

# Dispersion law of surface plasmons

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A study is made of the reason for the appearance of a nonanalytic linear term in the dispersion law of surface plasmons at a metal–vacuum boundary. A model of an unsharp boundary makes it possible to calculate explicitly the coefficient in front of the linear term. In this model the unsharp boundary of the metal is a transition layer whose thickness is large compared with the Debye screening radius, so that the hydrodynamic approximation can be used. The imaginary part of the linear term is associated with processes such as the decay of plasmons at the surface into single-particle excitations, which is allowed for (in the model employed) by introducing an inelastic reflection coefficient.

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## 1. INTRODUCTION. FORMULATION OF THE PROBLEM

It is well known (see, for example, the reviews in Refs. 1 and 2) that the dispersion law of surface plasmons

$$\omega_s(k) = 2^{-1/2} \omega_p [1 + A|k| + Bk^2 + \dots]$$

shows nonanalytic behavior manifested by a linear dependence of the frequency on the modulus of the wave vector. (Here,  $\omega_p$  is the plasma frequency and  $k$  is the wave vector in the surface plane. The coefficient  $B$  represents the usual quadratic dispersion of plasma waves, which follows from the continuum theory and which we shall not consider here. The coefficient  $A$  is generally complex and it represents the anomalous linear dispersion.)

Much work has been done on the origin of the linear term and on calculation of the coefficient  $A$ . In the majority of cases this has been done on the basis of the well-known general expression for the polarizability of an inhomogeneous electron gas in the random phase approximation (see, for example, Kittel's book<sup>3</sup>). Calculation of the coefficient  $A$  then requires self-consistent computation of the electron density on the boundary, which is an important and independent task. In other cases the linear plasmon dispersion is calculated for a given distribution of the surface electron density, which makes it possible to complete the numerical calculations. We shall not review these investigations but simply note that there has been a tendency to lose the physical picture in these detailed calculations and that it is desirable to elucidate some general aspects of the situation using a simpler model. In particular, the following questions are of importance:

1) the physical and mathematical mechanisms responsible for the nonanalyticity associated with the linear terms;

2) the factors governing the value of the coefficient  $A$  and, in particular,

2a) the physical damping processes represented by the imaginary part of the coefficient  $A$ .

We shall point out immediately that the model used

below is of limited validity and, moreover, the answer to the question 2a) obtained in this model can only be partial and based on certain assumptions. Nevertheless, it seems that the discussion given below is of some interest because the model employed makes it possible to carry out a detailed quantitative analysis and to provide a simple physical interpretation, which may be useful in dealing with more realistic situations.

The adopted model is as follows. It is assumed that the ion masses are infinite and the distribution of the positive charge of the ions can be specified arbitrarily so that the distribution of the electron density is deduced from the solution of self-consistent equations (for example, in the random phase approximation). If, as is the case in a real metal, the ion density terminates abruptly, the electron density falls over distances of the order of the Debye radius it is difficult to determine the nature of this fall. On the other hand, we can imagine a situation when the ion density falls in a distance  $d$  much greater than the Debye radius. Then, the scale of the inhomogeneity of the electron density distribution is of the same order of magnitude, i.e.,  $d \gg r_D$ .

It may be that this model is not without some justification in the case of gaseous plasmas or inhomogeneously doped semiconductors, but we shall use this model only to deal with the questions stated above.

The model is convenient because it allows us to use the hydrodynamic approximation subject to the condition

$$\lambda \gg d \gg r_D, \quad (1)$$

where  $\lambda = 2\pi k^{-1}$  is the wavelength along the surface. However, it should be pointed out that the hydrodynamic approach is unacceptable on an outer boundary of a metal in a region of the order of  $r_D$ , where the velocity of sound vanishes. In our model the influence of this layer can be allowed for by a single quantity, which is the complex reflection coefficient  $r$ . It can be calculated by the kinetic approach and we hope to carry out such a kinetic analysis later.

The imaginary part of the coefficient  $A$  differs from zero for  $|r| < 1$ . The nature of the inelastic processes responsible for the deviation of the reflection coefficient from unity cannot be discussed in the hydrodynamic

approximation but—as shown below—the coefficient  $r$  can be interpreted as the reflection coefficient of bulk plasmons from a metal–vacuum boundary. We shall assume that the inelastic nature of this reflection is associated with the decay of plasmons into electrons and holes in collisions with the boundary. In the bulk case this process represents the Landau damping and occurs only on condition  $q \geq \omega/v_F$ , where  $q$  is the plasmon wave vector and  $v_F$  is the Fermi velocity, which is associated with the need to satisfy the laws of conservation of energy and momentum simultaneously. However, when a plasmon collides with the surface, the momentum may not be conserved and the decay described above may take place. We shall assume that this process is the principal contribution to the value of  $A$  and it is implicitly allowed for in the numerical calculations based on the random phase approximation.

