Transition radiation of bulk plasmons

V. V. Rumyantsev and B. N. Libenson

Leningrad Polytechnic Institute
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Generation of bulk plasmons by electrons near the interface of a medium and vacuum is investigated theoretically. The possibility of bulk-plasmon emission as a result of momentum exchange between the plasmon and the interface is shown. By analogy with the generation of transverse electromagnetic quanta by an electron moving uniformly from a vacuum into a medium, this process is called transition radiation of bulk plasmons. Along with features common to transition radiation of longitudinal and transverse quanta, they have also significant differences. In particular, transition radiation of bulk plasmons is a spatial-dispersion effect. A characteristic feature of this phenomenon is an increase of the probability of electron scattering with emission of a bulk plasmon with increasing angle of incidence of the electron on the interface. The most favorable conditions for the observation this transition radiation are discussed. It should occur when intermediate-energy electrons are reflected from polycrystals.

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1. INTRODUCTION

It is usually assumed that bulk plasmons are generated by a Čerenkov mechanism. This assumption is in fact the basis of all the fundamental papers on the theory of excitation of bulk plasmons.[1,2] However, the cross section for excitation plasmons by intermediate-energy electrons is large. This means that excitation of bulk plasmons in condensed media takes place near the surface, rarely at distances exceeding one or two dozen angstroms, and sometimes at a distance of only several angstroms. The interface between the medium and vacuum not only ensures an electron-energy loss to excitation of surface plasmons, but should influence also the generation of bulk plasmons. This influence is known as the size effect and manifests itself when the plasmons are excited in thin films. The size effect decreases the cross section for the excitation of a bulk plasmon. When plasmons are excited in a semi-infinite medium there is no size effect. Even then, however, the interface can influence the bulk-plasmon generation.

We show in this paper that there exists a new bulk-plasmon excitation mechanism. The corresponding process is "non-Čerenkov" and is due to momentum exchange between the electrons participating in the plasma oscillations together with the corresponding electromagnetic field and the surface. This process takes place also when the change of the electron velocity as it crosses the interface can be neglected. Generally speaking, the electron velocity changes when it crosses the interface. This should also cause the emission of bulk plasmons, which can be called bremsstrahlung of bulk plasmons at the interface. This radiation, however, is not of great interest, since it can consist of only shortest-wavelength strongly attenuating plasmons. On the other hand momentum exchange between the bulk plasmon and the boundary makes possible generation of long-wave plasmons, even such whose wave vector **Q** satisfies the condition $Q < \omega_p/v_p$; and can therefore not be generated by the "Čerenkov" mechanism (here ω_p is the bulk-plasmon frequency and v_p is the velocity of the plasmon-exciting electron). In analogy with the generation of transverse electromagnetic-field quanta on a surface, for which a theory was constructed in Ref. 3 and further developed in a number of succeeding papers,[4] the proposed method of generating longitudinal quanta can be called transition radiation of bulk plasmons. One must bear in mind here, however, not only the analogy between these two processes, but also the substantial differences both between the generation processes proper and the fact that whereas the light is radiated into the vacuum, the plasmons remain in the medium and no radiation to the vacuum is produced. What is measured in experiment is only the cross section for electron scattering with a characteristic energy loss. Transition radiation of plasmons should therefore manifest itself in the fact that the cross section for excitation of volume plasmon, which disregarding the surface should not depend upon the angle α of incidence of the electrons on the interface between the vacuum and an infinite medium, acquires a definite dependence on α. This process can be particularly important in the scattering of intermediate-energy electrons through large angles in semi-infinite media.

2. PROBABILITY OF PLASMON GENERATION ON A SURFACE, AND RESONANCE LINE SHAPE

According to Refs. 5 and 6, the probability, per unit time, of the transition of an incident electron from a state with momentum **p** into a state with momentum **p** −**Q**, with the electron losing an energy ω, is connected with the retarded Green's function D^R of the field of the electrons of the medium by the relation

$$W_{\mathbf{p},\mathbf{p}-\mathbf{Q}} = 2\,\mathrm{Im}\,D^R(\mathbf{Q}, -\mathbf{Q}, \omega). \tag{1}$$

This equation is valid if the medium is inhomogeneous, fills the half-space and interfaces with the vacuum, provided that the change of the incident-electron velocity as a result of its direct interaction with the interface can be neglected. It is also valid at any incidence angle of the electron on the interface and presupposes only applicability of perturbation theory to the description of the interaction between the incident electron and the electron subsystem of the solid. The Green's function in this equation is the Fourier transform of the Green's function $D(x, x')$ defined by the equation

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$$\Delta D(x,x')+4\pi\int d^4x_1\Pi(x,x_1)D(x_1,x')=-4\pi\delta(x-x'). \qquad (2)$$

