) is replaced by $\varepsilon(0, \omega)$. This is justified by the fact that small-Q scattering dominates in (23*a*). This implies small $q_\parallel$ and $\omega/v \ll 1$. The first term, $\mathrm{Im}(1/\tilde{\varepsilon})$, is the usual energy-loss function and the second term on the right-hand side of (23*a*) which includes energy loss to surface excitations as described by Δ_l. The zeros of Δ_l give the surface plasmon dispersion in agreement with Apell and Ljungbert (1982a). In addition to entering the expression for the surface plasmon dispersion and damping $d_r^{(l)}$ determines the effective radius of the sphere since

$$\left(1 - d_r^{(l)} \frac{\partial}{\partial r}\right) j_l(QR) \simeq j_l(Q(R - d_r^{(l)})).$$

The quantity $d_r^{(l)}$ measures the position of the induced charge relative to the surface of the sphere as shown in equation (14). In the classical treatment the induced charge is at the surface of the sphere (and $d_r^{(l)} = 0$). In fact the induced charge is shifted to $(R - d_r^{(l)})$ and consequently the effective size of the sphere is changed.

It is clear from equation (21) that the dominant scattering occurs for small $q_\parallel$, i.e., for small $QR = [q_\parallel^2 + (\omega/v)^2]^{1/2} R$. This is because of the leading factors of Q^{-2} and Q^{-4} in the bulk and surface terms. Following Fukimoto and Komaki (1968) we replace $\varepsilon(Q, \omega)$ by $\varepsilon(0, \omega)$ in equation (23) and use the Drude form of the dielectric function

$$\varepsilon(0, \omega) = 1 - \frac{\omega_p^2}{\omega(\omega - ig)}. \tag{24}$$

We now find from equation (23)

$$P(q_\parallel, \omega) = P_B(q_\parallel, \omega) + P_S(q_\parallel, \omega) \tag{25a}$$

$$P_B(q_\parallel, \omega) = \frac{N}{\rho^2}\left[\mathrm{Im}\left(\frac{1}{\bar{\omega}(\bar{\omega} - i\bar{g}) - 1}\right) \times \left(1 - \bar{d} - \frac{3}{\rho^2}\sum_l l(2l+1)\,\mathrm{Im}(j_l(\rho) - \bar{d}\rho j_l'(\rho))^2(1 - l\bar{d})^{-1}\right)\right] \tag{25b}$$

$$P_S(q_\parallel, \omega) = \frac{N}{\rho^4}\sum_l l(2l+1)\,\mathrm{Im}\,\frac{(j_l(\rho) - \bar{d}\rho j_l'(\rho))^2(1 - l\bar{d})^{-1}}{\bar{\omega}(\bar{\omega} - i\bar{g}) - \bar{\omega}_l^2} \tag{25c}$$

$$N = 4e^2R^5/(3\pi\hbar v^2) \tag{25d}$$

$$\rho = QR \tag{25e}$$

$$\bar{\omega} = \omega/\omega_p \tag{25f}$$

$$\bar{g} = 1/\omega_p\tau \tag{25g}$$

$$\bar{d} = d_-/R \tag{25h}$$

$$\bar{\omega}_l^2 = [l/(2l+1)][1 + (l+1)\bar{d})]. \tag{25i}$$

In equation (23) the separation into bulk and surface terms is on the basis of the denominators $\bar{\omega}(\bar{\omega} - i\bar{g}) - 1$ and $\bar{\omega}(\bar{\omega} - i\bar{g}) - \bar{\omega}_l^2$ the zeros of which describe bulk and surface plasmons respectively.