## 2. PHENOMENOLOGICAL DERIVATION OF THE DISPERSION LAW OF SURFACE PLASMONS

The origin of the nonanalyticity in the spectrum of surface plasmons can easily be understood from simple physical considerations bearing in mind the different nature of the Coulomb forces on both sides of the boundary. In the metal the Coulomb interaction is weakened by the Debye screening but outside the metal (in vacuum) the interaction remains of the long-range type. An inhomogeneity of the medium associated with the boundary invalidates the usual continuum approach and it gives rise to discontinuities of the potential and electric induction proportional to the wave vector.

General considerations of this type were developed by Agranovich in connection with a study of surface polaritons.<sup>4</sup> We shall use these considerations to show how the unsharp nature of the boundary gives rise to nonanalyticity. We shall consider the behavior of the potential and induction near the surface transition layer and we shall derive the appropriate boundary conditions. The usual boundary conditions, specifying continuity of the potentials and normal components of the induction, are in this case insufficient. The point is that, in the presence of a transition layer, additional surface currents and polarization of the surface region are observed. Allowance for these currents and polarization and the corresponding derivation of the boundary conditions can be made microscopically but for our purpose it is sufficient to obtain the boundary conditions directly by integrating the macroscopic equations.

We shall consider a metal–vacuum boundary characterized by a decreasing electron density  $n(z)$ , as shown in Fig. 1. We shall identify the quantities referring to vacuum by the minus subscript and those to the metal by the plus subscript. We shall represent the coordinate dependences of all the quantities in the form  $\exp(kz + ik \cdot \rho)$ , where  $\rho$  is a two-dimensional vector in the boundary plane. We shall introduce the electric field potential  $E = -\nabla\phi$ . In the macroscopic approach we shall replace a surface transition layer with an infinitely thin surface located at the origin  $z=0$ .

The potential  $\phi$  and the function  $D$  in vacuum (–) and in the metal (+) can be represented in the form

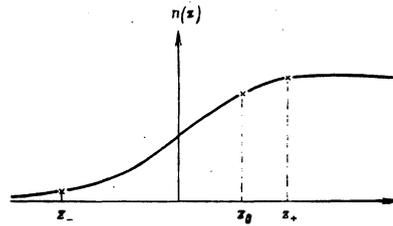


FIG. 1.

$$\phi_- = e^{kz}, \quad \phi_+ = B_+ e^{kz} + B_- e^{-kz}, \quad (2)$$

$$D_- = -\epsilon_- \frac{d\phi_-}{dz} = -k\epsilon_-, \quad (3)$$

The field amplitude in vacuum is selected to be unity and allowance is made for the fact that the field in vacuum decreases in the limit  $z \rightarrow -\infty$ ;  $B_-$  and  $B_+$  are the amplitudes of the field in the metal representing the decreasing and increasing components, respectively.

The presence of the transition layer can be allowed for by the following boundary conditions

$$\phi_+ - \phi_- = - \int_{z_-}^{z_+} E_z dz = -\alpha D_+ + \alpha D_- + O(k^2), \quad (4)$$

$$D_+ - D_- = ik\beta E_t + O(k^2), \quad (5)$$

where  $\alpha$  and  $\beta$  are phenomenological coefficients relating the distribution of the field  $E_z$  normal to the layer and of the longitudinal induction  $D_z$  inside the layer with the normal component of the induction  $D_z$  and the tangential field  $E_t$ :

$$\int E_z(z) dz = \alpha D_z, \quad \int D_z(z) dz = \beta E_t. \quad (6)$$

Integration is carried out here over the thickness of the transition layer.

The condition (4) is obtained from (6) by the substitution  $E_z = -d\phi/dz$ , whereas the condition (5) is found from (6) using the continuity equation

$$\frac{\partial D_z}{\partial z} + ikD_t = 0.$$

The following clear interpretation of the conditions (6) can be applied: if we introduce the displacement current  $I$  in accordance with the formula  $D = I/i\omega$ , we find that the conditions (6) correspond to the transition layer representation in the form of an infinitely thin conducting sheet of resistivity  $\rho_1 = \alpha/i\omega$  in the normal direction and a surface conductivity  $\sigma_t = i\omega\beta$  in the tangential direction.