As shown in Ref. 7, the polarization operator $\Pi(x,x')$, which is the kernel of Eq. (2), and with it also the dielectric constant $\varepsilon(\mathbf{Q},\omega)$ of the medium, can be obtained in the classical approximation from the solution of the Boltzmann kinetic equation with a boundary condition that takes into account the character of the scattering of the electrons of the medium by the interface. In this paper we choose this condition to be the well known Fuchs boundary condition. The phenomenological Fuchs condition, which relates the nonequilibrium distribution functions $f_1^{(1)}$ and $f_1^{(2)}$ of the medium electrons incident on the interface and leaving it, respectively, is written in the form

$$f_0+f_1^{(2)}(v_x,v_y,v_z,z=0)=P[f_0+f_1^{(1)}(v_x,v_y,-v_z,z=0)]+(1-P)f_0.$$

The medium occupies the half-space $z>0$. The plane $z=0$ coincides with the interface. Here P is the coefficient of specular reflection of the electrons of the medium from the boundary, and f_0 is the equilibrium (Fermi) distribution function. The Fuchs condition was chosen because it is a rather general and at the same time sufficiently simple classical boundary condition. The form of the boundary condition, generally speaking, influences the character of the plasmon transition radiation, but is not decisive. The very existence of the boundary is more important than the properties of the scattering of the electrons of the medium from it. With this approach, we take into account in the function $D(x,x')$, which is a solution of Eq. (2), both the very existence of the interface and the character of the scattering of the medium electrons from it. The probability of the transition (1) is thereby expressed in terms of so lucid a classical parameter as the coefficient P.

It can be shown that the polarization operator $\Pi(x,x')$ consists of two parts. The first is the polarization operator of a medium with a specularly reflecting boundary, multiplied by the coefficient P of the specular reflection of the electrons of the medium from the interface. The second part is the polarization operator of a medium with a diffusely reflecting interface, multiplied by $1-P$.

The function $D(\mathbf{Q},-\mathbf{Q},\omega)$ which determines the transition probability, is the Fourier transform of a function that has an integral dependence on the coordinate z normal to the surface. The Dyson equation for this function $D(z,z',\mathbf{q},\omega)$ will take the form (the $\mathbf{q}$ in the argument of D is the tangential (to the surface) part of the wave vector $\mathbf{Q}$, and the normal component of the momentum $\mathbf{Q}$ will hereafter be designated k)

$$\begin{aligned}
&\frac{d^2D(z,z',\mathbf{q},\omega)}{dz^2}-q^2D(z,z',\mathbf{q},\omega)\\
&+4\pi P\int_0^\infty dz_1D(z_1,z',\mathbf{q},\omega)\{\Pi_0(|z-z_1|,\mathbf{q},\omega)+\Pi_0(z+z_1,\mathbf{q},\omega)\}\\
&+4\pi(1-P)\int_0^\infty dz_1D(z_1,z',\mathbf{q},\omega)\Pi_0(|z-z_1|,\mathbf{q},\omega)\\
&+(1-P)\delta(z-s)\left\{2\int_0^\infty dz_1D(z_1,z',\mathbf{q},\omega)\int_{-\infty}^{+\infty}\frac{du\,u\exp(iuz_1)}{u^2+q^2}\Pi_0(u,\mathbf{q},\omega)\right.\\
&+2D(0,z',\mathbf{q},\omega)\int_{-\infty}^{+\infty}\frac{du}{u^2}\Pi_0(u,\mathbf{q}=0,\omega)\\
&\left.-2D(0,z',\mathbf{q},\omega)\int_{-\infty}^{+\infty}\frac{du\,u\exp(iuz)}{u^2+q^2}\Pi_0(u,\mathbf{q},\omega)\right\}=-4\pi\delta(z-z'). \qquad (3a)
\end{aligned}$$

This equation is valid at $z>0$. At $z<0$ the function D is given by the trivial equation

$$d^2D(z,z',\mathbf{q},\omega)/dz^2-q^2D(z,z',\mathbf{q},\omega)=0. \qquad (3b)$$

At the classical kinetic approach used here to describe the properties of the electrons of a metal, the polarization operator of an unbounded medium is of the form

$$\begin{aligned}
\Pi_0(|z|,\mathbf{q},\omega)&=\frac{3\omega_p^2(\nu+i\omega)}{8\pi v_F^3}\int_1^\infty du\frac{\mathscr{E}(u)}{[u^2-(qv_F/(\omega-i\nu))^2]^{1/2}}-\frac{3\omega_p^2}{v_F^2}\delta(z),\\
\mathscr{E}(u)&=\exp\left\{-|z|\left(\frac{\nu+i\omega}{v_F}\right)\left[u^2-\left(\frac{qv_F}{\omega-i\nu}\right)^2\right]^{1/2}\right\}. \qquad (3c)
\end{aligned}$$

In Eqs. (3), ω_p is the plasma frequency, $s\to+0$, v_F is the electron velocity on the Fermi surface, and ν is the frequency of the electron collisions in the medium.

Equations (3) were solved by the Wiener-Hopf method, which is based on the possibility of factorizing the kernel of the equation. This method works if this factorization of the kernel is actually feasible. When the coefficient P is not equal to zero, this factorization is possible only if the dielectric constant of an infinite medium is simple enough. In particular, the dielectric constant must not have logarithmic singularities. It is best for it to have only simple poles. This condition is satisfied by a model dielectric function of the form

$$\varepsilon(\mathbf{Q},\omega)=\left[\varepsilon(\omega)-b^2(\omega)\left(\frac{Qv_F}{\omega-i\nu}\right)^2\right]\Big/\left[1-b^2(\omega)\left(\frac{Qv_F}{\omega-i\nu}\right)^2\right]. \qquad (4)$$

This function is the hydrodynamic limit of Eq. (3c). The function (4) ensures correct asymptotics of ε at large and at small $\mathbf{Q}$. The coefficient b^2 that depends on the frequency increases with frequency in the interval $\omega_p/\sqrt{2}\lesssim\omega\lesssim\omega_p$. At $\omega=\omega_p/\sqrt{2}$ its value is close to 4/9. At $\omega\approx\omega_p$ we have $b^2\approx 2/5$. It can be verified that the value $b^2(\omega=\omega_p/\sqrt{2}=4/9$ ensures the correct form of that part of the Green's function (obtained by the Wiener-Hopf method) which describes surface plasmons. At $P=0$, i.e., for pure diffuse reflection of the electrons of the medium from the interface, Eqs. (3) can be solved by the Wiener-Hopf method not with the hydrodynamic dielectric constant, but with $\varepsilon(\mathbf{Q},\omega)$ taken in the kinetic approximation and therefore having logarithmic singularities. This solution will be discussed below.

