

Energy loss by a fast charged particle moving parallel to a surface

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A theory has been constructed for the excitation of bulk and surface plasmons by a fast charged particle in the case in which it is moving parallel to the boundary between a medium and a vacuum, remaining at all times either above the surface in vacuum or below the surface in the medium. It is found that surface effects turn out to be important even when the fast particle is moving at a significant distance from the boundary of the order of the ratio of the fast-particle velocity to the plasma frequency. We consider the effect of coupling of the reaction cross sections in different inelastic channels, which appears in particular in a decrease of the cross section for excitation of bulk plasmons with increase of the cross section for excitation of surface plasmons. It is shown that the dependence of the bulk-plasmon excitation cross section on the velocity of a particle moving parallel to the surface of the medium differs from the corresponding dependence in the case of motion of the particle in an infinite medium and coincides with the same dependence for the surface-plasmon excitation intensity but at normal incidence of the particle onto the surface. In turn the dependence of the surface-plasmon excitation cross section on the particle energy in its motion along the surface turns out to be analogous to the same dependence for the case of excitation of bulk plasmons in an unbounded medium. In motion of the particle near the surface of a thin film the shape of the energy-loss spectrum, it turns out, depends substantially on the distance of the particle from the surface, so that on change of this distance, resonance peaks in the spectrum can be transformed to minima. Finally, it is shown that the existence of a spatial dispersion of the permittivity leads to the possibility of emission of bulk plasmons by a particle which does not penetrate inside the medium at all.

1. INTRODUCTION

The surface of the medium is responsible for a number of properties of the inelastic scattering of fast particles interacting with matter. In addition to the fact that surface states are excited in this case, the surface affects the excitation of bulk states. This appears in particular in the existence of coupling between the cross sections for excitation of bulk and surface plasmons. The corresponding phenomenon was observed experimentally by Allie *et al.*¹ and was interpreted in the manner described above in a previous article by the present authors.² Surface effects can appear also in wake phenomena.^{3,4} The transition radiation of bulk plasmons is due entirely to the surface.²

Recently published experiments have studied the energy loss of fast nonrelativistic electrons moving in vacuum parallel to the surface of a medium at distances from it of the order of atomic distances, and also directly along a surface.⁵ In particular, Cowley⁵ observed a complicated dependence of the inelastic-scattering cross section on the value of the energy lost and on the degree of separation of the particle beam from the surface. In his opinion⁵ these dependences should be explained by the transition radiation of transverse electromagnetic quanta. We consider that experiments of this type must be interpreted first of all in the framework of the theory of excitation of surface and bulk plasmons with allowance for the coupling of the cross sections in these inelastic scattering channels. Particularly complicated dependences can arise here in the case in which the particle beam moves above a thin film. However, the same type of coupling

of reaction cross sections in different inelastic channels can occur already in a semi-infinite medium. Here new physical effects can arise. An interesting fact which develops is that the existence of a spatial dispersion of the permittivity of the medium makes possible the excitation of bulk plasmons by electrons which move only in the vacuum and which do not penetrate inside the medium at all. As far as we know this phenomenon in the theory has not been discussed up to this time.

In addition to experiments of the type which has been described in Ref. 5, there is another reason to consider theoretically the energy loss of a particle moving parallel to the surface of a medium. Ordinarily in intersection of the surface by the particle one calculates the scattering intensity, which is determined as the integral over all time of the interaction of the fast particle with the surface, taken from the transition probability per unit time. At normal incidence and at angles of incidence not too close to grazing, the intensity of scattering of the particle by the surface can be sufficiently small that the scattering can be discussed on the basis of perturbation theory. At large angles of incidence the fast particle interacts with the surface for such a long time that the scattering intensity can become large and the condition of applicability of perturbation theory is destroyed. Discussion of the energy loss of fast particles moving parallel to a surface permits one to use as the main characteristic of the scattering process not the scattering intensity, but the scattering probability per unit time, in working with which one can use perturbation theory. This approach turns out to be useful not only in the

case of experiments of the type described in Ref. 5, but also at grazing angles of incidence in general.

In the present work we construct a theory of the excitation by a fast charged particle of bulk and surface plasmons which are responsible for the main contribution to the energy loss of a particle moving in vacuum above the medium and a substantial contribution in motion of the particle in the medium. The particle is assumed to move parallel to the surface of the medium. We consider also the case of motion of a particle along the surface itself. It is found that surface effects turn out to be important even when the fast-particle beam is at a significant distance from the boundary, of the order of v/ω_p , where v is the fast-particle velocity and ω_p is the plasma frequency. At a fast-particle energy $E \approx 50$ keV this is a distance of the order of 100 Å.

