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# Auxiliary surface polaritons in the region of resonance with oscillations in a transition layer

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It is shown that an auxiliary surface wave is produced in the region of a resonance between a surface polariton and oscillations in a transition layer and is due to spatial dispersion with respect to the parameter  $kd$  ( $k$  is the wave vector of the surface wave and  $d$  is the thickness of the transition layer). The law of dispersion of the surface waves is investigated and their propagation lengths are determined. The additional boundary condition is obtained, corresponding to the case of a dielectric film in the vicinity of the resonance of the surface polariton with the frequency of the longitudinal oscillations in the film.

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## I. INTRODUCTION

The presence of the so-called transition layer on the surface of a medium or on an interface between media influences the dispersion law of the surface polaritons (SP). Since the dispersion of the SP is presently determined by various methods (the method of attenuated total reflection, Raman scattering, and others), this circumstance uncovers new possibilities of studying the physical properties of the surfaces in thin films.<sup>1</sup> The influence of the transition layer is particularly strong when the frequency  $\omega_0$  of the dipole oscillations in the transition layer<sup>11</sup> lands in the SP frequency—restructuring region. As shown earlier<sup>2</sup> (see also Ref. 1), in this case a gap  $\Delta$  is produced in the SP frequency spectrum, with a depth of the order of  $(d/\lambda_0)^{1/2}$ ,  $\lambda_0 = 2\pi c/\omega_0$ ,  $d$  is the thickness of the transition layer. This effect of the splitting of the SP dispersion curve, as well as the square-root dependence of  $\Delta$  on  $d$ , was first observed in Ref. 3 for the IR region of the spectrum in a study of SP propagating along a sapphire surface covered with an LiF film ( $\Delta \approx 20 \text{ cm}^{-1}$  at  $d \approx 100 \text{ \AA}$ ). The width of the gap increases substantially in the visible part of the spectrum.<sup>4</sup> In the last reference, the splitting effect was observed for SP propagating along an aluminum surface coated with silver films ( $d \approx 20 - 60 \text{ \AA}$ ). The splitting  $\Delta$  at  $d = 26 \text{ \AA}$  turns out, in accord with the theory, to be  $\approx 0.4 \text{ eV}$ .

It appears that the resonance of the oscillations in the transition layer with the SP is a rather common phenomenon. In particular, its possible occurrence must be taken into account also in the analysis of the spectra of reflection of light from surfaces of molecular crystals (e.g., anthracene<sup>5</sup>), and also in the study (see Ref. 6) of Fermi resonance with SP.

In view of the foregoing, further study of the dispersion of SP in presence of resonance with oscillations

in the transition layer becomes vital, particularly an analysis of the possible effects brought about by allowance for spatial dispersion. For the nonresonant situation this analysis was carried out in the author's earlier paper<sup>1</sup> (see also Ref. 7, where energy dissipation in the transition layer was taken into account with the aid of a certain model). It was shown,<sup>1</sup> in particular, that in the region of the Coulomb frequency  $\omega_s$  of the surface polariton on the interface with vacuum (the frequency  $\omega_s$  satisfies the condition  $\epsilon(\omega_s) = -1$ ,  $\epsilon(\omega)$  is the dielectric constant of the substrate) the transition layer produces an  $\omega_s(k)$  dependence linear in the wave vector  $k$  of the SP, and this leads to the appearance of an additional surface electromagnetic wave. In the frequency region  $\omega \approx \omega_s$ , however, the damping is large, and should hinder in particular the propagation of precisely this additional surface wave.