The solution obtained in this manner for Eqs. (3) with dielectric function (4) is of the form

$$\begin{aligned}
D(z,z',\mathbf{q},\omega)&=2\pi\theta(z)\theta(z')\left\{\frac{e^{-q|z-z'|}-e^{-q(z+z')}}{q\varepsilon(\omega)}\right.\\
&-\frac{i[\varepsilon(\omega)-1]}{r\varepsilon(\omega)}[e^{-ir|z-z'|}-e^{-ir(z+z')}]-\frac{2i(r-l)(l+iq)}{(r^2+q^2)(r+iq)}[e^{-qz}-e^{-irz}]\\
&\left.\times[e^{-qz'}-e^{-irz'}]\right\}+\frac{4\pi}{q[1+\Xi(q,\omega)]}\left\{e^{-q|z|}+\theta(z)\frac{iq(l^2-r^2)}{(r+iq)}\right.\\
&\left.\times\frac{(1+P)[e^{-irz}-e^{-qz}]}{[l((1-P)r+l(1+P))-iq((1+P)r+(1-P)l)]}\right\}\left\{e^{-q|z'|}\right.\\
&\left.+\theta(z')\frac{iq(l^2-r^2)(1+P)[e^{-irz'}-e^{-qz'}]}{(r+iq)[l((1-P)r+(1+P)l)-iq((1+P)r+(1-P)l)]}\right\}. \qquad (5)
\end{aligned}$$

In this expression

$$r=\left\{\frac{k^2\varepsilon(\omega)[1-\varepsilon(\mathbf{Q},\omega)]+q^2\varepsilon(\mathbf{Q},\omega)[1-\varepsilon(\omega)]}{\varepsilon(\omega)-\varepsilon(\mathbf{Q},\omega)}\right\}^{1/2},$$

$$l=\left\{\frac{k^2[1-\varepsilon(\mathbf{Q},\omega)]+q^2[1-\varepsilon(\omega)]}{\varepsilon(\omega)-\varepsilon(\mathbf{Q},\omega)}\right\}^{1/2}$$

with the function

$$1+\Xi(\mathbf{q},\omega)=1+\varepsilon(\omega)-\frac{iqv_Fb(1-P)[1-\varepsilon(\omega)]}{2\omega\{1+[1-b^2(qv_F/\omega)^2]^{1/2}\}}$$
$$-\frac{q(1+P)[1-\varepsilon(\omega)]\{q+Z\}}{(1+P)\{(\omega/bv_F)^2-q^2-qZ\}-i(1-P)[(\omega/bv_F)^2-q^2]^{1/2}\{Z-q\}}$$
$$Z=[q^2-(\omega/bv_F)^2\varepsilon(\omega)]^{1/2}.$$

We note incidentally that the condition for the existence of surface plasmons is the vanishing of just this function. In the approximation linear in the spatial dispersion, the frequency of the surface plasmons in a medium with a rough interface is

$$\omega_{surf}=\frac{\omega_p}{\sqrt{2}}\left\{1+\left[\frac{(1+P)^2}{3\sqrt{2}(1+P^2)}+i\frac{\sqrt{2}}{12}(1-P)\frac{3+2P+P^2}{1+P^2}\right]\frac{qv_F}{\omega_p}\right\}. \tag{6}$$

At $P=0$ this formula goes over directly into a previously obtained expression.[7]

The solution (5) of the Dyson equation makes it possible to obtain in accord with (1) the probability of electron scattering with transfer of a momentum $\mathbf{Q}$ and an energy ω to the medium. This probability contains two terms, on describing the probability of electron scattering per unit time with excitation of a plasmon within the volume

$$W_B(\mathbf{Q},\omega)=\frac{8\pi e^2}{\hbar \mathbf{Q}^2 V}\operatorname{Im}\frac{1}{\varepsilon(\mathbf{Q},\omega)}. \tag{7a}$$

The second term describes the excitation of bulk plasmons on the surface. It is convenient to rewrite it in a way that it yields not the scattering probability per unit time, that the probability that the electron crossing the interface emit (because of the existence of this interface) a bulk plasmon. This latter probability will be designated P_S:

$$P_S(\mathbf{Q},\omega)=\frac{8\pi e^2}{\hbar \mathbf{Q}^4 V v_{pz}}\operatorname{Re}\left\{\frac{[\varepsilon(\omega)-\varepsilon(\mathbf{Q},\omega)]^{1/2}}{\varepsilon(\mathbf{Q},\omega)[1-\varepsilon(\omega)]}\Big[(k^2[1-\varepsilon(\mathbf{Q},\omega)]\right.$$
$$+q^2[1-\varepsilon(\omega)])^{1/2}-\frac{(k^2\varepsilon(\omega)[1-\varepsilon(\mathbf{Q},\omega)]+q^2\varepsilon(\mathbf{Q},\omega)[1-\varepsilon(\omega)])^{1/2}}{\varepsilon(\omega)}\Big]$$
$$+\frac{[\varepsilon(\omega)-\varepsilon(\mathbf{Q},\omega)]^{1/2}\{k^2[4\varepsilon(\omega)-\varepsilon(\mathbf{Q},\omega)(1+3\varepsilon(\omega))]}{2\varepsilon(\omega)\varepsilon^2(\mathbf{Q},\omega)[1-\varepsilon(\omega)][k^2\varepsilon(\omega)[1-\varepsilon(\mathbf{Q},\omega)]+q^2\varepsilon}\rightarrow$$
$$\left.\frac{3q^2\varepsilon(\mathbf{Q},\omega)[1-\varepsilon(\omega)]\}}{(\mathbf{Q},\omega)[1-\varepsilon(\omega)]]^{1/2}}\right\}+\frac{32\pi e^2 q}{\hbar Q^4 V v_{pz}}\operatorname{Im}\left\{\frac{\Lambda(k,\mathbf{q},\omega)\Lambda(-k,\mathbf{q},\omega)}{1+\Xi(q,\omega)}\right\}. \tag{7b}$$

Here v_{pz} is the fast-electron velocity component normal to the interface. Expression (7a) describes the "Čerenkov" excitation of the plasmon, wherein all the momentum transferred to the medium goes to the electron subsystem. This means that the only dielectric constant in this term depends on the total momentum $\mathbf{Q}$ transferred to the medium. In (7b) the momentum $Q=(k^2+q^2)^{1/2}$ transferred to the medium is divided in some way between the electron subsystem and the interface. This subdivision of the total momentum transfer is ensured both by direct collisions between the electrons participating in the plasma oscillations and the interface, and by the electric part of the stress tensor. The bulk plasmons are therefore excited near the interface not "according to Čerenkov," and we can speak of transition radiation of bulk plasmons. Since, however, the procedure of calculating the plasmon excitation probability includes summation over all the final states of the electron subsystem, the momentum transferred to the electron subsystem is not directly fixed in the left-hand side of (7b). On the other hand the separation of those terms of the bulk-plasmon excitation probability, which describe their transition radiation, should be based on the fact that these terms contain a dielectric function ε that depends not on the total value of $\mathbf{Q}$, but only on part of it. In our theory, such a function is found to be $\varepsilon(k=iq,q,\omega)=\varepsilon(\omega)$. In (7b) this dielectric constant enters in the first and partly in the second term. The second term contains $\varepsilon(iq,q,\omega)$ in the functions $\Lambda(k,\mathbf{q},\omega)$ and $\Lambda(-k,\mathbf{q},\omega)$, which are given by

$$\Lambda(k,\mathbf{q},\omega)=1-\frac{(1+P)(q-ik)(q^2+k^2)[1-\varepsilon(\omega)][1-\varepsilon(\mathbf{Q},\omega)]}{[\varepsilon(\omega)-\varepsilon(\mathbf{Q},\omega)]\{k+\mathscr{L}_1\}}$$
$$\times\{\mathscr{L}_1[(1-P)\mathscr{L}_2-iq(1+P)]+\mathscr{L}_2[(1+P)\mathscr{L}_2-iq(1-P)]\}^{-1},$$
$$\mathscr{L}_1=\left(\frac{k^2\varepsilon(\omega)[1-\varepsilon(\mathbf{Q},\omega)]+q^2\varepsilon(\mathbf{Q},\omega)[1-\varepsilon(\omega)]}{\varepsilon(\omega)-\varepsilon(\mathbf{Q},\omega)}\right)^{1/2},$$
$$\mathscr{L}_2=\left(\frac{k^2[1-\varepsilon(\mathbf{Q},\omega)]+q^2(1-\varepsilon(\omega))}{\varepsilon(\omega)-\varepsilon(\mathbf{Q},\omega)}\right)^{1/2}. \tag{8}$$

The redistribution of the momentum among the electrons that participate in the oscillations of frequency ω_p and the interface, and also among the electromagnetic field of these electrons and the interface, takes place effectively in a region having a dimension on the order of v_F/ω_p. This applies mainly to momentum exchange between the electrons and the interface on account of the mechanical collisions of the electrons with the surface. Exchange of the electron momentum with the interface on account of the electromagnetic field takes place over the wavelength λ of the plasmon, which usually exceeds v_F/ω_p. With increasing wavelength, however, the momentum carried by the field decreases, so that a certain effective momentum exchange occurs again between the electrons of the medium and the interface. The transition radiation of the bulk plasmon is therefore a spatial-dispersion effect. Correspondingly, the transition probabilities contain combination of the functions ε in the form $[\varepsilon(\omega)-\varepsilon(\mathbf{Q},\omega)]^{1/2}$.