2. EXCITATION PROBABILITY

Electrons moving in a beam parallel to the surface of a medium can be described by the wave function

$$\Psi_i(\mathbf{r}) = (\sigma S V \pi)^{-1/2} \exp\{i\mathbf{k}\mathbf{r} - (z-z_0)^2/2\sigma^2\}. \quad (1)$$

Here \mathbf{k} is the wave vector corresponding to motion of the electron in the plane parallel to the boundary, z is the normal with respect to the surface coordinate, z_0 is the average distance from the surface to the electrons in the beam, σ is the dispersion of the distribution of the electrons in the beam along the normal to the surface coordinate, and S is the surface area. According to Refs. 6 and 7 the probability per unit time of transition of a fast charged particle from the initial state $\Psi_i(\mathbf{r})$ to the final state $\Psi_f = V^{-1/2} \exp(i\mathbf{k}' \cdot \mathbf{r}')$, where V is the normalization volume, can be written in the form

$$W_{i \rightarrow f} = 2 \int d\mathbf{r} \int d\mathbf{r}' \langle \Psi_i | \hat{\rho}(\mathbf{r}') | \Psi_f \rangle \text{Im } D(\mathbf{r}, \mathbf{r}', \omega), \quad (2)$$

where the energy transferred to the medium is $\hbar\omega > 0$. The matrix elements of the operator $\hat{\rho}(\mathbf{r})$ of the charge density of the fast particle have the form

$$\begin{aligned} \langle \Psi_f | \hat{\rho}(\mathbf{r}) | \Psi_i \rangle &= (\sigma S V \pi)^{-1/2} \exp\{-i\mathbf{Q}\mathbf{r} - (z-z_0)^2/2\sigma^2\}, \\ \langle \Psi_i | \hat{\rho}(\mathbf{r}') | \Psi_f \rangle &= (\sigma S V \pi)^{-1/2} \exp\{i\mathbf{Q}\mathbf{r}' - (z'-z_0)^2/2\sigma^2\}. \end{aligned} \quad (3)$$

Thus, from Eqs. (2) and (3) it follows that

$$\begin{aligned} W_{i \rightarrow f} &= \frac{2}{\sigma S V \pi} \\ &\times \int_{-\infty}^{+\infty} dz \int_{-\infty}^{+\infty} dz' e^{i\mathbf{h}_z'(z-z')} \exp\left\{-\frac{(z-z_0)^2 + (z'-z_0)^2}{2\sigma^2}\right\} \\ &\times \text{Im } D(\omega, \mathbf{q}, -\mathbf{q}, z, z'). \end{aligned} \quad (4)$$

The quantity \mathbf{q} occurring in the argument of the Green's function $D(\omega, \mathbf{q}, -\mathbf{q}, z, z')$ of the electric field created by the medium is the component parallel to the surface of the wave vector \mathbf{Q} transferred to the medium.

In the limiting case $\sigma \rightarrow 0$ (a thin current layer) the transition probability does not depend on k_z' , the component normal to the surface of the wave vector \mathbf{k}' characterizing the final state of the fast particle. In this limiting case of an infinitely thin current layer the component normal to the surface of the wave vector of a fast particle which is in the initial state is not determined and in accordance with the uncertainty principle it can take on any value.

In the upshot, we are interested in the total cross section for scattering of a fast particle and the energy spectrum of the scattered particles corresponding to integration of the differential scattering cross section over all momentum transfers (to find the total cross section for scattering it will be necessary to integrate also over all transferred energies). Having this in mind, it is convenient to carry out immediately the integration over the z component of the momentum transfer. After this the transition probability per unit time takes the form

$$W = (2/S) \text{Im } D(\mathbf{q}, \omega, z_0, z_0). \quad (5)$$

The Green's function $D(\mathbf{q}, \omega, z_0, z_0)$ which enters into this equation contains information on the electronic excitations of the medium with allowance for the nature of the scattering of the electrons of the medium by the surface. The Green's function D was found in Ref. 2 for an arbitrary degree of roughness of the surface. It has the form

$$\begin{aligned} D(\mathbf{q}, \omega, z_0, z_0) &= \frac{2\pi e^2}{\hbar} \theta(z_0) \left\{ \frac{1-e^{-2qz_0}}{qe(\omega)} - i \frac{[\epsilon(\omega)-1](1-e^{-2iqz_0})}{re(\omega)} - \frac{2i(r-l)(l+iq)}{(r^2+q^2)(r+iq)} (e^{-qz_0}-e^{-irz_0})^2 \right\} \\ &+ \frac{4\pi e^2}{\hbar q \{1+\Xi_s(\mathbf{q}, \omega)\}} \left\{ e^{-q|z_0|} + \theta(z_0) \frac{iq(l^2-r^2)(1+P)(e^{-irz_0}-e^{-qz_0})}{(r+iq)[l((1-P)r+(1+P)l)-iq((1+P)r+(1-P)l)]} \right\}^2. \end{aligned} \quad (6)$$

In this formula θ is a step function and

$$r = \left[\left(\frac{\omega-i\nu}{bv_F} \right)^2 \epsilon(\omega, \nu) - q^2 \right]^{1/2}, \quad l = \left[\left(\frac{\omega-i\nu}{bv_F} \right)^2 - q^2 \right]^{1/2}.$$