The scattering is largest for small ρ and it is useful to expand equation (25) in powers of ρ^{-2}. One finds that P_B is of the order of ρ^0 while

$$P_S(q_\parallel, \omega) \simeq -\frac{4e^2}{2\pi\hbar v^2}\frac{R^3}{Q^2}\,\mathrm{Im}\,\alpha(\omega) \tag{26a}$$

where

$$\alpha(\omega) = \frac{\tilde{\varepsilon}(0, \omega) - 1}{\tilde{\varepsilon}(0, \omega) + 2} \tag{26b}$$

and $\tilde{\varepsilon}(0, \omega)$ is given by equation (23*b*). We note that the probability of energy loss is the product of the Coulomb interaction, $4\pi e^2/Q^2$, the volume of the sphere and the optical polarisability of the sphere in the limit of small Q. The imaginary part of α now contains a term proportional to $\bar{d} = d_\perp/R$ due to electron–hole excitation which results in a term in P_S proportional to R^2 which is the expected result for surface scattering.

In order to compare the non-local scattering probability with the classical one given by $\tilde{d} = 0$ we assume $\tilde{g} \ll |\operatorname{Im} \tilde{d}|$ and $|\operatorname{Re} \tilde{d}| \ll 1$ in equation (25). Taking the small-Q limit yields

$$P_S(q_\parallel, \omega) \simeq \tfrac{1}{3}(N/\rho^2) \frac{(1 - \tilde{\omega}^2) \operatorname{Im} \tilde{d}}{(\tilde{\omega}^2 - \frac{1}{3})^2 + (\frac{2}{3} \operatorname{Im} \tilde{d})^2} \tag{27a}$$

while for $\tilde{d} = 0$ question (25) gives the classical result

$$P_S^C(q_\parallel, \omega) = \tfrac{1}{3}(N/\rho^2) \frac{\tilde{g}\tilde{\omega}}{(\tilde{\omega}^2 - \frac{1}{3})^2 + (\tilde{\omega}\tilde{g})^2}. \tag{27b}$$

It will be seen that $\operatorname{Im} \tilde{d} \propto \tilde{\omega}$ so that the non-local expression (27*a*) and the classical one (27*b*) are very similar but with different effective electron lifetimes. The vanishing of P_S at $\tilde{\omega} = 1$ as indicated by equation (27*a*) is a characteristic of the non-local treatment.

In order to evaluate $P(q_\parallel, \omega)$ from equation (25) we require knowledge of $\tilde{d} = d_\perp/R$ where $d_\perp(\omega)$ is a response function with real and imaginary parts. It has been shown by Apell (1981b) that

$$d_\perp = \frac{1}{1 - (1/\varepsilon)} \int_{z_1}^{z_2} dz \left(\int dz' [\varepsilon^{-1}(z, z', \omega) - (\theta(-z) + \varepsilon^{-1}\theta(z))] \right) \tag{28}$$

where ε^{-1} is the true inverse dielectric function for a solid with a plane surface located in the region $z > 0$ and $z_1 < 0 < z_2$ and z_1, z_2 are such that $z < z_1$ and $z_2 < z$ describes the region in which the fields have reached their asymptotic values. Assuming (Apell 1981b) that

$$\varepsilon^{-1}(z, z', \omega) = 1/\varepsilon(z, \omega)\, \delta(z - z') \tag{29a}$$

where

$$\varepsilon(z, \omega) = 1 - (\omega_p^2/\omega^2) n_0(z) \tag{29b}$$

and where $n_0(z)$ is the electron ground-state density of the solid relative to the bulk value at $z = \infty$. This approximation for $\varepsilon^{-1}(z, z', \omega)$ has the virtue of being simple and of fulfilling the f-sum rule. It corresponds to placing all the oscillator strength in a local plasmon of frequency $\omega^2 = \omega_p^2 n_0(z)$. While this gives a rather poor approximation to the spatial variation of the induced density it accounts reasonably well for the frequency dependence of $d_\perp$ in equation (29*c*). It has been used with some success in predicting the overall trend in the photoabsorption spectra of atoms (Lundqvist and Mukhopadhyay 1980). Equation (29) yields an approximate form of $d_\perp$ having the essential features of the non-local response

$$d_\perp = \int_{z_1}^{z_2} dz \frac{n_0(z)(1 - n_0(z))}{\tilde{\omega}(\tilde{\omega} - i0^+) - n_0(z)}. \tag{29c}$$