We note in connection with the foregoing that for SP propagating along dielectric surfaces, a rather strong damping can occur not only at  $\omega \approx \omega_s$  but also at  $\omega < \omega_s$ , i.e., for the entire region of the SP spectrum. However, for SP propagating along metal surfaces the situation is, generally speaking, different. Inasmuch as for waves of frequency  $\omega \approx \omega_s \approx \omega_p/\sqrt{2}$  ( $\omega_p$  is the frequency of the volume plasmon) the surface-polariton field penetrates markedly into the metal, the SP is strongly damped in this spectral region. In the frequency region  $\omega \ll \omega_p/\sqrt{2}$ , however, the surface-wave field penetrates only insignificantly into the metal, the damping of the SP is weak, and its propagation length turns out to be macroscopically large (on the order of several centimeters, see Ref. 8; a review of later experiments is contained in the book of Ginzburg and the author<sup>9</sup>). As will be shown below, the relative smallness of the damping is preserved in many cases, and in the region of the resonance of the oscillations in the transition layers with the SP, provided that the frequency of these os-

cillations is  $\omega_0 \ll \omega_p/\sqrt{2}$ , and the transition layer is thin enough (see below). Therefore the observation and study of the additional surface waves are apparently easiest to perform precisely under conditions when these waves propagate along surfaces of metals.

## 2. DISPERSION OF SURFACE POLARITONS IN THE REGION OF RESONANCE WITH OSCILLATIONS IN A TRANSITION LAYER

We assume that an isotropic medium (II) with dielectric constant  $\varepsilon(\omega)$  occupies the region of space  $z < 0$ , and borders on vacuum (I) along the plane  $z = 0$ . When account is taken of the transition layer, whose thickness is  $d \ll \lambda$ , where  $\lambda$  is the wavelength, the analysis of the dispersion of the surface waves can be carried out by using, instead of the field boundary conditions that follow from Maxwell's equations on an abrupt boundary, the following boundary conditions on the surface  $z = 0$ , in an approximation linear in  $d/\lambda$  (see also Ref. 1):

$$\begin{aligned} D_n(\text{II}) - D_n(\text{I}) &= i\gamma k_t E_t(\text{I}), \\ E_t(\text{II}) - E_t(\text{I}) &= -i\mu E_n(\text{I}) k_t + ik_0 d [\mathbf{n} \times \mathbf{H}_t(\text{I})], \\ H_t(\text{II}) - H_t(\text{I}) &= -idH_n(\text{I}) k_t - ik_0 \gamma [\mathbf{n} \times \mathbf{E}_t(\text{I})], \\ H_n(\text{II}) - H_n(\text{I}) &= idH_t(\text{I}) k_t, \end{aligned} \quad (1)$$

where  $n$  and  $t$  denote the vector components normal and tangential to the plane  $z = 0$ ,  $k_0 = \omega/c$ , and  $\omega$  is the field frequency.

The phenomenological quantities  $\gamma(\omega)$  and  $\mu(\omega)$  in (1) are determined by the properties of the transition layer. If this layer can be regarded as macroscopic, then  $\gamma = d\bar{\varepsilon}$  and  $\mu = d/\bar{\varepsilon}$  where  $\bar{\varepsilon}(\omega)$  is the dielectric constant of the layer. But if the layer thickness is of the order of the lattice constant, then macroscopic theory must be used to find  $\gamma$  and  $\mu$ . What is important to us here is only that the resonances  $\gamma(\omega)$  and  $\mu(\omega)$  correspond, generally speaking, to different values of the frequencies. For the vicinities of these frequencies it suffices to retain in (1), in the case of sufficiently weak damping, only the resonant terms. In particular, for the frequency regions  $\omega \approx \omega_0$ ,  $\mu(\omega_0) = \infty$  (this is precisely the case corresponding to the experiments of Refs. 3 and 4) we can assume that the only discontinuity occurs in  $E_t$ , with

$$E_t(\text{II}) - E_t(\text{I}) = -i\mu E_n(\text{I}) k_t. \quad (2)$$

Assuming that in the considered frequency region  $\omega \approx \omega_0$  and the dielectric constant is  $\varepsilon(\omega) < 0$ , we have for the surface waves the dispersion law

$$F(\omega, k) = \kappa/\varepsilon + \kappa_t + \mu k^2 = 0, \quad (3)$$

where  $k$  is the two-dimensional wave vector of the surface wave,

$$\kappa = [k^2 - \omega^2 \varepsilon/c^2]^{1/2}, \quad \kappa_t = (k^2 - \omega^2/c^2)^{1/2}.$$