The frequency dependence of P_S is given by Eq. (7b) and is quite complicated in form. It becomes simpler if we go in (7b) to the limit of weak spatial dispersion. Expanding all the functions $\varepsilon(\mathbf{Q},\omega)$ in the real part of (7b) in powers of $\mathbf{Q}$ and retaining only the terms of lowest power, we obtain

$$P_S(\mathbf{Q},\omega)=\frac{8\pi e^2 v_F b(\omega)}{\hbar v_{pz} V Q^2\omega}\operatorname{Re}\left\{\frac{1-[\varepsilon(\omega)]^{1/2}}{[\varepsilon(\omega)]^{1/2}}\right\}. \tag{9}$$

If we neglect the dependence of the coefficient $b(\omega)$ on ω, the spectral distribution of the radiation probability is given by

$$\frac{1}{\omega}\operatorname{Re}\left\{\frac{1-[\varepsilon(\omega)]^{1/2}}{[\varepsilon(\omega)]^{1/2}}\right\}. \tag{9a}$$

If the function $\varepsilon(\omega)$ near the plasma frequency is written in the form $\varepsilon(\Delta)=2(\Delta-i\nu)/\omega_p$ ($\Delta=\omega-\omega_p$ is here the frequency detuning), Eq. (9a) assumes an even more concrete form:

$$2^{-3/2}(\omega_p+\Delta)^{-1}\left(\frac{\Delta^2+\nu^2}{\omega_p^2}\right)^{-3/4}\left\{\left[1-\sqrt{2}\left(\frac{\Delta^2+\nu^2}{\omega_p^2}\right)^{1/4}\right.\right.$$
$$\left.\times\cos\left(\frac{1}{2}\operatorname{arctg}\frac{\nu}{\Delta}\right)\right]\cos\left(\frac{3}{2}\operatorname{arctg}\frac{\nu}{\Delta}\right)-\sqrt{2}\left(\frac{\Delta^2+\nu^2}{\omega_p^2}\right)^{1/4}$$
$$\left.\times\sin\left(\frac{1}{2}\operatorname{arctg}\frac{\nu}{\Delta}\right)\sin\left(\frac{3}{2}\operatorname{arctg}\frac{\nu}{\Delta}\right)\right\}. \tag{9b}$$

This function becomes negative in the narrow frequency region $|\Delta| < \nu$. At all other Δ it is positive. It can be stated that at $|\Delta| > \nu$ this function describes well enough the plasmon transition-radiation spectrum. If $\nu \to 0$, then (9a) goes over into

$$\frac{1}{\omega}\mathrm{Re}\frac{1-[\varepsilon(\omega)]^{1/2}}{[\varepsilon(\omega)]^{3/2}} \to \frac{1}{\omega(2\Delta/\omega_p)^{3/2}}\left[1-\left(2\frac{\Delta}{\omega_p}\right)^{1/2}\right]. \qquad (9c)$$

The reason for the dip in the spectrum as $\Delta \to 0$ is that the momentum transfer cannot be too small, since the transition radiation is due entirely to spatial dispersion. The plasmon localization region at the instant of its production near the interface should not be longer than the path of the electrons of the medium between collisions in the volume. Transition radiation therefore ensures generation of plasmons with wave vectors that satisfy the condition

$$k^2 > A\omega_p\nu/v_F^2, \quad A \sim 1.$$

In the frequency range near $\omega = \omega_p$ the spectrum of the plasmons generated in the transition radiation should therefore have a dip. This frequency range is determined from the condition

$$|\varepsilon(\omega)| < B\nu/\omega_p, \quad B \sim 1.$$

The fact that the probability (9b) at $|\Delta| < \nu$ is not zero but negative is due entirely to our discarding all but the linear term in the expansion of (7b) in powers of Q. The convergence of the series becomes worse as $\Delta \to 0$ because the coefficients of this expansion turn out to contain in the denominators the quantities $\varepsilon(\omega)$ raised to an increasing power as $\Delta \to 0$.

In the particular case when the coefficient P of specular reflection of the medium electrons from the interface is zero, it becomes possible to solve Eq. (3) with a dielectric constant obtained by solving the kinetic equation and given by[8]

$$\varepsilon(\mathbf{Q},\omega) = 1 - \frac{3\omega_p^2}{(\omega - i\nu)^2}\int_1^\infty \frac{dz}{z^2[z^2 - (Qv_F)^2/(\omega - i\nu)^2]}. \qquad (10)$$

The transition probability calculated on the basis of this solution yields a transition-radiation line shape quite close to (9) on the line wings, with a dip in the region $|\Delta| < \nu$, but this dip does not go negative. This result is obtained without going to the limit of weak spatial dispersion.

All this means that the transition-radiation spectrum can be described by Eq. (9) at all $|\Delta| > \nu$. If ν is small, the dip of the spectrum in the region $|\Delta| < \nu$ is most likely not observable in experiment, for it is precisely at $\Delta = 0$ that the bulk plasmons are intensively generated by the "Čerenkov" method. It makes sense therefore to "cut off" the peak of the function (9c) at a height corresponding to the frequency $|\Delta| = \nu$. The plot of the function (9a) modified in this manner is shown in Fig. 1. For comparison, the figure shows also the spectral distribution of the plasmon-radiation probability distribution in a collisionless plasma far from the interface. This latter curve, due to the "Čerenkov" radiation, is described by the formula

$$\frac{1-\varepsilon(\omega)}{\varepsilon(\omega)}\theta\left[\varepsilon(\omega) - \left(b\frac{v_F}{v_p}\right)^2\right], \qquad (11)$$

where θ is the step function. The bulk-plasmon transi-

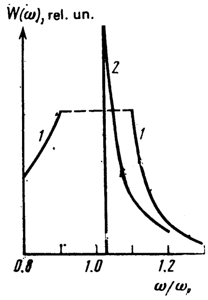

FIG. 1. Line contour of bulk plasma resonance: 1) transition radiation of bulk plasmon in the frequency range $|\omega - \omega_p| > \nu$. It is assumed that $\nu/\omega \ll 1$; 2) Čerenkov radiation of bulk plasmon in a collisionless electron plasma. It is assumed that $E_F/E_p = 0.1$ and $b_2 = 3/5$.