Assuming the density profile is given by (Apell 1983)

$$n_0(z) = \tanh^2 \beta z \qquad 0 < z \leqslant \infty \tag{30}$$

one finds from use of (30) in (29) and $z_1 = 0$, $z_2 = \infty$

$$\operatorname{Re} d_\perp(\omega) = \operatorname{Re} d_\perp(0) \left(1 + \frac{\tilde{\omega}}{2} \ln \left| \frac{\tilde{\omega} - 1}{\tilde{\omega} + 1} \right| \right) \tag{31a}$$

$$\operatorname{Im} d_\perp(\omega) = -\frac{\pi}{2} \operatorname{Re} d_\perp(0)\, \tilde{\omega}\theta(1 - \tilde{\omega}). \tag{31b}$$

The Re $d_\perp(0)$ can be chosen to agree with the calculations of Lang and Kohn (1973) who obtained Re $d_\perp(0) = -0.85, -0.69, -0.63$ Å for $r_s = 2, 4, 6$.

In figure 1 we have plotted the quantity P_s/N given by equation (25*c*) as a function of $\bar{\omega} = \omega/\omega_p$ for various values of $\rho = [q_{\parallel}^2 + (\omega/v)^2]^{1/2}R$ for the non-local theory (present work) and for the classical theory as obtained from equation (25*c*) by setting $d_\perp = 0$. We have chosen $\bar{g} = 1/\omega_p\tau = 10^{-3}$ and for the non-local case $\mathrm{Re}(d_\perp(0)/R) = -0.03$. For $\rho = 0.1$ only the $l = 1$ term in equation (25*c*) is important and the curves are adequately represented by equations (27*a*) and (27*b*). For $\rho = 1$ the main contribution is again from $l = 1$ while for $\rho = 4$ the terms $l = 2$ and $l = 3$ are the most important and as a result the surface plasmon peak given by $\bar{\omega}^2 = \bar{\omega}_l^2$ is shifted to higher energy. Note that the $\rho = 4$ result is roughly four orders of magnitude less than $\rho = 0.1$; the Coulomb scattering is forward.

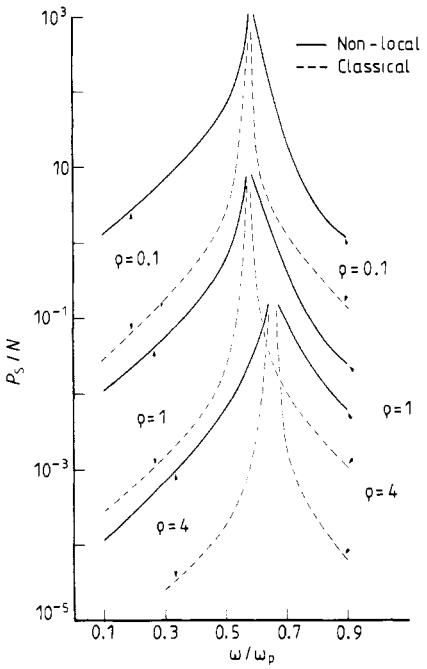

Figure 1. Surface scattering probability P_s as a function of energy loss $\hbar\omega$ for different values of $\rho = QR$. P_s is normalised to N which is defined in equation (25*e*). Small ρ implies forward-scattering. The full curves are the results of the present calculations and the broken curves refer to the classical results.

In summary we find the case of energy loss by fast electrons quite similar to that of photon absorption, particularly for small scattering angles. In both cases the excitation of electron–hole pairs near the surface of the sphere leads to very large enhancements of the scattering cross section relative to that calculated in the classical approximation which neglects such effects.

Appendix 1.