At  $\mu = 0$ , Eq. (3) leads to the known relation

$$k^2 = \frac{\omega^2}{c^2} \frac{\varepsilon(\omega)}{\varepsilon(\omega) + 1}. \quad (4)$$

If, on the other hand,  $\mu \neq 0$ ,  $\mu = -Ad(\omega^2 - \omega_0^2)^{-1}$ , where

$A$  is a positive quantity that depends little on  $\omega$  in the resonance region, then at  $\omega \approx \omega_0$  the dispersion law of the surface waves becomes substantially altered. We consider hereafter, for the sake of argument, the dispersion of surface waves under conditions that correspond to the experiments of Ref. 4. Namely, we assume that the medium II is a metal with plasma frequency  $\omega_p \gg \omega_0$ , that the transition layer is macroscopic and is obtained by depositing on the surface  $z = 0$  a film of another metal with plasma frequency  $\omega_0$ .

Since  $k_F \approx 10^8 \text{ cm}^{-1}$  for electrons on the Fermi surface, such a film can be regarded as macroscopic if the inequality  $k_F d \gg 1$  is satisfied, and this will now be assumed. Assuming, in addition that the normal skin effect takes place, we postulate thereby

$$\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega + i\Gamma)}, \quad \bar{\varepsilon}(\omega) = 1 - \frac{\omega_0^2}{\omega(\omega + i\tilde{\Gamma})}, \quad (5)$$

where  $\Gamma$  and  $\tilde{\Gamma}$  are the electron collision frequencies in the metal II and in the transition layer. Substitution of expressions (5) in (3), with account taken of the fact that  $\mu = d/\bar{\varepsilon}$ , leads to a relation that enables us to determine the dispersion of the surface waves in the presence of damping.

We are interested here only in a situation wherein the frequency of the surface wave is a real quantity specified by the surface-wave pump source. Under these conditions the quantity  $k = k' + ik''$  is complex, and the surface-wave propagation length is  $L = (2k'')^{-1}$ . If the damping of the surface wave is weak enough (i.e., if  $k' \gg k''$ ) then in first order approximation the damping can be completely neglected in the derivation of the dispersion law. This law, i.e., the function  $\omega_s(k)$ , is then determined from the equation

$$F(\omega) = -cdk^2 \omega^2 (\omega^2 - \omega_0^2)^{-1}, \quad (6)$$

where

$$F(\omega) = \frac{\omega^2 (k^2 c^2 + \omega_p^2 - \omega^2)^{1/2}}{\omega^2 - \omega_p^2} + (k^2 c^2 - \omega^2)^{1/2}. \quad (6a)$$

If  $\omega_{0s}(k)$  is the frequency of the surface polariton on the abrupt boundary (i.e., under conditions when the presence of the transition layer is disregarded), then  $F(\omega_{0s}^2) = 0$  and in the frequency region  $\omega \approx \omega_{0s}(k)$  we have

$$F = (dF/d\omega^2)_0 [\omega^2 - \omega_{0s}^2].$$

From relation (6a) it follows that at  $\omega^2 \ll \omega_p^2$  and  $k^2 c^2 \ll \omega_p^2$  we get

$$F(\omega^2) = -\omega^2/\omega_p^2 + (k^2 c^2 - \omega^2)^{1/2},$$

so that

$$\omega_{0s}^2(k) \approx k^2 c^2 - k^2 c^4/\omega_p^2 + \dots, \quad (7a)$$

$$(dF/d\omega^2)_0 \approx -\omega_p/2\omega_{0s}^2 \quad (7b)$$

and consequently

$$F(\omega^2) \approx -\omega_p [\omega^2 - \omega_{0s}^2(k)]/2\omega_{0s}^2(k). \quad (8)$$

Thus, Eq. (6) can be written for the considered frequency region  $\omega \approx \omega_0 = \omega_{0s}(q_0)$  in the form

$$(\omega^2 - \omega_0^2) [\omega^2 - \omega_{0s}^2(k)] = 2cdk^2 \omega^2 \omega_{0s}^2(k) / \omega_p.$$

Solving this equation for  $\omega^2$ , we get two solutions  $\omega_{1,2}^2(k)$ :

$$\omega_{1,2}^2(k) = \frac{1}{2} [\omega_0^2 + \omega_{0s}^2(k) + 2cdk^2 \omega_{0s}^2(k) / \omega_p] \pm \frac{1}{2} \{ [\omega_0^2 - \omega_{0s}^2(k)]^2 + 4cdk^2 \omega_{0s}^2(k) [\omega_0^2 + \omega_{0s}^2(k)] / \omega_p \}^{1/2}. \quad (9)$$

We have left out under the square-root sign in (9) the relatively small terms proportional to  $k^2 d^2$ .