Here P is the coefficient of mirror reflection of electrons of the medium from its surface and b is a coefficient characterizing the spatial dispersion of the longitudinal permittivity of an unbounded medium, which in the hydrodynamic ap-

proximation can be written in the form

$$\epsilon(\mathbf{Q}, \omega) = \left[\epsilon(\omega) - b^2 \left(\frac{Qv_F}{\omega-i\nu} \right)^2 \right] \left[1 - b^2 \left(\frac{Qv_F}{\omega-i\nu} \right)^2 \right]^{-1}. \quad (7)$$

In Eq. (7) $\epsilon(\omega) = 1 - \omega_p^2/(\omega - i\nu)^2$, ν is the effective collision frequency of the electrons of the medium and v_F is the velocity of electrons at the Fermi surface. Regions occupied by the medium correspond to $z > 0$. In Eq. (6)

$$1 + \Xi_s(\mathbf{q}, \omega) = 1 + \varepsilon(\omega) - \frac{i q v_F b (1-P) [1-\varepsilon(\omega)]}{2\omega \{1+[1-b^2(qv_F/\omega)^2]^{1/2}\}} - \frac{q(1+P)[1-\varepsilon(\omega)](q+ir)}{(1+P)(l^2-iqr)+i(1-P)l(q-ir)}. \quad (8)$$

The Green's function is particularly simple in the case in which the fast particle moves only in the vacuum, not penetrating into the medium. In this case

$$D(\mathbf{q}, \omega, z_0, z_0) |_{z_0 < 0} = \frac{4\pi e^2 \exp(-2q|z_0|)}{\hbar q [1 + \Xi_s(q, \omega)]}. \quad (9)$$

For $P = 1$ (the case of pure mirror reflection of the electrons of the medium from the boundary) Eq. (9) is simplified. The function $\Xi_s(q, \omega)$ is given by Eq. (8) in the form of an algebraic function. However, to understand its meaning better, it is useful also to write this function in the form of an integral over the wave vectors u that the boundary exchanges with the system of electrons of the medium. For $P = 1$ we have

$$1 + \Xi_s(\mathbf{q}, \omega) = 1 + \frac{q}{\pi} \int_{-\infty}^{+\infty} \frac{du (u^2 - l^2)}{(u^2 + q^2)(u^2 - r^2)}. \quad (10)$$

3. EXCITATION OF BULK AND SURFACE PLASMONS BY A CHARGED PARTICLE MOVING IN VACUUM PARALLEL TO THE INTERFACE

It is well known that if we do not take into account the spatial dispersion of the permittivity, the electric field due to the existence of bulk plasmons is nonvanishing only inside the material. This means that a fast particle moving in vacuum parallel to a sharp medium-vacuum interface cannot excite bulk plasmons. Of course, a real interface is not completely abrupt, even if the medium is a solid. The potential barrier which simulates the potential acting on the electrons of the medium from the direction of the surface has a finite width and height. This leads to the existence of tails of the density of the electrons of the medium near the surface. At the present time it is considered that a fast particle moving in vacuum along a surface can excite bulk plasmons only as the result of interaction with these tails. In the present section we shall show that even if such tails of the electron density did not exist at all and the fast particle were moving in a vacuum in the complete sense of this word, nevertheless excitation of bulk plasmons would be possible as a result of a new mechanism of generation, in description of which it is necessary to take into account spatial-dispersion effects which lead to extension of the electric field of bulk plasmons beyond the material. In this section we shall also find the dependence of the probability of emission of surface plasmons on the value of $|z_0|$ and the other parameters of the problem.

In the previous section it was shown that if a fast particle is moving in vacuum at a distance $|z_0|$ from the medium-vacuum interface, the Green's function $D(\mathbf{q}, \omega, z_0, z_0)$ is described by Eq. (9). This formula can be further simplified if we write it in the approximation linear in the vector \mathbf{q} . In this approximation

$$D = \frac{4\pi e^2}{\hbar q} e^{-2q|z_0|} \left[\theta(\omega_p - \omega) \left\{ 1 + \varepsilon(\omega) - \frac{bqv_F}{\omega} [1 - \varepsilon(\omega)] \right. \right. \\ \times \left[\frac{(1+P)^2 (-\varepsilon(\omega))^{1/2}}{(1+P)^2 + (1-P)^2 \varepsilon(\omega)} \right] \left. \right]^{-1} \\ + i(1-P) \left[\frac{1}{4} - \frac{\varepsilon(\omega) (1+P)}{(1+P)^2 + (1-P)^2 \varepsilon(\omega)} \right] \left. \right]^{-1} \\ + \theta(\omega - \omega_p) \left\{ 1 + \varepsilon(\omega) - \frac{bqv_F}{\omega} i [1 - \varepsilon(\omega)] \right. \\ \times \left[\frac{1-P}{4} + \frac{\varepsilon''(\omega) (1+P)}{1+P+(1-P)\varepsilon''(\omega)} \right] \left. \right]^{-1}. \quad (11)$$

The first term in this formula describes first of all the excitation of a surface plasmon. The appearance of this term in D is quite natural. It is known that the electric field associated with a surface plasmon is present both in the medium and in the vacuum, and that the potential of the plasmon field falls off exponentially with distance from the surface. Therefore a fast particle moving in vacuum has the possibility of interacting directly with this field. This is true even in the case when one does not take into account the spatial dispersion of the permittivity.