tion radiation probability receives also a contribution from that term of (7b) which contains the factors Λ. A feature of the last term is its dependence on the roughness of the interface. In the limit of weak spatial dispersion this part of the transition probability is given by

$$P_s' = -\frac{128\pi e^2 v_F q^2(1+P)[1-\varepsilon(\omega)]}{3\hbar v_p V\omega Q^4[1+\varepsilon(\omega)]\cos\alpha}\mathrm{Re}\frac{1}{\sqrt{\varepsilon(\omega)}[(1-P)\sqrt{\varepsilon(\omega)}+1+P]}. \qquad (12)$$

It was assumed here that $b^2 = 4/9$. Equation (12) is a correction to expression (9). It is seen from (12) that the transition radiation depends on the degree of surface roughness. In particular, transition radiation of bulk plasmons on a boundary with diffuse scattering of the electrons is somewhat more intense than in the case of specular reflection. The nonconservation of the tangential component of the momentum at the interface contributes to the transition probability which now, as seen from (12), takes the form

$$P_s'(P=0) - P_s'(P=1) = \frac{128\pi e^2 v_F q^2[1-\sqrt{\varepsilon(\omega)}]}{3\hbar v_p Q^4\omega V[1+\varepsilon(\omega)]\cos\alpha}. \qquad (13)$$

The bulk-plasmon transition radiation probability increases with increasing angle α between the electron incidence and the normal to the surface of the solid. This dependence takes the form $1/\cos\alpha$, so long as the Born approximation holds in the interaction of the incident electron with the electron subsystem of the medium, i.e., at $e^2/\hbar v_p\cos\alpha < 1$.

Transition radiation of bulk plasmons can be identified by the dependence, on the angle α, of the ratio of the height of the peak on the spectrum of the electrons that excited the bulk plasmon to the height of the peak of the electrons reflected from the medium without loss of energy, at intermediate incident-electron energies.

3. TOTAL PROBABILITY OF TRANSITION RADIATION OF BULK PLASMONS

Before we compare the total probability of the transition radiation of bulk plasmons with the probability of plasmon radiation far from the interface, we must compare the intensity of the transition radiation with the intensity of the competing bulk-plasmon bremsstrahlung due to the change in the velocity of the electron incident on the medium–vacuum interface. From the energy and momentum conservation laws it follows directly that

$$\mathbf{Q}_{pl}\mathbf{v}_p=\omega_p-\mathbf{Q}_S\mathbf{v}_p.$$

Here $\mathbf{Q}_{pl}$ is the momentum transferred to the bulk plasmon, and $\mathbf{Q}_S$ is the momentum transferred to the interface. From this formula we get

$$Q_{pl}>\frac{\omega_p}{v_p}-\frac{\mathbf{v}_p\mathbf{Q}_S}{v_p}. \tag{14}$$

This condition is valid regardless of whether we are considering transition radiation of bulk plasmons or their bremsstrahlung at the interface. The sign of $\mathbf{Q}_S\cdot\mathbf{v}_p$ in these cases, however, is different. In bremsstrahlung the electron is accelerated when it crosses the interface and Q_S is then of the order of U/v_p, where U is the difference between the potential energies of the electron outside and inside in the medium, while $\mathbf{v}_p\cdot\mathbf{Q}_S<0$, i.e., the components of $\mathbf{v}_p$ and $\mathbf{Q}_S$ normal to the interface are of opposite sign. For bremsstrahlung Eq. (14) therefore takes the form

$$Q_{pl}>\omega_p/v_p+Q_S\cos\alpha.$$

At not too large incidence angles, the second term is of the same order as the first. This means that the longest-wavelength plasmons that can be generated via this mechanism have λ shorter than in the case of "Čerenkov" mechanism. For transition radiation, however, the sign of the product $\mathbf{v}_p\cdot\mathbf{Q}_S$ can generally speaking be arbitrary. The results of the preceding section show that actually $\mathbf{v}_p\cdot\mathbf{Q}_S>0$, so that for transition radiation

$$Q_{pl}>\omega_p/v_p-Q_S\cos\alpha,$$

and the "transition" mechanism can generate also long-wave plasmons, something that neither the "Čerenkov" nor the bremsstrahlung mechanisms can do at all. It follows that a plasmon going from the interface into the interior of the medium "pulls" the interface with it via its electric field, and the momentum transferred thereby to the interface exceeds the momentum (of opposite sign) transferred to the interface by the mechanical collisions of the electrons that take part in the plasma oscillations of the medium with the interface.

We compare now the probability of the transition radiation of the plasmons with the probability of their Čerenkov radiation in the bulk, due to large-angle scattering of the intermediate-energy electrons by the medium and their return to the vacuum. The probability of the transition radiation of the plasmons should be compared with the probability of their Čerenkov radiation in the bulk precisely for large-angle-scattering of the intermediate-energy electrons for several reasons. First, if the electron moves in the volume long enough, no matter how low the probability of the Čerenkov radiation of the plasmon per unit time, it exceeds after a sufficiently long time the intensity of plasmon radiation "on the interface." the transition radiation competes therefore with the Čerenkov radiation in a semi-infinite medium only when the electrons stay in the medium a relatively long time. This is precisely the case of reflection of the electrons from the medium. Second, the transition radiation is a spatial-dispersion effect. Consequently the parameter v_F/v_p that characterizes this dispersion should not be very small, and increases with decreasing energy E_p. The transition radiation can thus not be intense in transmission-type experiments, which are possible only at high electron energies. Finally, transmission-type experiments can be masked by the size effect. We must therefore understand how the transition radiation will manifest itself in large-angle scattering of the electrons in the medium.