At  $k = q_0$ , where the frequency  $\omega_{0s}(k) = \omega_0$ , a second splitting of the branches takes place. In fact, at  $k = q_0$  at the frequencies  $\omega_{1,2}(q_0)$  we get

$$\omega_{1,2}(q_0) \approx \omega_0^2 [1 \pm (2cdq_0^2 / \omega_p)^{1/2}],$$

so that the size of the gap  $\Delta \equiv \omega_1(q_0) - \omega_2(q_0)$  is determined by the relation

$$\Delta \approx \omega_0 (2d\omega_0^2 / c\omega_p)^{1/2}, \quad (10)$$

or in terms of wavelengths<sup>20</sup>

$$d\lambda / \lambda_0 = 2(\pi d / \lambda_p)^{1/2} (\lambda_p / \lambda_0), \quad (10a)$$

where  $\lambda_p = 2\pi c / \omega_p$ ,  $\lambda_0 = 2\pi c / \omega_0$ .

Besides the gap  $\Delta = \omega_1(q_0) - \omega_2(q_0)$ , we can introduce also the quantity  $\Delta_1 = \omega_1(\min) - \omega_2(\max)$ . In this relation  $\omega_1(\min)$  is the minimal frequency on the lower branch and corresponds to that value  $k_{\min} = \omega_{\min} / c$ , where the upper branch comes up against the asymptote  $\omega = ck$  [see (7a)]. It follows from (6) that at  $\omega_0 \ll \omega_p$

$$\omega_1(\min) - \omega_0 \approx \frac{d}{2c} \omega_0 \omega_p.$$

We note also that  $\Delta_1 < \Delta$  (see Fig. 1). The gap decreases with increasing  $\omega_0$ , namely  $\Delta \sim \omega_p^{-1/2}$ . The reason for the latter is that when  $\omega_p$  is increased the electric field intensity at  $z \approx 0$  decreases and accordingly the interaction of the wave with the transition layer decreases. If, on the contrary,  $\omega_p \rightarrow 0$  (this case obviously corresponds to a metallic film in vacuum), Eq. (6) takes the form

$$2(k^2 c^2 - \omega^2)^{1/2} = cdk^2 \omega^2 (\omega_0^2 - \omega^2)^{-1/2}. \quad (11)$$

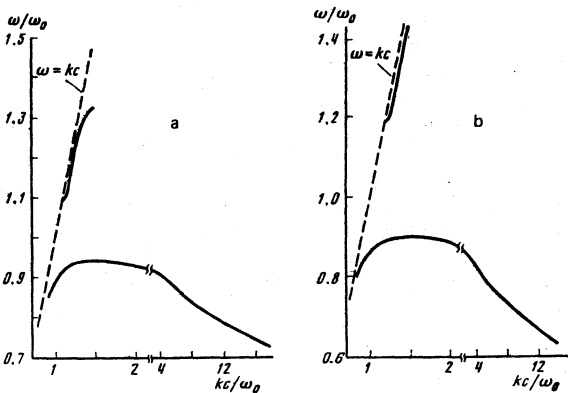


FIG. 1. Dispersion of surface polaritons propagating along an aluminum surface ( $\hbar\omega_p \approx 15.8$  eV) coated with a silver film ( $\hbar\omega_0 \approx 3.8$  eV,  $y_p = \omega_p^2 / \omega_0^2 \approx 15$ ), at different values of the parameter  $\alpha = \omega_0 d / c$ : a— $\alpha = 0.05$ , b— $\alpha = 0.1$ .