In this Appendix we derive equation (9). From equations (6), (7) and (8) one has

$$d_r^{(lm)} = \int_{r_1}^{r_2} \mathrm{d}r \left(\frac{\partial}{\partial r} \varphi_{lm}(r) - \frac{\partial}{\partial r} \tilde{\varphi}_{lm}(r) \right) \left(\frac{\partial}{\partial r} \tilde{\varphi}_{lm}(R^+) - \frac{\partial}{\partial r} \tilde{\varphi}_{lm}(R^-) \right)^{-1}. \tag{A1.1}$$

Now $\tilde{\varphi}$ is discontinuous at $r = R$ while φ is continuous so we write $\tilde{\varphi}$ in the form

$$\tilde{\varphi}_{lm}(r) = \theta(R - r)\, \tilde{\varphi}_{lm}^{<}(r) + \theta(r - R)\, \tilde{\varphi}_{lm}^{>}(r). \tag{A1.2}$$

Use of (A1.2) in (A1.1) yields

$$d_r^{(lm)} = [\varphi_{lm}(r_2) - \varphi_{lm}(r_1) - (\tilde{\varphi}_{lm}^{>}(r_2) - \tilde{\varphi}_{lm}^{>}(R)) + (\tilde{\varphi}_{lm}^{<}(r_2) - \tilde{\varphi}_{lm}^{<}(R))] \times \left(\frac{\partial}{\partial r} \tilde{\varphi}_{lm}^{>}(R) - \frac{\partial}{\partial r} \tilde{\varphi}_{lm}^{<}(R) \right)^{-1}. \tag{A1.3}$$

From the definition of $\tilde{\varphi}$; $\varphi_{lm}(r_i) = \tilde{\varphi}_{lm}^{>}(r_i)$ for $i = 1, 2$. Thus equation (9) follows directly from (A1.3).

Appendix 2.

In this appendix we derive equation (14). Our starting point is equation (9). We wish to relate $\tilde{\varphi}(R)$ and $\partial\tilde{\varphi}(R)/\partial R$ to the induced charge. The total charge is $\rho(\boldsymbol{r}) = \rho_0(\boldsymbol{r}) + \delta\rho(\boldsymbol{r})$ where ρ_0 is given by equation (1*a*) and ρ is the sum of the charge density of the fast electron, ρ_0, and $\delta\rho$, the charge induced in the system by the fast electron. Writing

$$\varphi(\boldsymbol{r}) = \sum_{lm} \varphi_{lm}(r)\, Y_{lm}(\hat{\Omega}_r) \tag{A2.1a}$$

$$\rho(\boldsymbol{r}) = \sum_{lm} \rho_{lm}(r)\, Y_{lm}(\hat{\Omega}_r) \tag{A2.1b}$$

and using

$$\nabla^2 \varphi(\boldsymbol{r}) = -4\pi\rho(\boldsymbol{r}) \tag{A2.2}$$

gives

$$\frac{1}{r^2} \frac{\partial}{\partial r} r^2 \frac{\partial}{\partial r} \varphi_{lm}(r) - l\left(\frac{l+1}{r^2} \right) \varphi_{lm}(r) = -4\pi\rho_{lm}(r) \tag{A2.3}$$

multiply by r^2 and integrate from r_1 to r_2 to obtain

$$r_2^2 \frac{\partial}{\partial r} \varphi_{lm}(r_2) - r_1^2 \frac{\partial}{\partial r} \varphi_{lm}(r_1) - l(l+1) \int_{r_1}^{r_2} \mathrm{d}r\, \varphi_{lm}(r) = -4\pi \int_{r_1}^{r_2} \mathrm{d}r\, r^2 \rho_{lm}(r). \tag{A2.4}$$