The total probability of excitation of all electronic states in the medium is related to the Green's function $D(\mathbf{q}, \omega, z_0, z_0)$ by the expression

$$W_t = \frac{1}{2\pi^2} \int_0^\infty d\omega \int d\mathbf{q} \delta(\omega - v\mathbf{q}) \text{Im } D(\mathbf{q}, \omega, z_0, z_0). \quad (12)$$

Calculating the contribution to W_t from the right-hand side of the function (11), we must integrate over ω from 0 to ω_p . From the structure of the denominator of the first term in (11) it follows that even if we neglect the imaginary part of the denominator, the pole of this denominator turns out to be on the integration path. Therefore the contribution of surface plasmons (more precisely, of excitations whose energies lie in the interval $0 < \hbar\omega < \hbar\omega_p$) in (12) is finite even for an infinitely small imaginary part of the denominator of the first term in (11). Thus, the total probability of excitation of a surface plasmon takes the form

$$W_{ts} = \frac{e^2 \omega_p}{\sqrt{2} \hbar} \int_{\omega_p/\sqrt{2}v}^\infty dq \exp(-2q|z_0|) \left[q^2 v^2 - \frac{\omega_p^2}{2} \right]^{-1/2}. \quad (13)$$

This formula is obtained from (11) and (12) after integration over ω and over the angle between the vectors \mathbf{v} and \mathbf{q} . This expression does not contain v_F , which means that the spatial dispersion of the permittivity makes only a small contribution to W_{ts} in the nature of a correction. The integral in (13) is equal to the Macdonald function K_0 . Thus,

$$W_{ts} = \frac{e^2 \omega_p}{\sqrt{2} \hbar v} K_0 \left(\frac{\sqrt{2} \omega_p |z_0|}{v} \right). \quad (14)$$

As we have already mentioned, this formula was obtained with neglect of the spatial dispersion. The smallness of the latter means that $qv_F/\omega < 1$. The field of a surface plasmon falls off exponentially with distance from the surface into the vacuum, and the characteristic length has a

value q^{-1} . Therefore a fast particle interacts with surface plasmons whose wave vectors are $q \lesssim 1/|z_0|$. Thus, smallness of the spatial dispersion corresponds to the inequality $|z_0| > v_F/\omega_p$. It is just for such z_0 values that Eq. (14) is valid. One can see from (14) that a surface plasmon is rather intensively excited when the argument of the Macdonald function is of the order of or less than unity. In the argument of the function K_0 we have the ratio of the lengths $|z_0|/(v/\omega_p)$, and therefore a fast particle can excite surface plasmons in moving from the surface of the medium to a distance which considerably exceeds atomic dimensions. Equation (14) agrees with the results obtained for the specific energy loss by a fast particle to surface plasmons by another method in Ref. 4.

The second term in (11) describes a less obvious effect—the excitation of plasma waves whose frequencies correspond to bulk plasmons by a particle which does not penetrate into the medium at all. From (11) and (12) it is evident that a finite integral over ω with the second term of Eq. (11) under the integral cannot be provided by an infinitely small imaginary addition to $\epsilon(\omega)$, since the pole of the integrand turns out to be outside the integration contour. Consequently the total probability of excitation of bulk plasmons W_{tb} will be due to the imaginary part of the denominator of the second term in (11), which is related to the frequency v of collision of the electrons of the medium and to the imaginary part of the above-mentioned denominator, which is proportional to v_F . This latter imaginary part will be most important and most interesting if $qv_F > v$. The minimum value of q is due to the conservation laws (which correspond to the δ function in Eq. (12)) and is equal to ω/v . Consequently $\text{Im } D$ will be due to the spatial dispersion if $\omega/v > v/v_F$. In what follows we shall assume that this condition is satisfied. Then the effect of excitation of bulk plasmons by a fast particle moving outside the medium can be considered as the effect of spatial dispersion of the permittivity. In this case

$$\begin{aligned} \text{Im } D(q, \omega, z_0, z_0) = & \frac{4\pi e^2 b v_F [1 - \epsilon(\omega)]}{\hbar \omega [1 + \epsilon(\omega)]^2} \\ & \times \left[\frac{\epsilon^{1/2}(\omega) (1+P) \theta(\omega - \omega_p)}{1+P+(1-P)\epsilon^{1/2}(\omega)} + \frac{1-P}{4} \right] e^{-2q|z_0|}. \end{aligned} \quad (15)$$

The first term in (15) is due to the imaginary part of the integral in (10). It can be seen from (10) that it is determined by the contribution from the poles at the points $u^2 = r^2$. The wave vector u which follows from this equation corresponds to the momentum transferred to the boundary (in a direction normal to it) by the electrons of the medium. Here the possibility arises of removal of the plasma wave from the boundary into the bulk. In this case the total momentum transferred to the electrons of the medium is greater than ω/v . This is evident directly from conservation of energy-momentum in such a process, from which it follows that $\omega = \mathbf{Q} \cdot \mathbf{v}$. Therefore the minimum momentum transferred to the electrons of the medium is tangential with respect to the surface and is equal to ω/v . Consequently this process of emission of a bulk plasmon for $P = 1$ is a Čerenkov process, but the surface takes part in this process.