It follows from this equation that the nonradiative surface waves discussed here occur only for values of  $\omega$  and  $k$  such that  $kc > \omega$ ,  $\omega < \omega_0$ . There is no splitting of the surface-wave spectrum. As to the function  $\omega(k)$ , it is the same here as for two-dimensional systems (see Refs. 10 and 11).

We return, however, to the analysis of the dispersion of surface waves in the case  $\omega_p \gg \omega_0$ . We note first that at  $k \gg q_0$  when  $kc \gg \omega_p$ , i.e., in the nonrelativistic limit, Eq. (6) takes the simpler form

$$\frac{\omega_p^2 - 2\omega^2}{\omega^2 - \omega_p^2} = \frac{\omega^2 kd}{\omega^2 - \omega_0^2}. \quad (12)$$

It must be borne in mind that this nonrelativistic equation is valid only if the inequality  $\omega_p d / c \ll 1$  holds. Only in this case does the transition in (6) to the nonrelativistic limit not contradict the inequality  $kd \ll 1$  used to write down the boundary conditions (1).

It follows from (12) that for the upper frequency branch, at large  $k$  (see also Refs. 1 and 7)

$$\omega_1(k) \approx 2^{-1/2} \omega_p (1 + 1/2 kd), \quad (13a)$$

whereas for the lower branch

$$\omega_2(k) \approx \omega_0 (1 - 1/2 kd). \quad (13b)$$

The fact that for the lower SP frequency branch we have a linear dispersion law with a negative slope leads in the frequency region  $\omega \leq \omega_0$  to the appearance of an additional (see below) surface waves. In this frequency region we have not one but two SP, having the same frequency but different wave vectors.

The polariton dispersion law for the considered case (metallic film on bulk metal), without allowance for the damping, is shown in Fig. 1 for different values of the parameters  $\alpha \equiv \omega_0 d / c$  and  $y_p = \omega_p^2 / \omega_0^2$ . We note that for aluminum coated with a silver film we have  $y_p \approx 15.2$  and  $\omega_0 / c = 2 \cdot 10^5$  cm<sup>-1</sup>. For an LiF film on a silver substrate,  $y_p = 45$  and  $\omega_0 / c = 4.2 \cdot 10^3$  cm<sup>-1</sup> (see Fig. 2). In the already cited Ref. 4, the gap in the polariton spectrum for the Ag/Al pair was investigated for films with parameters  $\alpha = 5 \cdot 10^{-2}$ ,  $8 \cdot 10^{-2}$  and  $12 \cdot 10^{-2}$ .

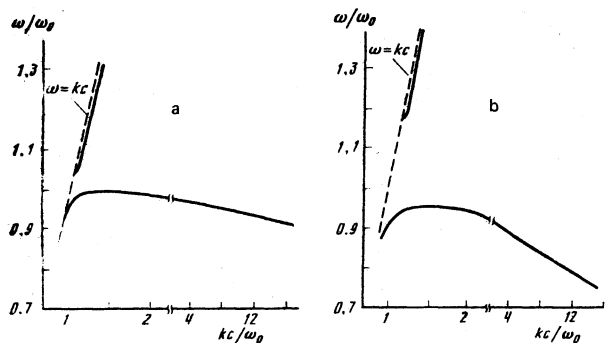


FIG. 2. Dispersion of surface polaritons propagating along a silver surface coated with a LiF film in the vicinity of the frequency  $\omega_0 \approx 670$  cm<sup>-1</sup> (we used for the dielectric constant of LiF the relation  $\epsilon(\omega) = 1.9(\omega^2 - \omega_0^2)(\omega^2 - \omega_1^2)^{-1}$ ,  $\omega_1 = \omega_0 = 670$  cm<sup>-1</sup>,  $\omega_1 = 310$  cm<sup>-1</sup>), at various values of the parameter  $\alpha = 0.4\omega_0 d / c$ : a— $\alpha = 0.01$ , b— $\alpha = 0.05$ .