From equation (5) and the definition of $\tilde{\Phi}$:

$$\nabla^2 \tilde{\varphi}^{<} = -4\pi\rho^{(\mathrm{in})} \tag{A2.5a}$$

$$\nabla^2 \tilde{\varphi}^{>} = -4\pi\rho^{(\mathrm{ex})}. \tag{A2.5b}$$

Thus use of equation (A1.2) in (A2.2) gives

$$\left(r_2^2\frac{\partial}{\partial r}\tilde{\varphi}_{lm}^{>}(r_2) - R^2\frac{\partial}{\partial r}\tilde{\varphi}_{lm}^{>}(R)\right) - \left(r_1^2\frac{\partial}{\partial r}\tilde{\varphi}_{lm}^{<}(r_1) - R^2\frac{\partial}{\partial r}\tilde{\varphi}_{lm}^{<}(R)\right)$$
$$- l(l+1)\int_{r_1}^{r_2} \mathrm{d}r\,\tilde{\varphi}_{lm}(r)$$
$$= -4\pi\int_{r_1}^{r_2}\mathrm{d}r\,r^2(\theta(R-r)\rho_{lm}^{(\mathrm{in})} + \theta(r-R)\rho_{lm}^{(\mathrm{ex})}) \tag{A2.6}$$

Recalling that $\tilde{\varphi}_{lm}^{>}(r_2) = \varphi_{lm}^{>}(r_2)$ and $\tilde{\varphi}_{lm}(r_1) = \varphi_{lm}^{<}(r_1)$, approximating φ_l in the integral in (A2.4) by $\tilde{\varphi}_l$, and using (A2.6) in (A2.5) yields

$$R^2\left(\frac{\partial}{\partial r}\tilde{\varphi}_{lm}^{>}(R) - \frac{\partial}{\partial r}\tilde{\varphi}_{lm}^{<}(R)\right) - 4\pi\int_{r_1}^{r_2}\mathrm{d}r\,r^2(\theta(R-r)\rho_{lm}^{(\mathrm{in})} + \theta(r-R)\rho_{lm}^{(\mathrm{ex})})$$
$$= -4\pi\int_{r_1}^{r_2}\mathrm{d}r\,r^2\rho_{lm}(r) \tag{A2.7}$$

where $\rho^{(\mathrm{ex})} = \rho_0$, $\rho^{(\mathrm{in})} = (1/\varepsilon^{(\mathrm{in})})\rho_0$, and $\rho = \rho_0 + \delta\rho$. Use of these relations in equation (A2.7) gives

$$R^2\left(\frac{\partial}{\partial r}\tilde{\varphi}_{lm}^{>}(R) - \frac{\partial}{\partial r}\tilde{\varphi}_{lm}^{<}(R)\right) = -4\pi\int_{r_1}^{r_2}\mathrm{d}r\,r^2\left[\delta\rho_{lm} - \left(\frac{1}{\varepsilon^{(i)}} - 1\right)(\rho_0)_{lm}\right] \tag{A2.8}$$

where $\varepsilon^{(i)}$ is defined in equation (3*d*). The quantity

$$\left(\frac{1}{\varepsilon^{(i)}} - 1\right)\rho_0$$

is the charge density induced in the sphere by the fast electron in the classical approximation while $\delta\rho$ is the actual charge induced in the sphere.

Next one writes equation (A2.3) as

$$\frac{2}{r}\frac{\partial}{\partial r}\varphi_{lm}(r) + \frac{\partial^2}{\partial r^2}\varphi_{lm}(r) - l\frac{(l+1)}{r^2}\varphi_{lm}(r) = -4\pi\rho_{lm}(r) \tag{A2.9}$$

multiplying by r and integrating from r_1 to r_2 gives

$$\varphi_{lm}(r_2) - \varphi_{lm}(r_1) + r_2\frac{\partial\varphi_{lm}(r_2)}{\partial r} - r_1\frac{\partial\varphi_{lm}(r_1)}{\partial r} - l(l+1)\int_{r_1}^{r_2}\mathrm{d}r\,\frac{\varphi_{lm}(r)}{r}$$
$$= -4\pi\int_{r_1}^{r_2}\mathrm{d}r\,r\rho_{lm}(r). \tag{A2.10}$$