As shown in Ref. 9, the spectral density of the radiation intensity (we define radiation intensity as the total probability of radiation during the entire time of interaction of the electron with the medium and, as in the derivation of Eq. (7b), we shall use for it the symbol P_S when speaking of the intensity of the transition radiation, and P_B in the case of Čerenkov radiation in the bulk) of the bulk plasmons excited by the "Čerenkov" mechanism under the assumption that the plasmon-emission and electron large-angle scattering by the lattice take place in succession, can be written in the form

$$P_B^{(bs)}(\omega_p)=Rw_{pl}/\gamma_l. \tag{15}$$

We shall be interested in this intensity principally at $\omega=\omega_p$. The total intensity is connected with (15) by the natural relation

$$P_B^{(bs)}=\int d\omega P_B^{(bs)}(\omega).$$

The superscript (bs) labels backscattered electrons. The factor R in this equation can be regarded as the coefficient of elastic reflection of the electrons from the medium. As shown in Ref. 9, $R=\gamma_{bs}/\gamma l$, where γ_{bs} is the probability of elastic incoherent large-angle scattering of the electron in the medium per unit time and γ_l is the total probability of electron scattering in the medium at any angle per unit time. The quantity γ_l defines in essence the coherence length of an electron moving in the medium. Finally,

$$w_{pl}=\frac{e^2\omega_p}{\pi\hbar v_p\nu}\ln\frac{v_p}{v_F} \tag{16}$$

is the spectral density of the probability of the "Čerenkov" emission of a plasmon in the volume, and ν in this formula is the collision frequency of the electrons of the medium.

The analogous formula for the spectral intensity of the transition radiation of a plasmon followed by elastic backscattering of an electron from the atoms of the medium is

$$P_S^{(bs)}(\omega_p)=\frac{RV}{(2\pi)^3}\int d^3Q\delta(\omega_p-\mathbf{v}_p\mathbf{Q})P_S(\mathbf{Q},\omega_p); \tag{17}$$

here $P_S(\mathbf{Q},\omega)$ is given by (9). The corresponding electron-backscattering intensity in the alternate process, when the electron is first backscattered and a bulk plasmon is then emitted via the "transition" mechanism, can be represented in a form similar to that cited in Ref. 10:

$$P_S^{(bs,a)}=\frac{RV}{(2\pi)^3}\int\limits_{U^{1/2}/(U+E_p)^{1/2}}^{1} d\cos\alpha'\int dQ\delta(\omega_p-\mathbf{v}_p\mathbf{Q})P_S(\mathbf{Q},\omega_p). \tag{18}$$

The lower limit of integration with respect to the angle

α' takes into account the cone in which the electrons are emitted from the medium into the vacuum and due to the surface barrier. The total spectral density of the intensity of emission of a bulk plasmon at a frequency close to ω_p is thus

$$P_{pl}^{(bs)}=R\frac{e^2}{\pi\hbar v_p}\left\{\frac{\omega_p}{\nu\gamma_l}+\left(\frac{3}{5}\right)^{1/2}\frac{v_F\omega_p^{1/2}}{v_p\nu^{1/2}}\right.$$
$$\left.\times\left[\frac{1}{\cos\alpha}+\ln\left(\frac{U+E_p}{U}\right)^{1/2}\right]\right\}\ln\frac{v_p}{v_F}. \qquad (19)$$

In the derivation of this equation we used expression (9c). The first term of (19) is due to the Čerenkov emission of the electrons scattered into the rear hemisphere of solid angles. The next two are due to transition radiation.

4. CONCLUSIONS

In this section we examine how to identify transition radiation in an experiment. It is seen from Eq. (19) that the transition radiation has angular and energy dependences different from those of Čerenkov radiation. Besides the obvious dependence of the term in the curly brackets on v_F and on $\cos\alpha$, the energy dependence and in a certain sense the frequency dependence are also contained in γ_l. The collision frequency γ_l, as it is customarily introduced, characterizes exclusively a bulk process and it might seem that it should be completely independent of α. It must be borne in mind, however, that the probabilities of excitation of bulk and surface plasmons are not independent, just as the cross sections for reactions in different channels are not at all independent. This connection is present also in theories of kinetic type. It manifests itself in particular in the presence of the second term of Eq. (4.2) of Ref. 7. This term causes the following change in the bulk-plasmon excitation probability over the entire time of interaction of the electron with the interface:

$$P_B'=-\frac{4\pi e^2(3q^2-k^2)}{\hbar v_p V Q^4 q\cos\alpha}\operatorname{Im}\frac{1}{\varepsilon(\omega)}. \qquad (20)$$

It is seen from this equation that if $3q^2>k^2$ the increase of the surface-plasmon generation probability leads to a decrease in that of the bulk plasmon. Since the surface-plasmon generation probability depends obviously on the angle, the bulk-plasmon excitation probability acquires an angular dependence. This angular dependence has no bearing on the transition radiation of bulk plasmons, but must be taken into account in the interpretation of the experiments. We carry out for this purpose the following estimates.