### 3. DAMPING OF SURFACE POLARITONS IN THE REGION OF RESONANCE WITH THE TRANSITION LAYER AND THE AUXILIARY BOUNDARY CONDITION

When account is taken of the energy dissipation in the substrate and in the film, the quantities  $\varepsilon$  and  $\mu$  in (3) are [see (5)] complex even for real  $\omega$ :  $\varepsilon = \varepsilon' + i\varepsilon''$ ,  $\mu = \mu' + i\mu''$ . In this case we can determine with the aid of (3) the real and imaginary parts of  $k = k' + ik''$  as functions of  $\omega$ . Since the electron collision frequencies in the metal are  $\Gamma \approx 10^{14}$  and  $\bar{\Gamma} \approx 10^{14} \text{ sec}^{-1}$  (see Ref. 12), we have  $\omega \sim \omega_0 \sim 10^{15} \text{ sec}^{-1}$ ,  $\Gamma/\omega \ll 1$ ,  $\bar{\Gamma}/\omega \ll 1$ , for the considered frequency region, so that  $|\varepsilon'| \gg \varepsilon''$  and  $|\bar{\varepsilon}'| \gg \bar{\varepsilon}''$ . This means that to find the values of  $k''$  in this case we can use<sup>3</sup> the first-order approximation in  $\varepsilon''$  and  $\bar{\varepsilon}''$ . Taking the foregoing into consideration, we obtain from (3)

$$k''(\omega) = -\frac{k^2 \mu'' - \varepsilon''(\kappa/\varepsilon^2 + \omega^2/2c^2 \varepsilon \kappa)}{k(1/\varepsilon \kappa + 1/\kappa_1 + 2\mu)} \quad (14)$$

but now the quantity  $k = k(\omega)$ , determined by the dispersion law (3), (6), as well as the values of  $\varepsilon$  and  $\mu$ , must be regarded as real (with  $\Gamma = \bar{\Gamma} = 0$ ).

At  $\mu = 0$ , i.e., under conditions when the transition layer is not taken into account

$$k_0'' = \frac{\varepsilon''(\kappa/\varepsilon^2 + \omega^2/2c^2 \varepsilon \kappa)}{k(1/\varepsilon \kappa + 1/\kappa_1)} \quad (15)$$

and when account is taken of (5), at frequencies such that  $\omega_p \gg \omega \gg \Gamma$ , we have

$$k_0''(\omega) = \frac{\Gamma}{2c} \left( \frac{\omega}{\omega_p} \right)^2.$$

If the transition layer is considered in the frequency region  $\omega \leq \omega_0$ , each value of  $\omega$  corresponds, as already mentioned (see Figs. 1 and 2) to two SP (ordinary and auxiliary), and relation (14) enables us to obtain for them the corresponding damping length  $L$ . We estimate first the value of  $k''(\omega)$  corresponding to the auxiliary solution, bearing in mind first that frequency region  $\omega < \omega_0$  where the nonrelativistic approximation is already suitable. Inasmuch as in this spectral region  $\kappa \approx \kappa_1 \approx k$ , we obtain from (14), taking the inequality  $|\varepsilon| \gg 1$  into account

$$k_2''(\omega) = -1/3 \mu''(\omega) k^2 + \varepsilon'' k / 3 \varepsilon^2$$

and, using (5) and (13b), we get

$$k_2''(\omega) \approx \Gamma/4 \omega_0 d. \quad (16)$$

In this case of extremely large  $k$  (but still  $k \ll 1/d$ ) and  $\omega < \omega_p$ , the SP field hardly penetrates into the metal. It is therefore not surprising that (15) does not contain the characteristics of the substrate and that this relation, apart from a factor of 2, can also be obtained (when damping is taken into account) for a metallic foil in vacuum from the dispersion relation (11) taken at  $k \gg \omega/c$ . At  $\bar{\Gamma}/\omega_0 \approx 3 \cdot 10^{-2}$  and  $d = 30 \text{ \AA}$ ,  $k_2'' \approx 2 \cdot 10^4 \text{ cm}^{-1}$ , which corresponds to propagation lengths  $L$  of the order of tenths of a micron.