Use of (A1.2) in (A2.2) and writing the resulting equation in a form similar to (A2.7) gives

$$\left(1 + r_2\frac{\partial}{\partial r}\right)\tilde{\varphi}_{lm}^{>}(r_2) - \left(1 + R\frac{\partial}{\partial r}\right)\tilde{\varphi}_{lm}^{>}(R) - \left(1 + r_1\frac{\partial}{\partial r}\right)\tilde{\varphi}_{lm}^{<}(r)$$
$$+ \left(1 + R\frac{\partial}{\partial r}\right)\tilde{\varphi}_{lm}^{<}(R) - l(l+1)\int_{r_1}^{r_2}\frac{\mathrm{d}r}{r}\,\tilde{\varphi}_{lm}(r)$$
$$= -4\pi\int_{r_1}^{r_2}\mathrm{d}r\,r(\theta(R-r)\rho_{lm}^{(\mathrm{in})}(r) + \theta(r-R)\rho_{lm}^{(\mathrm{ex})}(r)). \tag{A2.11}$$

Approximating φ in the integral in (A2.10) by $\bar{\varphi}$ and using (A2.11) in (A2.10) gives

$$\left(1 + R\frac{\partial}{\partial r}\right)\bar{\varphi}^{>}_{lm}(R) - \left(1 + R\frac{\partial}{\partial r}\right)\bar{\varphi}^{<}_{lm}(R)$$
$$= -4\pi \int_{r_1}^{r_2} \mathrm{d}r\, r\left[\delta\rho_{lm} - \left(\frac{1}{\varepsilon} - 1\right)(\rho_0)_{lm}\right]. \tag{A2.12}$$

Use of (A2.8) and (A2.12) in (9) yields equation (14) where

$$\delta\rho^{(lm)} = \delta\rho_{lm} - \left(\frac{1}{\varepsilon^{(i)}} - 1\right)(\rho_0)_{lm}. \tag{A2.13}$$

We show next that $\delta\rho^{(lm)}$ is a function of l only and not m. The actual susceptibility of the sphere must be a function of r, r' and the angle between $\boldsymbol{r}$ and $\boldsymbol{r}'$. Thus

$$\delta\rho(\boldsymbol{r}) = \int \mathrm{d}^3 r'\, \chi(\boldsymbol{r}, \boldsymbol{r}')\, \varphi(\boldsymbol{r}') \tag{A2.14a}$$

where

$$\chi(\boldsymbol{r}, \boldsymbol{r}') = \sum_l \chi_l(r, r')\, P_l(\cos\theta_{r,r'}) \tag{A2.14b}$$

and φ has the form

$$\varphi(\boldsymbol{r}') = \sum_{lm} \varphi_{lm}(r')\, Y_{lm}(\hat{\Omega}_{r'}). \tag{A2.14c}$$

It follows from (A2.14) that

$$\delta\rho(\boldsymbol{r}) = \sum_{lm} \delta\rho_{lm}(r)\, Y_{lm}(\hat{\Omega}_r) \tag{A2.15a}$$

where

$$\delta\rho_{lm}(r) = \sum_{lm} \int \mathrm{d}r'\, r'^2 \chi_l(r, r')\, \varphi_{lm}(r'). \tag{A2.15b}$$

Now $\nabla^2\varphi = -4\pi\rho$ gives

$$\frac{1}{r^2}\frac{\partial}{\partial r} r^2 \frac{\partial}{\partial r}\varphi_{lm}(r) - \frac{l(l+1)}{r^2}\varphi_{lm}(r) = -4\pi \int \mathrm{d}r'\, r'^2 \chi_l(r, r')\, \varphi_{lm}(r'). \tag{A2.16}$$