The second term in the square brackets in (15) exists only for $P \neq 1$, i.e., in the case of a rough surface. For $P \neq 1$

the scattering cross section, as can be seen from (15), increases. For $P \neq 1$ the conservation of energy and momentum takes the form

$$\omega = (\mathbf{Q}_e + \mathbf{Q}_{\text{surf}}) \cdot \mathbf{v}.$$

Here \mathbf{Q}_e is the tangential component of the momentum transferred to the electron subsystem and \mathbf{Q}_{surf} is the tangential component of the momentum transferred to the surface. The sum $|\mathbf{Q}_e + \mathbf{Q}_{\text{surf}}|$ coincides with the quantity ω/v , and the electrons of the medium can receive also a smaller momentum. Consequently roughness of the surface leads to transition radiation of bulk plasmons.

The total probability of radiation of bulk plasmons is

$$\begin{aligned} W_{tb} = & \frac{2e^2 b v_F \omega_p}{\pi \hbar v^2} \\ & \times \int_0^1 \frac{dx}{(1-x)^{1/2} (1+x)^2} \left[\frac{(1-P)}{4} + \frac{(1+P)\sqrt{x}}{1+P+(1-P)\sqrt{x}} \right] \\ & \times K_1 \left(\frac{2\omega_p}{v} \frac{|z_0|}{(1-x)^{1/2}} \right). \end{aligned} \quad (16)$$

In the region $(\omega_p/v)|z_0| < 1$

$$W_{tb} \approx \frac{e^2}{\pi \hbar v} \frac{b v_F}{|z_0|} \left\{ \frac{1-P}{8} - \frac{1-P}{1+P} \left[\ln 2 - \frac{1}{2} \right] + \frac{\pi-2}{4} \right\}. \quad (16')$$

This formula can be compared with W_{ts} taken for the same values of z_0 . For these z_0 values the probability W_{ts} can be represented in the form

$$W_{ts} \approx \frac{e^2 \omega_p}{\hbar v \sqrt{2}} \left[\ln \left(\frac{v \sqrt{2}}{\omega_p |z_0|} \right) + C \right], \quad (17)$$

where C is Euler's constant.

The ratio of the probabilities is given by

$$\begin{aligned} \frac{W_{tb}}{W_{ts}} \approx & \frac{\sqrt{2} b v_F}{\pi \omega_p |z_0|} \left\{ \frac{1}{8} (1-P) - \frac{(1-P)}{(1+P)} \left[\ln 2 - \frac{1}{2} \right] + \frac{\pi-2}{4} \right\} \\ & \times \left\{ C + \ln \left(\frac{v \sqrt{2}}{\omega_p |z_0|} \right) \right\}^{-1}. \end{aligned} \quad (18)$$

For $v_F/\omega_p \approx z_0 \ll v/\omega_p$ this ratio can be the order of one tenth if z_0 , as is reported in the experimental study of Ref. 5, is of the order of one angstrom or smaller. It is evident from this that the energy-loss spectrum of particles moving just above the surface of a semi-infinite medium can have a rather complicated form.

In concluding this section we note that in motion of a fast particle in the immediate vicinity of the surface of a medium when the fast particle feels the tails of the electron density, both mechanisms of bulk plasmon generation (that proposed by us and that due to the tails) can contribute comparably to the total cross section for scattering of a fast particle (integrated over frequency). We can suppose, however, that these two mechanisms determine different parts of the bulk-plasmon energy spectrum. The new mechanism found by us is responsible for the generation of waves whose frequencies are greater than ω_p , as follows directly from the

form of the second part of Eq. (11). This follows even more clearly from Eq. (16). The integration in (16) is over the variable $x = 1 - \omega_p^2/\omega^2$, so that in essence this corresponds to integration over the frequencies of the generated excitations. The integral in (16) is determined substantially by the value of the integrand for all x , and not only for $x = 0$.

In regard to the contribution of the tails of the electron density, this problem is most simply solved in the case in which the electron concentration in the medium at the boundary changes so slowly that one can make use of the concept of a local bulk-plasmon frequency (this situation is realized, for example, in the region of a *p-n* junction in semiconductors). In this case there will be generated plasmons whose frequency ω is less than the frequency ω_p characteristic of the main bulk of the material. In the case of a sharply inhomogeneous boundary this question is not so simple and presents independent interest. However, even in this case apparently as the result of the tails of the electron density there are generated plasmons with frequencies less than those which are due to the new mechanism. The wavelength of a plasmon is usually significantly greater than the atomic length in which the tails of the electron density die out. Therefore an excess of the frequency ω of the generated bulk excitations over ω_p can be due most likely to the usual dependence of ω on the wave vector \mathbf{q} . This correction is quadratic in the spatial-dispersion parameter qv_F/ω . However, the correction to $\text{Im } D$ due to the new mechanism, as follows from (11) and (16), is linear in this parameter. If this is the case, then the new mechanism and the more trivial mechanism of bulk-plasmon production are not so much competitive as they are complementary.