Substituting now the expressions (2) and (3) in the boundary conditions (4) and (5) and using also the relationship  $E_t = -d\phi/d\rho = -ik$ , we obtain

$$\left. \begin{aligned} B_+ - B_- - \epsilon_-/\epsilon_+ &= -k\beta/\epsilon_+, \\ B_+ + B_- - 1 &= \alpha k\epsilon_-. \end{aligned} \right\} \quad (7)$$

The dispersion equation of surface plasmons is obtained by equating to zero the amplitude  $B_+$  of the field component which increases with depth in the metal:

$$2B_+ = 1 + \epsilon_-/\epsilon_+ + (\alpha\epsilon_- - \beta/\epsilon_+)k = 0. \quad (8)$$

If the permittivity of vacuum is  $\epsilon_- = 1$  and that of the metal is  $\epsilon_+ = 1 - \omega_p^2/\omega^2$ , the final expression for the spectrum of surface plasmons is

$$\omega = 2^{-1/2} \omega_p (1 + A|k|) + O(k^2), \quad (9)$$

where  $A = (\alpha + \beta)/4$  and we have allowed for the fact that the permittivity of the metal at the surface plasmon frequency is minus unity.

It thus follows that the linear term in the plasmon spectrum can be obtained even from the phenomenological equations if the existence of the transition layer is allowed for in the long-wavelength approximation. However, the boundary conditions give nothing more and the quantity  $A$  is defined only in respect of its order of magnitude. Further analysis requires consistent solution of the appropriate equations in vacuum, in the transition layer, and in the metal; the solutions then have to be matched. The hydrodynamic equations are employed to describe the transition layer because of the assumption that the dimensions of this layer are large compared with the Debye radius. We shall assume that the transition layer is thin compared with the plasmon wavelength, so that all the quantities can be expanded in terms of the small parameter  $kd$ . This method has been used earlier to investigate potential surface waves in a plasma<sup>5</sup> and surface plasmons considered here are a special case of this. A calculation is made of the damping decrement of surface waves proportional to  $kd$  and associated either with total reflection of the wave at the point  $\varepsilon(\omega, r) = 0$  or with allowance for collisions.

We shall now consider in greater detail the quantity  $A$  and we shall calculate it.

### 3. EQUATIONS IN THE PRESENCE OF A TRANSITION LAYER AND SURFACE PLASMON SPECTRUM

The complete system of equations for our problem consists of the Poisson equation

$$\nabla^2 \varphi = -4\pi ne, \quad (\nabla E = 4\pi ne, \quad E = -\nabla \varphi), \quad (10)$$

the continuity equation

$$-i\omega n + \nabla j = 0 \quad (j = nv), \quad (11)$$

and the equation of motion

$$i\omega m_i = en_0 \nabla \varphi + \nabla \cdot \Pi_{ik} \quad (\Pi_{ik} = p\delta_{ik}), \quad (12)$$

supplemented by a linearized equation of the state of the electron gas, which we shall take in the form  $p = ms^2 n$ , where  $s = s(z)$  is the velocity of sound in the electron gas (it is of the order of the Fermi velocity).

Eliminating the quantities  $j$  and  $n$  from Eqs. (10)–(12) and assuming that  $\varphi(r) = \varphi(z) \exp(i\mathbf{k} \cdot \boldsymbol{\rho})$ , we obtain generally a fourth-order equation for the potential  $\varphi(z)$ :

$$\frac{d}{dz} \left( \varepsilon(z) \frac{d\varphi}{dz} \right) - k^2 \varepsilon(z) \varphi + \left( \frac{d^2}{dz^2} - k^2 \right) \frac{s^2}{\omega^2} \left( \frac{d^2}{dz^2} - k^2 \right) \varphi = 0, \quad (13)$$

where  $\varepsilon(z) = 1 - 4\pi e^2 n(z) / m\omega^2$  is the permittivity of the transition layer. We recall that in vacuum we have  $\varepsilon_v = \varepsilon(-\infty) = 1$  (in the limit  $z \rightarrow -\infty$ , whereas for the metal (in the limit  $z \rightarrow +\infty$ ) we obtain

$$\varepsilon_m = \varepsilon(+\infty) = 1 - \frac{4\pi e^2 n(+\infty)}{m\omega^2} = 1 - \frac{4\pi e^2 n_0}{m\omega^2} = 1 - \frac{\omega_p^2}{\omega^2}.$$