The normal modes of the sphere are determined by the solutions of (A2.16) when there is no external potential in which case $\varphi^{\mathrm{NM}} = \varphi^{\mathrm{NM}}_{lm}(r)$ is a function of l but not m because X in (A2.16) is a function of l only. The actual charge $\delta\rho$ in (A2.13) that is induced by the incoming electron can be expanded in the normal modes

$$\delta\rho(\boldsymbol{r}) = \sum_{lm} \delta\rho_l^{\mathrm{NM}}(r)\, Y_{lm}(\hat{\Omega}_r) \tag{A2.17a}$$

where

$$\delta\rho_l^{\mathrm{NM}}(r) = -(1/4\pi)\left(\frac{1}{r^2}\frac{\partial}{\partial r} r^2 \frac{\partial}{\partial r}\varphi_l^{\mathrm{NM}}(r) - \frac{l(l+1)}{r}\varphi_l^{\mathrm{NM}}(r)\right) \tag{A2.17b}$$

and consequently $\delta\rho_{lm}$ of (A2.13) is not a function of m.

Appendix 3.

In this Appendix we derive equation (21) by the use of (19) in (18). We first use equation (13) in (19) to obtain

$$\Delta\varphi^{(i)}(\boldsymbol{r}) = (4\pi)^2 \frac{e}{v} \sum_{lm} \mathrm{i}^l \int \frac{\mathrm{d}^2\boldsymbol{q}_\parallel}{(2\pi)^2} \frac{\exp(-\mathrm{i}\boldsymbol{q}_\parallel \cdot \boldsymbol{\rho}_0)}{Q^2} \times \left(\frac{1}{\varepsilon(Q,\omega)} - 1\right) Y^*_{lm}(\hat{\Omega}_Q)\, Y_{lm}(\hat{\Omega}_r)\, G_l^{(i)}(Q,r) \tag{A3.1}$$

where (i) denotes $>$ or $<$ and

$$G_l^{<}(Q,r) = j_l(Qr) - \frac{(l+1)}{\Delta_l}\left(\frac{r}{R}\right)^l \left(1 - d_r^{(l)}\frac{\partial}{\partial r}\right) j_l(QR) \tag{A3.2a}$$

$$G_l^{>}(Q,r) = \frac{\varepsilon l}{\Delta_l}\left(\frac{R}{r}\right)^{l+1} \left(1 - d_r^{(l)}\frac{\partial}{\partial r}\right) j_l(QR). \tag{A3.2b}$$

To evaluate P from (18) we require

$$I \equiv \int \mathrm{d}^2\rho_0 \exp(\mathrm{i}\boldsymbol{q}_\parallel \cdot \boldsymbol{\rho}_0)\, \Delta Q(\boldsymbol{Q},\omega) = \int \mathrm{d}^3 r \exp(\mathrm{i}\boldsymbol{q}_\parallel \cdot \boldsymbol{\rho}_0)\, \Delta Q(\boldsymbol{r},\omega). \tag{A3.3}$$

Use of (A3.1) in (A3.3) gives

$$\begin{aligned} I &= (4\pi)^2 \frac{e}{v} \sum_{lm} \mathrm{i}^l \frac{1}{Q^2}\left(\frac{1}{\varepsilon(Q,\omega)} - 1\right) Y^*_{lm}(\hat{\Omega}_Q) \int \mathrm{d}^3 r \exp(-\mathrm{i}\boldsymbol{Q}\cdot\boldsymbol{r})\, Y_{lm}(\hat{\Omega}_r) \\ &\quad \times \left(\theta(R-r)\, G_l^{<}(Q,r) + \theta(r-R)\, G_l^{>}(Q,r)\right) \\ &= (4\pi)^3 \frac{e}{v}\frac{1}{Q^2}\left(\frac{1}{\varepsilon(Q,\omega)} - 1\right) \sum_{lm} |Y_{lm}(\hat{\Omega}_Q)|^2 \int_0^\infty \mathrm{d}r\, r^2 j_l(Qr) \\ &\quad \times \left(\theta(R-r)\, G_l^{<}(Q,r) + \theta(r-R)\, G_l^{>}(Q,r)\right). \end{aligned} \tag{A3.4}$$