4. EXCITATION OF PLASMONS BY A CHARGED PARTICLE MOVING IN A MEDIUM PARALLEL TO THE SURFACE

If a charged particle is moving in a medium below the surface, the Green's function $D(\mathbf{q}, \omega, z_0, z_0)$ corresponding to this case has the form

$$D(\mathbf{q}, \omega, z_0, z_0) = \frac{2\pi e^2}{\hbar q \epsilon(\omega)} - \frac{2\pi e^2 [1 - \epsilon(\omega)]}{\hbar q \epsilon(\omega) [1 + \epsilon(\omega)]} e^{-2qz_0}. \quad (19)$$

This Green's function is written with neglect of the spatial dispersion of the permittivity. If the fast particle is moving in the medium, then in contrast to the case which was discussed in the previous section, for coupling of a bulk plasmon and a fast particle there is no need of taking into account the spatial dispersion of the permittivity. The total probability of scattering of a fast particle by plasmons, obtained by means of Eq. (19), has the form

$$W = \frac{2e^2}{\pi \hbar v} \left\{ \ln \left(\frac{2v}{v_F} \right) \int_0^\infty d\omega \text{Im} \frac{1}{\epsilon(\omega)} - \int_0^\infty d\omega \text{Im} \frac{1 - \epsilon(\omega)}{\epsilon(\omega) [1 + \epsilon(\omega)]} \left[K_0 \left(\frac{2\omega z_0}{v} \right) - E_1 \left(\frac{2\omega z_0}{v_F} \right) \right] \right\}. \quad (20)$$

The function E_1 is the integral

$$E_1(x) = \int_1^\infty dt t^{-1} e^{-tx}.$$

The formula (20) describes the excitation of both bulk and surface plasmons. From (20) it follows that the probability of excitation of bulk plasmons is

$$W_e = \frac{e^2 \omega_p}{\hbar v} \left[\ln \left(\frac{2v}{v_F} \right) - K_0 \left(\frac{2\omega_p z_0}{v} \right) + E_1 \left(\frac{2\omega_p z_0}{v_F} \right) \right]. \quad (21)$$

Let's analyze this formula. As $z_0 \rightarrow \infty$, which corresponds to motion of a particle in an unbounded medium, we obtain the well known result:

$$W_e = \frac{e^2 \omega_p}{\hbar v} \ln \left(\frac{2v}{v_F} \right). \quad (22)$$

As $z \rightarrow 0$ we obtain at $W_e \rightarrow 0$. This means that in motion of a fast particle immediately along the surface, bulk plasmons can be radiated only as the result of spatial dispersion of the permittivity. From (20) it can be seen that an increase of the probability of excitation of a surface plasmon leads to a decrease of the probability of excitation of a bulk plasmon. This effect is especially important for $z_0 < v/\omega_p$. Here the argument of the function K_0 in (20) is small in comparison with unity, and the value of K_0 is relatively large. The argument of the function E_1 in this case will depend on the Fermi velocity v_F . If $v/\omega_p > z_0 \gg v_F/\omega_p$, then

$$W_e = \frac{e^2 \omega_p}{\hbar v} \left[\ln \left(\frac{2\omega_p z_0}{v_F} \right) - C \right]. \quad (23)$$

Now, in contrast to (22), the velocity of the fast particle does not appear in the argument of the logarithm. This means that the dependence of the probability of excitation of bulk plasmons by a particle moving parallel to the surface on the energy has the same form as the analogous dependence of the intensity P_s of excitation of a surface plasmon, but at normal incidence of the particle onto the surface. The decrease of the value of W_e with increase of the angle of incidence of the electrons on the surface of aluminum observed experimentally in Ref. 1, we assume, can be explained mainly by the mutual influence of the bulk and surface channels of the reactions. Since in experiments of this type the electron traverses some path in the medium below the surface at an effective distance z_0 from the boundary, and the quantity z_0 will depend on the angle of incidence of the electron onto the surface, then the observed angular dependence of the cross section for excitation of a bulk plasmon is due to the second term and partly to the third term in Eq. (21).