We shall now consider the three consecutive regions  $z < z_1$  (vacuum),  $z_- < z < z_+$  (transition layer), and  $z > z_+$  (metal), defined by the fact that at  $z > z_+$  we can assume

that  $n(z) = n_+$ , whereas for  $z < z_-$ , we have  $n(z) = 0$ . Moreover, we shall postulate that the condition  $d \leq |z_+ - z_-| \leq k^{-1}$  is satisfied. In other respects the quantities  $z_{\pm}$  are arbitrary and drop out from the final formulas.

In these three regions, Eq. (13) becomes

1) in vacuum,

$$\frac{d^2 \varphi}{dz^2} - k^2 \varphi = 0, \quad (14)$$

2) in the metal,

$$\varepsilon_+ \left( \frac{d^2 \varphi}{dz^2} - k^2 \varphi \right) - \frac{s^2}{\omega^2} \left( \frac{d^2}{dz^2} - k^2 \right)^2 \varphi = 0, \quad (15)$$

where the ratio of the second to the first term is of the order of  $(ks/\omega_p)^2 \approx (kr_D)^2 \ll 1$ , because  $s \sim v_F$  and  $r_D \sim v_F/\omega \sim v_F/\omega_D$ .

It follows that the equation for the potential in the metal retains the same form as in vacuum

$$\frac{d^2 \varphi}{dz^2} - k^2 \varphi = 0, \quad (16)$$

and we find that

3) in the transition region

$$\frac{d}{dz} \left( \varepsilon(z) \frac{d\varphi}{dz} \right) - k^2 \varepsilon(z) \varphi + \frac{s^2}{\omega^2} \frac{d^4 \varphi}{dz^4} = 0 \quad (17)$$

the ratio of the second to the first term is of the order of  $(s/\omega_p d)^2 \sim (r_D/d)^2 \lesssim 1$  (we recall that  $r_D/d \leq 1$  is a small parameter of the model).

Thus, everywhere with the exception of the vicinity of the point  $z_0$ , where  $\varepsilon(z_0) = 0$ , we can ignore the second term. For the time being, we shall not consider this point and confine our attention to the equation

$$\frac{d}{dz} \left( \varepsilon(z) \frac{d\varphi}{dz} \right) - k^2 \varepsilon(z) \varphi = 0. \quad (18)$$

We shall seek the solution in the transition region as a series in powers of  $k$ ,

$$\varphi = \varphi_0 + k\varphi_1 + k^2\varphi_2 + \dots, \quad (19)$$

and find the coefficient  $\varphi_n$  by matching Eq. (19) to the potential in vacuum and in the metal.

It follows from Eqs. (15) and (16) that in vacuum

$$\varphi_v = e^{kz}, \quad (20)$$

whereas in the metal

$$\varphi_m = B_d e^{-kz} + B_i e^{kz} \quad (21)$$

or, expanding the exponential functions and amplitudes  $B$  in Eqs. (20) and (21) in powers of  $kz$ , we obtain

$$\varphi_v = 1 + kz + \frac{1}{2}(kz)^2 + \dots, \quad (22)$$

$$\begin{aligned} \varphi_m = & (B_d^{(0)} + kB_d^{(1)} + k^2 B_d^{(2)}) \left( 1 - kz + \frac{1}{2}(kz)^2 \right) \\ & + (B_i^{(0)} + kB_i^{(1)} + k^2 B_i^{(2)}) \left( 1 + kz + \frac{1}{2}(kz)^2 \right) + \dots \end{aligned} \quad (23)$$

where the subscripts  $d$  and  $i$  refer to the decreasing and rising components. Equating the terms with the same powers of  $k$  in Eqs. (20), (22), and (23), we obtain the values of the coefficients  $\varphi_n$  near the matching points in vacuum

$$\varphi_0(-\infty)=1, \quad \varphi_1(-\infty)=z, \quad \varphi_2(-\infty)=\frac{1}{2}z^2, \quad (24)$$

and in the metal

$$\left. \begin{aligned} \varphi_0(+\infty) &= B_d^{(0)} + B_i^{(0)}, & \varphi_1(+\infty) &= B_d^{(1)} + B_i^{(1)} + (-B_d^{(0)} + B_i^{(0)})z, \\ \varphi_2(+\infty) &= B_d^{(2)} + B_i^{(2)} + (-B_d^{(1)} + B_i^{(1)})z + \frac{1}{2}(B_d^{(0)} + B_i^{(0)})z^2. \end{aligned} \right\} (25)$$