We next make use of the following relationships

$$\int_0^R \mathrm{d}r\, r^2 j_l(Qr)\left(\frac{r}{R}\right)^l = \frac{R^2}{Q}\left(\frac{l}{QR} j_l(QR) - j_l'(QR)\right) \tag{A3.5a}$$

$$\int_R^\infty \mathrm{d}r\, r^2 j_l(QR)\left(\frac{R}{r}\right)^{l+1} = \frac{R^2}{Q}\left(\frac{(l+1)}{QR} j_l(QR) + j_l'(QR)\right) \tag{A3.5b}$$

$$\sum_m |Y_{lm}(\hat{\Omega}_q)|^2 = (2l+1)/4\pi \tag{A3.5c}$$

$$\sum_l (2l+1) j_l^2(QR) = 1 \tag{A3.5d}$$

in equation (A3.4) to obtain

$$\begin{aligned} I &= (4\pi)^2 \frac{e}{v}\frac{1}{Q^2}\left(\frac{1}{\varepsilon(Q,\omega)} - 1\right)\left[\frac{R^3}{3} - \frac{R}{Q^3}\sum_l \frac{l(l+1)(2l+1)}{\Delta_l}(1-\varepsilon) j_l(QR)\right. \\ &\quad \times \left(1 - d_r^{(l)}\frac{\partial}{\partial r}\right) j_l(QR) + \frac{R^2}{Q}\sum_l \frac{(2l+1)}{\Delta_l} j_l'(QR)(l+1+\varepsilon l) \\ &\quad \left. \times \left(1 - d_r^{(l)}\frac{\partial}{\partial r} j_l(QR)\right)\right]. \end{aligned} \tag{A3.6}$$

Use of

$$(l+1+\varepsilon l)/\Delta_l = 1 - l(l+1)\,\Delta_l^{-1}(d_r^{(l)}/R)(\varepsilon - 1) \qquad P = -2e/((2\pi)^3\hbar v),\ \mathrm{Im}I$$

from (18) yields equation (21).

References

Ahlqvist P and Apell P 1982 *Phys. Scr.* **25** 587
Apell P 1978 *Phys. Scr.* **17** 535
—— 1981a *Phys. Scr.* **23** 284
—— 1981b *Phys. Scr.* **24** 795
—— 1982 *Phys. Scr.* **25** 57
—— 1983 *Solid State Commun.* submitted
Apell P and Ljungbert Å 1982a *Phys. Scr.* **26** 113
—— 1982 *Solid State Commun.* **44** 1367
—— 1983 *Solid State Commun.* **46** 47
Feibelman P J 1975 *Phys. Rev.* B **12** 1319
—— 1976 *Phys. Rev.* B **14** 762
—— 1982 *Prog. Surf. Sci.* **12** 287
Flores F and Garcia-Moliner F 1972 *Solid State Commun.* **11** 1295
Fujimoto E and Komaki K 1968 *J. Phys. Soc. Japan* **25** 1679
Kliewer K L 1980 *Surf. Sci.* **101** 57
Lang N and Kohn W 1973 *Phys. Rev.* B **7** 3541
Lundqvist S and Mukhopadhyay G 1980 *Phys. Scr.* **21** 503
Lushnikov A A and Simonov A J 1975 *Z. Phys.* B **21** 357
Mukhopadhyay G and Lundqvist S 1978 *Phys. Scr.* **17** 69
Penn D R and Rendell R W 1981 *Phys. Rev. Lett.* **47** 1067
—— 1982 *Phys. Rev.* B **26** 3047
Schmidt-Ott A and Siegmann H C 1980 *Phys. Rev. Lett.* **45** 1284