From Eq. (20) we obtain also the total probability for excitation of surface plasmons

$$W_s = \frac{4e^2}{\pi \hbar v} \int_0^\infty d\omega \text{Im} \frac{1}{1 + \epsilon(\omega)} \left[K_0 \left(\frac{2\omega z_0}{v} \right) - E_1 \left(\frac{2\omega z_0}{v_F} \right) \right] = \frac{e^2 \omega_p}{\sqrt{2} \hbar v} \left[K_0 \left(\frac{\sqrt{2} \omega_p z_0}{v} \right) - E_1 \left(\frac{\sqrt{2} \omega_p z_0}{v_F} \right) \right]. \quad (24)$$

For $v_F/\omega_p \ll z_0 < v/\omega_p$ this probability is

$$W_s \approx \frac{e^2 \omega_p}{\sqrt{2} \hbar v} \left[\ln \left(\frac{\sqrt{2} v}{\omega_p z_0} \right) - C \right]. \quad (25)$$

In other words, the dependence of W_s on the energy turns

out to be the same as the dependence of W_e on E in an unbounded medium. Thus, in motion of a fast particle below the surface of the medium and parallel to it, the dependence of the total probability for excitation of bulk plasmons on the particle energy turns out to be that which exists in the case of excitation of surface plasmons in normal incidence of a particle onto the surface. And on the other hand the total probability of excitation of surface plasmons turns out to depend on the energy of a particle moving parallel to the surface in the same way as occurs in excitation of bulk plasmons by particles without taking into account the boundary.

Let us consider now the motion of fast particles near a thin (in the electrodynamic sense) film. The special feature of this problem is that in this case there are, not two main channels, but a number of channels of inelastic scattering, so that all reaction channels can influence each other. Consider a film of thickness d with permittivity $\epsilon(\omega)$, with interfaces with vacuum on both sides. We shall take the origin of coordinates at the surface above which the fast particle is moving. The Green's function D for $z_0 < 0$ has in this case the form

$$D(q, \omega, z_0, z_0) = \frac{4\pi e^2}{\hbar q} e^{-2q|z_0|} \frac{1 + \epsilon(\omega) - [1 - \epsilon(\omega)]e^{-2qd}}{[1 + \epsilon(\omega)]^2 - [1 - \epsilon(\omega)]^2 e^{-2qd}}. \quad (26)$$

The transition probability obtained by means of (26) for a fixed value of energy transfer $\hbar\omega$, and integrated over all momentum transfers is

$$\begin{aligned} w_s(\omega, z_0) &= \frac{4e^2}{\pi\hbar v} \int_{\omega_p}^{2k} dq \frac{e^{-2q|z_0|}}{(q^2 - \omega^2/v^2)^{1/2}} \operatorname{Im} \frac{1 + \epsilon(\omega) - [1 - \epsilon(\omega)]e^{-2qd}}{[1 + \epsilon(\omega)]^2 - [1 - \epsilon(\omega)]^2 e^{-2qd}} \\ &= -\frac{4e^2\epsilon(\omega)}{\hbar v[1 - \epsilon(\omega)][1 + \epsilon(\omega)]} \theta \left(\ln \left| \frac{1 - \epsilon(\omega)}{1 + \epsilon(\omega)} \right| - \frac{\omega d}{v} \right) \\ &\times \theta \left(kd - \ln \left| \frac{1 - \epsilon(\omega)}{1 + \epsilon(\omega)} \right| \right) \left\{ \left[\ln \left| \frac{1 - \epsilon(\omega)}{1 + \epsilon(\omega)} \right| \right]^2 - \left(\frac{\omega d}{v} \right)^2 \right\}^{-1/2} \\ &\times \exp \left\{ -\frac{2|z_0|}{d} \ln \left| \frac{1 - \epsilon(\omega)}{1 + \epsilon(\omega)} \right| \right\}. \end{aligned} \quad (27)$$

In this case we intentionally did not integrate the transition probability over the energy transfer $\hbar\omega$, since the presence of several inelastic channels is manifested primarily in the energy spectra of the scattered particles. The scattered-particle spectrum will depend substantially on the ratio $|z_0|/d$. From Eq. (27) it is evident that when $2|z_0|/d > 1$, the exponential contained in w_s is not small only when $\ln|(1 - \epsilon(\omega))/(1 + \epsilon(\omega))|$ is small. The minimum possible value of this logarithm is determined by the fact that the following condition must be satisfied:

$$\ln \left| \frac{1 - \epsilon(\omega)}{1 + \epsilon(\omega)} \right| \geq \frac{\omega d}{v}. \quad (28)$$

If (28) is considered as an equation, this expression will coincide with the well known equation which determines the frequencies of plasmons which are called normal and tangential