We shall seek the solution in the transition region employing, as the boundary conditions, the continuity of the potential and its derivatives at the matching points. This can be done by substituting Eq. (19) into Eq. (18) and then the various approximations with respect to  $k$  give the following results.

1) In the zeroth approximation, we obtain

$$\varepsilon(z) \frac{d\varphi_0}{dz} = C, \quad \varphi_0 = C \int \frac{dz}{\varepsilon(z)} + C'$$

Assuming that the upper limit of  $z$  is  $z_+$ , we obtain  $C'=1$  from the first of the two conditions in Eq. (24). The continuity of the derivatives at  $z=z_+$  gives  $C=0$  and, therefore, we obtain

$$\varphi_0=1, \quad B_d^{(0)} + B_i^{(0)}=1, \quad (26)$$

2) In the first approximation, the same procedure gives

$$\varepsilon(z) \frac{d\varphi_1}{dz} = C, \quad \varphi_1 = C \int \frac{dz}{\varepsilon(z)} + C'$$

The second of the boundary conditions (25) at  $z=z_+$  gives  $C'=z_+$  and if the derivatives  $d\varphi/dz$  are equated at this point, then  $C=\varepsilon_-$ . Thus, we have

$$\varphi_1 = \varepsilon_- \int \frac{dz}{\varepsilon(z)} + z_+ \quad (27)$$

At  $z=z_+$ , we find from the second condition in Eq. (18)

$$\varepsilon_- \int \frac{dz}{\varepsilon(z)} + z_+ = B_d^{(1)} + B_i^{(1)} + (-B_d^{(0)} + B_i^{(0)})z_+,$$

and equating of the derivatives gives

$$\varepsilon_-/\varepsilon_+ = -B_d^{(0)} + B_i^{(0)}. \quad (28)$$

We finally have

$$\varepsilon_- \int \frac{dz}{\varepsilon(z)} + z_+ = B_d^{(1)} + B_i^{(1)} + \frac{\varepsilon_-}{\varepsilon_+} z_+. \quad (29)$$

3) In the second approximation, still following the same procedure, we obtain

$$\varepsilon(z) \frac{d\varphi_2}{dz} = \int \varepsilon(z) \varphi_0 dz + C = \int \varepsilon(z) dz + \varepsilon_- z_+$$

and at  $z=z_+$ ,

$$\varepsilon_+ (-B_d^{(1)} + B_i^{(1)}) + \varepsilon_+ z_+ = \int \varepsilon(z) dz + \varepsilon_- z_+. \quad (30)$$

From Eqs. (29) and (30) we obtain  $B_i^{(1)}$ , which is the amplitude of the solution that increases in the limit  $z \rightarrow \infty$ :

$$2B_i^{(1)} = \int \left( \frac{\varepsilon(z)}{\varepsilon_+} + \frac{\varepsilon_-}{\varepsilon(z)} \right) dz + \left( 1 + \frac{\varepsilon_-}{\varepsilon_+} \right) (z_- - z_+). \quad (31)$$

The quantity  $B_i^{(0)}$  is found from Eqs. (26) and (28) and its value is

$$2B_i^{(0)} = 1 + \varepsilon_-/\varepsilon_+.$$

The total amplitude  $B_i$  obtained in the approximation linear in  $k$  is  $B_i = B_i^{(0)} + kB_i^{(1)}$ , whereas the spectrum of surface plasmons is found by equating this amplitude to zero, i.e.,

$$B_i^{(0)}(\omega) + kB_i^{(1)}(\omega) + \dots = 0. \quad (32)$$

Assuming that  $\omega = \omega_s + kA$ , we find (in the first approximation) the eigenfrequency of surface plasmons from the condition

$$B_i^{(0)}(\omega_s) = 0 \quad \text{or} \quad 1 + \varepsilon_-/\varepsilon_+(\omega_s) = 0, \quad (33)$$

and hence, as expected,  $\omega_s = 2^{-1/2}\omega_p$ . The quantity  $A$  is deduced from Eq. (32) and defined by

$$A = -B_i^{(1)}(\omega_s) / \frac{dB_i^{(0)}(\omega_s)}{d\omega} = \frac{1}{2} \omega_s B_i^{(1)}(\omega_s), \quad (34)$$

where  $B_i^{(1)}(\omega_s)$  is found from Eq. (31) by substituting  $\omega = \omega_s$ .