To estimate the propagation lengths of both the ordinary

and the auxiliary polariton, which correspond to small  $k$ , it is necessary to use the general relation (14). In dimensionless quantities this relation can be rewritten in the form

$$\begin{aligned} k'' &= \omega_0 A(x, y)/cB(x, y), \\ A(x, y) &= \alpha \bar{\nu} x y^2 (y-1)^{-2} + \nu y_p y^{-2} \varepsilon^{-1} [(x-\varepsilon y)^2 \varepsilon^{-1} \\ &\quad + y/2(x-\varepsilon y)^2]^{1/2}, \\ B(x, y) &= x^{1/2} \{ e^{-1}(x-\varepsilon y)^{-1/2} + (x-y)^{-1/2} + 2\alpha y(y-1)^{-1} \}, \end{aligned} \quad (14a)$$

where

$$\begin{aligned} x &= c^2 k^2 / \omega_0^2, \quad y = \omega^2 / \omega_0^2, \quad \nu = \Gamma / \omega_0, \quad \bar{\nu} = \bar{\Gamma} / \omega_0, \\ \alpha &= \omega_0 d / c, \quad \varepsilon = 1 - y_p / y. \end{aligned}$$

Since  $y_p \gg 1$  by assumption, in the region  $x \geq 1$  and  $y \approx 1$  (where  $|\varepsilon(y)| \approx y_p/y \gg 1$ , the expressions for  $A(x, y)$  and  $B(x, y)$  take the following simpler form

$$\begin{aligned} A(x, y) &\approx \alpha \bar{\nu} x y^{1/2} (y-1)^{-2} + \nu (y/y_p)^{1/2}, \\ B(x, y) &\approx x^{1/2} \{ (x-y)^{-1/2} + 2\alpha y (y-1)^{-1} \}. \end{aligned}$$

For example, at  $\alpha = 10^{-2}$  and  $y_p = 45$  (see Fig. 2a,  $\bar{\nu} = 10^{-2}$ ,  $\nu = 0.27$ ) the value  $y = 0.98$  ( $\omega = 0.98\omega_0$ ) corresponds to  $x = 1.1$  (ordinary wave) and  $x = 1.9$  (auxiliary wave). In this case we have for the ordinary wave  $k_1'' = \omega_0/16c$ , and for the auxiliary one  $k_2'' = \omega_0/10c$  and  $L_1 \approx L_2$ . On the other hand if  $y = 0.89$ , then  $x = 0.9$  and  $x_2 = 20$ , so that  $k_1'' = 10^{-3}\omega_0/c$ ,  $k_2'' = 0.2\omega_0/c$ . Consequently, with increasing distance from resonance, the meanfree path of the additional polariton is greatly shortened and  $L_1/L_2 \approx 200$  in our case (i.e., at  $y = 0.89$ ).

The presented estimates of the SP propagation lengths indicate that, just as in the case of volume polariton, the observation of the auxiliary wave calls for great efforts and can be realized only by choosing suitable substrates and films.

In the case of metal surfaces, thin dielectric films decrease the SP free path only insignificantly, even in the region of the resonance in the film (see Ref. 13). It appears that it is precisely under such conditions that one should search also for auxiliary surface waves.

Before we proceed to discuss the problem of the auxiliary boundary conditions (ABC), we make one remark concerning the damping of surface waves in the case of metallic transition layers (metallic films). In the region of very small thicknesses, metallic films are usually not solid but have an island-like structure. Then, besides with the wave damping due to the non-hermiticity of the dielectric tensor, the Landau damping mechanism can become significant in some cases.

As shown by Lozovik and Nishanov,<sup>14</sup> for metallic drops of radius  $R$  the Landau damping leads to a broadening

$$\Gamma \sim \varepsilon^2/R,$$

so that at  $R \sim 10 \text{ \AA}$  the value of  $\Gamma$  is of the order of an electron volt. This means apparently that in experiments<sup>4</sup> performed at very small silver-coating thicknesses ( $d = 2 \text{ \AA}$  and  $d = 6 \text{ \AA}$ ) no gap was observed in the SP spectra because under these conditions the films were not solid but consisted of islands with characteristic dimensions of the order of the coating thickness.