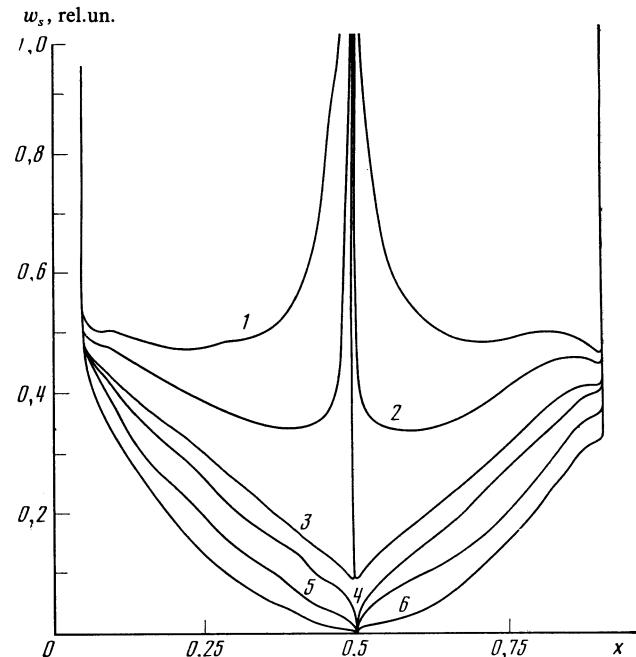


FIG. 1. Energy spectrum $w_s(\omega)$ of the scattered particles. The abscissa is $x = (\omega/\omega_p)^2$, and the ordinate is the probability of scattering with energy loss \sqrt{x} . The region $0 < x < 0.5$ corresponds to excitation of tangential plasmons, and the region $0.5 < x < 1$ is due to excitation of normal plasmons. Curve 1 corresponds to $|z_0|/d = 0.1$, curve 2 corresponds to 0.25, curve 3 corresponds to 0.45, curve 4 to 0.55, curve 5 to 0.75, and curve 6 to 1.0. The curves have been plotted for the case $E = 100$ keV, $d = 10^{-7}$ cm, and $\omega_p = 15$ eV. The nature of the curves is retained as long as the film can be considered thin, i.e., as long as $\omega_p d/v < 1$.

surface plasmons.^{8,9} The right hand side of Eq. (28) is always small by definition. Indeed, ω_p/v is, as follows directly from the conservation laws, the minimum wave vector which the produced plasmon can have. Therefore $\omega_p d/v = d/\lambda_{\max}$, where $\lambda_{\max} = v/\omega_p$ is the maximum wavelength of the plasmon. Since we are considering films which are thin in the electrodynamic sense, this means that $d/\lambda_{\max} < 1$. Thus, for $2|z_0|/d > 1$ from the condition of smallness of the logarithm mentioned above we obtain the estimate

$$\frac{1 - \epsilon(\omega)}{1 + \epsilon(\omega)} = \frac{\omega_p^2}{2\omega^2 - \omega_p^2} \sim 1,$$

from which it follows that plasmons with frequencies close to $\omega_p/\sqrt{2}$ are excited only weakly in this case.

When the particle moves comparatively close to the film surface so that $2|z_0|/d < 1$, then a rise of the part of the spectrum in the frequency region close to $\omega_p/\sqrt{2}$ occurs. This means that the shape of the scattered-particle energy spectrum can change very substantially on change of the distance $|z_0|$. As can be seen from Fig. 1, in which we have shown the function $W_s(\omega)$ plotted on the basis of Eq. (27), for a comparatively small change of $|z_0|$ the spectrum changes from the usual spectrum in which a resonance at the frequency $\omega_p/\sqrt{2}$ is distinctly expressed to a spectrum in which there is a clearly expressed minimum in the region of the frequency $\omega_p/\sqrt{2}$ of the surface plasmon. This change in the nature of the spectrum w_s at frequencies ω close to $\omega_p/\sqrt{2}$ on

variation of the parameter $|z_0|/d$ recalls the nature of variation of the function

$$|\omega^2/\omega_p^2 - 0.5|^{2|z_0|/d-1}$$

as a function of the value of $|z_0|/d$. The appearance of two peaks in the scattered-particle spectrum at small and large ($\sim \omega_p$) frequencies is apparently in qualitative correspondence with the results of the experiment of Ref. 5. The presence of a substrate on which the film is deposited can of course lead to still greater complication of the energy spectrum.

¹J. Allie, E. Blanc, and D. Duffayard, *Surf. Sci.* **62**, 215 (1977).

²V. V. Rumyantsev and B. N. Libenson, *Zh. Eksp. Teor. Fiz.* **83**, 247 (1982) [Sov. Phys. JETP **56**, 135 (1982)].

³J. Schilling and H. Raether, *J. Phys. C6*, L358 (1973).

⁴Yu. V. Kononets and G. M. Filippov, *Zh. Eksp. Teor. Fiz.* **83**, 971 (1982) [Sov. Phys. JETP **56**, 547 (1982)].

⁵J. M. Cowley, *Phys. Rev. B25*, 1401 (1982).

⁶B. N. Libenson and V. V. Rumyantsev, *Fiz. Tverd Tela (Leningrad)* **20**, 65 (1978) [Sov. Phys. Solid State **20**, 34 (1978)].

⁷Yu. A. Romanov, *Zh. Eksp. Teor. Fiz.* **47**, 2119 (1964) [Sov. Phys. JETP **20**, 1424 (1965)].

⁸Yu. A. Romanov, *Izv. vuzov, Radiofizika* **7**, 828 (1964).

⁹R. A. Ferrell, *Phys. Rev.* **111**, 1214 (1958).

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