The limits of integration in Eq. (31) are infinite because  $f = \varepsilon(z)/\varepsilon_+ + \varepsilon_-/\varepsilon(z)$  vanishes between the limits of  $z_+$  and  $+\infty$  and from  $-\infty$  to  $z_-$ . In fact, for  $z > z_+$  ( $z < z_-$ ) we have  $\varepsilon(z) = \varepsilon_+[\varepsilon(z) = \varepsilon_-]$  and  $f = 1 + \varepsilon_-/\varepsilon_+ = 0$  (and correspondingly  $f = \varepsilon_-/\varepsilon_+ + 1 = 0$ ) on the strength of Eq. (33).

In this way the final expression for the coefficient  $A$  becomes

$$A = \frac{1}{4} \omega_s \int_{-\infty}^{+\infty} \left( \frac{1}{\varepsilon(z)} - \varepsilon(z) \right) dz. \quad (35)$$

It is clear from the above formula that the main contribution to the coefficient  $A$  is due to the transition layer. In vacuum (in the limit  $z \rightarrow -\infty$ ), we find that  $\varepsilon(\omega) = 1$  and in the metal at the surface plasmon frequency  $\omega_s = 2^{-1/2}\omega_p$  (in the limit  $z \rightarrow +\infty$ ) we have  $\varepsilon(\omega_s) = -1$  and the integral considered without allowance for the transition layer vanishes identically. The integral (35) diverges in the vicinity of the point where  $\varepsilon(z) = 0$  and it should be additionally defined in this region by the more complete equation (17). Thus, the integral in Eq. (35) should be understood as the principal value; its value at a pole will be obtained below.

#### 4. REGION OF TURNING POINT $\varepsilon(z_0) = 0$

In the vicinity of a turning point, the complete equation is<sup>1)</sup>

$$\frac{d}{dz} \left( \varepsilon(z) \frac{d\varphi}{dz} \right) + \frac{s^2}{\omega^2} \frac{d^2\varphi}{dz^2} = 0. \quad (36)$$

If we assume that the velocity of sound varies little near the point  $z = z_0$  and apply the second of the conditions in Eq. (25), we obtain the first integral

$$\varepsilon(z) \frac{d\varphi}{dz} + \frac{s^2}{\omega^2} \frac{d^2\varphi}{dz^2} = \varepsilon_- \quad (37)$$

Near the point  $z = z_0$  we can expand the function  $\varepsilon(z)$  as a series, retaining the linear term,

$$\varepsilon(z) = 1 - \frac{\omega_p^2(z)}{\omega^2} \approx - \frac{d\omega_p^2(z)}{dz} \bigg|_{z=z_0} (z - z_0) = - \frac{z - z_0}{a}, \quad (38)$$

where

$$a = \frac{1}{4} \left( \frac{1}{n_0} \frac{dn}{dz} \right)^{-1}$$

is a quantity of the dimensions of length and of the same order as the thickness of the transition layer.

It is now convenient to modify Eq. (37) by replacing the potential with the field  $E = -d\phi/dz$  and by introducing a dimensionless variable  $\xi = (z - z_0)/a$ . Then, Eq. (37) becomes

$$\xi E - \mu^2 \frac{d^2 E}{d\xi^2} = e^{-\xi}$$

where a dimensionless parameter  $\mu$  represents

$$\mu = s/\omega a \approx r_D/d \ll 1. \quad (39)$$

Finally, assuming that  $\xi = \xi \mu^{2/3}$ , we can reduce the above equation to a form convenient for solution:

$$\frac{d^2 E}{d\xi^2} - \xi E = -\frac{e^{-\xi}}{\mu^{2/3}} = \kappa. \quad (40)$$

The solution of Eq. (36) (in terms of the corresponding potential) should be added to the expression  $\varepsilon_-/\varepsilon(z)$  in the formula for  $\varphi_1(z)$ , noting that this is important only near the point  $z_0$ .