It has already been emphasized<sup>1</sup> that to observe the optical effects due to the additional surface wave one can use SP excitation by diffraction of light (say from a laser) by a wedge. This excitation method was recently (see Ref. 15) realized for the IR band using a metallic wedge. Development of experiments of the type described in Ref. 15 will possibly permit also the study of effects due to interference between auxiliary and ordinary surface waves having the same frequency.

To find the field amplitudes with account taken of the auxiliary surface wave, the ordinary boundary conditions are no longer sufficient, and the ABC problem arises just as in three-dimensional crystal optics.

The form of the ABC should, generally speaking, depend on the type of the film and on the character of those dipole oscillations in it that lead to resonance with the SP. We confine ourselves therefore from now on to a discussion of the ABC for the case considered in Ref. 2 in the derivation of the dispersion relation (3).

We note first that the correction to the boundary condition (2) is necessitated by the polarization of the film along a direction perpendicular to the film. Therefore to find the ABC in the case when the film is dielectric it can be assumed that the dipole moment of the transition is directed in the film along the  $z$  axis. Since the variation of the field over the film thickness, in the approximation linear in  $d/\lambda$ , is disregarded, the film can be taken to be two-dimensional. If such a two-dimensional system (two-dimensional crystal) is bounded along the  $x$  axis and if we disregard the deformations of the edge molecules then, just as in the three-dimensional case (see Ref. 16), the boundary conditions at  $x=0$  can be taken for the polarization to be

$$P_x(x=0)=0, \quad (17)$$

where  $\mathbf{P}$  is the film polarization per unit area.

Satisfaction of condition (17) means that at  $x \approx 0$  the film can not lead to a discontinuity of  $E_t$  of the form (2), inasmuch as at  $x=0$  we have

$$E_t(\text{II}) - E_t(\text{I}) = -ik_t \int_0^d E_n(z, x=0) dz, \quad (18)$$

where  $E_n(z, x=0)$  is the normal component of  $E$  in the film, and since at  $x=0$  we have in the film  $P_n \approx 0$  [see (17)], we arrive at the conclusion that  $E_n(z, x=0) \approx D_n(z, x=0)$ , and consequently

$$\int_0^d E_n(z, x=0) dz = \int_0^d D_n(z, x=0) dz = dD_n(\text{I}, x=0) = dE_n(\text{I}, x=0).$$

Thus, at  $x \approx 0$  no term of order  $\mu = d/\bar{\epsilon}$  resonant at  $\bar{\epsilon} = 0$  appears in the right-hand side of (18). Since the nonresonant terms were omitted when (2) was derived from (1), it must be assumed that at  $x \approx 0$

$$E_t(\text{II}, x=0) - E_t(\text{I}, x=0) = 0. \quad (19)$$

Comparing now (19) and (2) we conclude that the condition (17) for the polarization in the film leads to the sought ABC in the form

$$E_n(\text{I}, x=0) = 0. \quad (20)$$

Since  $E(\text{I}) = D(\text{II})$ , relation (20) can be replaced by the equivalent condition

$$D_n(\text{II}, x=0) = 0. \quad (20a)$$

We can obtain in similar fashion the ABC  $E_t(\text{I}, x=0) = 0$  for dielectric films in the region of resonance of  $\bar{\epsilon}(\omega)$ . If, however, this resonance is due to a two- or three-dimensional Wannier-Mott exciton, then it may be important to introduce a dead layer of thickness  $l \sim r_B$  where  $r_B$  is the Bohr radius of the exciton, in analogy with procedure used in the theory of ABC for volume waves<sup>17</sup> (see also Ref. 9).

The question of the form of the ABC for metallic films calls for a separate study.

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- <sup>1</sup>Layers of this kind can be produced also artificially, for example by depositing very thin films on various substrates.
- <sup>2</sup>The damping was not taken into account in the determination of  $\Delta$ . Therefore relations (10) are valid if  $\Delta$  is large compared with the width of the polariton spectral line.
- <sup>3</sup>Estimates show that an analogous situation is realized in most cases also for dielectric films and dielectric substrates. (e.g., LiF films on sapphire).
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