It follows from Ref. 7 that the connection, described by Eq. (20), between the bulk and surface reaction channels can be taken into account by introducing in (15) an additional term, after which the latter formula takes the form

$$P_B^{(bs)}(\omega_p)=R\frac{w_{pl}}{\gamma_l}\left(1-\frac{\gamma_s}{\gamma_l\cos\alpha}\right). \qquad (21)$$

The ratio γ_s/γ_l is of the order of $\gamma_l[\omega_p\ln(v_p/v_F)]^{-1}$. Assuming the second term in (21) to be relatively small, we can represent (21) in the form

$$P_B^{(bs)}(\omega_p)=R\frac{w_{pl}}{\gamma_l+\gamma_s/\cos\alpha}. \qquad (22)$$

This means in fact that if the frequency γ_l in (19) is replaced by the sum $\gamma_l+\gamma_s/\cos\alpha$, this will take into account the fact that an increase of the surface-plasmon excitation intensity at electron glancing-angle incidence on the surface can decrease the bulk-plasmon excitation intensity. In a certain sense this means that it is possible to introduce formally an effective electron bulk collision frequency that depends on the angle α. Thus the angle α governs the intensity of the "Čerenkov" excitation and the part of the intensity of the transition radiation of the bulk plasma in the total excitation intensity of the bulk plasmon:

$$P_{pl}^{(bs)}(\omega_p)=R\frac{e^2\omega_p}{\pi\hbar v_p\nu}\left\{\frac{\cos\alpha}{\gamma_s+\gamma_l\cos\alpha}+\left(\frac{3}{5\nu\omega_p}\right)^{1/2}\right.$$
$$\left.\times\frac{v_F}{v_p}\left[\frac{1}{\cos\alpha}+\ln\left(\frac{U+E_p}{U}\right)^{1/2}\right]\right\}\ln\frac{v_p}{v_F}. \qquad (23)$$

The intensity of the "Čerenkov" excitation decreases with increasing incidence angle α, whereas the intensity of the transition excitation increases. Consequently, the existence of an intensity minimum when the angle α is varied is a feature that points to the significance of the transition radiation. Of course, this minimum exists also if, at normal electron incidence, the intensity of the Čerenkov radiation is larger than or commensurate with the intensity of the transition radiation. The cosine of the angle at which this minimum is observed is

$$\cos\alpha_0\approx\gamma_s/\omega_p\left\{\left[\frac{v_p}{v_F}\left(\frac{\nu}{\omega_p}\right)^{1/2}\frac{1}{\ln(v_p/v_F)}\right]^{1/2}-1\right\}\ln\frac{v_p}{v_F}, \qquad (24)$$

with $0<\cos\alpha_0<1$ at

$$E_p>E_F\left\{\frac{1}{4}\left(\frac{\omega_p}{\nu}\right)^{1/2}\ln\frac{E_p}{E_F}+\left[\frac{\gamma_0}{(\nu\omega_p)^{1/2}}+\frac{\omega_p}{16\nu}\ln^2\frac{E_p}{E_F}\right]^{1/2}\right\}^2. \qquad (25)$$

If the transition radiation prevails over the Cerenkov radiation, the radiation intensity depends on the angle simple like $\cos^{-1}\alpha$ and there is no longer any minimum. Assuming by way of estimate $\gamma_l\approx\gamma_0 v_F/v_p$, we find that the transition radiation exceeds the Čerenkov radiation at

$$E_p\leqslant E_F\gamma_0/(\nu\omega_p)^{1/2}\cos\alpha. \qquad (26)$$

It is seen thus that at incidence angles close to normal this characteristic energy is of the order of several Fermi energies. At larger angles α the value of E_p increases so that, say, at $\alpha=80°$ the transition radiation previals over the Čerenkov radiation up to an energy of the order of 200 eV.

In some experimental studies[10-12] attempts were made to observe the angular dependence of the probability of bulk plasmon emission. For a number of reasons (including the lack of a clear understanding of the origin of this angular dependence), they cannot be used for a theoretical analysis. To cast light on the character and the causes of such an angular dependence, preliminary experiments are needed on large-angle scattering of electrons of energies from 50 to 500 eV in polycrystals. Most prominent along all the experimental studies known to us is that of Allié *et al.*,[11] who investigated large angle electron scattering just at intermediate energies

(100–500 eV). Unfortunately, the medium in that study was not a polycrystal but a single crystal, whose anisotropy introduced an additional angular and energy dependence into the radiation probability. The results of that study can still not be unambiguously experiments. Apparently, however, one important conclusion can nevertheless drawn from it. According to Ref. 11, for a normal incidence angle at $E_p > 100$ eV the radiation intensity of the bulk plasmon is a growing function of E_p. This means, in accord with Eqs. (23) and (26), that in this case the plasmons are generated in by the "Čerenkov" mechanism. At $E_p > 500$ eV and at all α (with the probable exception of those close to $\pi/2$), the angular dependence of the intensity is described by the first term of Eq. (23). This agrees with our estimates, according to which the Čerenkov radiation already predominates over the transition radiation at all angles. On the other hand in the energy interval 100–200 eV and at incidence angles $\alpha = 45°$ and $\alpha = 80°$, the intensity is noticeably higher than at normal incidence. The agreement of this interval of E_p with the one expected on the basis of our Eqs. (25) and (26), together with the increase of the intensity with increasing incidence angle, gives grounds for assuming that we are probably dealing here with transition radiation of a bulk plasmon.

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