The expression (40) is the inhomogeneous equation for the Airy function. The inhomogeneous term on the right-hand side makes the asymptotes of its solution differ from the usual asymptotic behavior of the Airy functions. Therefore, we shall solve it anew by the Laplace method. Assuming that

$$E(\xi) = \int_0^{\infty} u(t) e^{ut} dt,$$

where the arrow at the upper limit denotes an arbitrary selection of the integration path in the complex plane on condition that the integral converges at an infinitely distant point, we obtain the particular solution of the inhomogeneous equation (40) in the form

$$E_{\text{inh}} = \kappa \int_0^{\infty} \exp(\xi t - t^3/3) dt.$$

As usual, the integration path should lie within sectors of the complex plane  $t$  defined by the condition  $\text{Re} t^3 > 0$  (Fig. 2). It is convenient to take one of the particular solutions in the form of an integral along the imaginary axis in the upper half-plane

$$E_{\text{inh}} = F(\xi) = \kappa \int_0^{\infty} \exp(\xi t - t^3/3) dt. \quad (41)$$

The general solution of Eq. (40) should consist of the solution (41) and two particular solutions of the homo-

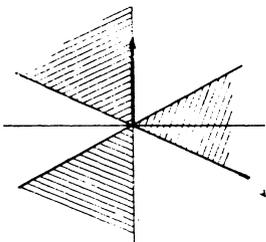


FIG. 2.

geneous equation, which are the Airy functions  $\text{Ai}(\xi)$  and  $\text{Ai}(-\xi)$ . Let us assume that  $\text{Ai}(\xi)$  is the Airy function oscillating at  $\xi < 0$  and decreasing exponentially for  $\xi > 0$ . Then,  $\text{Ai}(-\xi)$  has an asymptote which rises exponentially in the range  $\xi > 0$ . Since our solution is finite for all values of  $\xi$ , the function  $\text{Ai}(-\xi)$  should be absent from the general solution. Thus, the general solution of Eq. (40) finite for all values of  $\xi$  represents a linear combination of two functions  $F(\xi)$  and  $\text{Ai}(\xi)$ .

On the other hand, we can easily show that the function  $\text{Ai}(\xi)$  is the difference between two linearly independent particular solutions  $F(\xi)$  and  $F^*(\xi)$  (the asterisk denotes complex conjugacy):

$$\text{Ai}(\xi) = F(\xi) - F^*(\xi)$$

and, therefore, in deriving the general solutions we can confine ourselves to the two linearly independent particular solutions of the inhomogeneous equation.

Finally, we shall obtain the general solution of the inhomogeneous equation (40) in the form

$$E(\xi) = C_1 F(\xi) + C_2 F^*(\xi), \quad (42)$$

where the constants  $C_1$  and  $C_2$  are related by

$$C_1 + C_2 = 1, \quad (43)$$

and the function  $F(\xi)$  is given by Eq. (41). We shall now obtain asymptotic expressions for  $F(\xi)$  in the range of large values of  $\xi$ . We shall rotate the integration contour by the angle  $\pi/2$  in the clockwise direction. Then,

$$F(\xi) = i\kappa \int_0^{\infty} \exp[i(\xi u + u^3/3)] du. \quad (44)$$

If  $\xi > 0$ , the function  $f(\xi) = \xi u + u^3/3$  rises monotonically and the end point makes the principal contribution to the integral. If  $\xi < 0$ , the most important contribution is made by the saddle point, and this contribution is oscillatory; the contribution of the end point is asymptotically small although it decreases monotonically. A calculation of these contributions gives, for  $|\xi| \gg 1$ ,

$$F(\xi) \approx \begin{cases} -\kappa/\xi, & \xi > 0, \\ \kappa |\xi|^{-1/2} \pi^{1/2} \exp(-i^{2/3} |\xi|^{3/2} + 3/4 \pi i) - \kappa/\xi, & \xi < 0. \end{cases} \quad (45)$$

Since, in view of Eq. (38), in the vicinity of the point  $z_0$  we have  $\varepsilon^{-1}(z) \approx -a/(z - z_0) \approx -\kappa/\xi$ , it follows that  $F(\xi)$  in the range  $\xi \gg 1$  reduces to  $\varepsilon^{-1}(z) - \varepsilon(z)$  and, therefore, the integrand in Eq. (35) is replaced with  $F(\xi)$  in the vicinity of the point  $z_0$ .

We shall now return to the potential and calculate the contribution to the integral (35) originating from the vicinity of the point  $z_0$ . We shall do this by integrating Eq. (44) within the limits  $|z - z_0| < \delta$ , which reduces to the integration of the function  $F(\xi)$ :

$$I = \int_{-\delta}^{\delta} F(\xi) d\xi = i\kappa \int_0^{\delta} d\xi \int_0^{\infty} \exp\left[i\left(\xi u + \frac{u^3}{3}\right)\right] du = 2i\kappa \int_0^{\delta} \exp\left(\frac{iv^3}{3}\right) \frac{\sin v}{v} dv.$$

Here,  $\delta$  is the dimensionless thickness of the transition layer, which is of the order of  $\delta \sim \mu^{-2/3} \sim (s/w_0 a)^{-2/3} \sim (r_D/d)^{-2/3} \gg 1$ . Going to the limit  $\delta \rightarrow \infty$ , we obtain

$$I = 2i\kappa \int_0^{\infty} \exp\left(\frac{iv^3}{3\delta^3}\right) \frac{\sin v}{v} dv = 2i\kappa \int_0^{\infty} \frac{\sin v}{v} dv = i\pi\kappa. \quad (46)$$

The solution (44) now becomes

$$\int_{-d}^d E(\xi) d\xi = -i\pi\kappa(C_1 - C_2). \quad (47)$$

This expression should be added to the integral (35) which is understood to be the principal value. We shall now find the constants  $C_1$  and  $C_2$ . They are related by Eq. (43), and obtained, in fact, from the condition that there is no solution which increases with depth in the metal. The other relationship between  $C_1$  and  $C_2$  can be obtained by writing down the quasiclassical solution of the equation for the transition layer and matching it with the exact expression (45).

Turning back to Eq. (37), we shall rewrite it in the form

$$\frac{d^2 E}{dz^2} + \frac{\omega^2 - \omega_p^2(z)}{s^2(z)} E = -\left(\frac{\omega}{s}\right)^2 e_-. \quad (48)$$

Bearing in mind the smallness of the differential term, we can write the quasiclassical solution of the inhomogeneous equation in the form

$$E(z) = -\frac{e_-}{\varepsilon(z)} + \left\{ C_1' \exp\left(-i \int_{z_1}^z p(z) dz\right) + C_2' \exp\left(i \int_{z_1}^z p(z) dz\right) \right\} |p(z)|^{-1/2}, \quad (49)$$

where

$$p(z) = (\omega^2 - \omega_p^2(z))^{1/2} / s(z) \quad (50)$$

is, as usual, a smoothly varying function. The quasiclassical solution (49) is applicable to the transition layer in the interval  $z_1 < z < z_0$ , to the left of the turning point and up to the point  $z_1$  on the outer boundary of the metal corresponding to the vanishing of the velocity of sound.

The second and third terms in Eq. (49) thus describe two surface plasmon waves traveling in the transition layer parallel to the positive and negative directions of the  $z$  axis. The ratio of their amplitudes at the point  $z = z_1$  is governed by the surface reflection coefficient of plasmons

$$C_1'/C_2' = R = |r| e^{i\varphi_0}. \quad (51)$$

Here,  $|r|$  is the modulus of the reflection coefficient and  $\varphi_0$  is the phase of the reflected wave (taken at the point  $z_1$ ).

Matching of the solutions (49) and (42) subject to Eq. (45) gives

$$C_1' \exp\left[-i \int_{z_1}^{\infty} p(z) dz\right] = C_1 e^{2\pi i / 4},$$

$$C_2' \exp\left[i \int_{z_1}^{\infty} p(z) dz\right] = C_2 e^{-2\pi i / 4},$$

which gives

$$\frac{C_1}{C_2} = |r| e^{i\varphi_0}, \quad \varphi_0 = \varphi_1 - 2 \int_{z_1}^{\infty} p(z) dz - \frac{3\pi}{2}. \quad (52)$$

Solving Eqs. (43) and (52) simultaneously and substituting the resultant values of the constants in Eq. (47), we find that

$$\int_{-d/2}^{d/2} E(z) dz = i\pi a e_- \frac{|r| e^{i\varphi_0} - 1}{|r| e^{i\varphi_0} + 1}, \quad (53)$$

or, separating the real and imaginary parts,

$$\int_{-d/2}^{d/2} E(z) dz = -\pi a e_- [i(1 - |r|^2) + 2|r| \sin \varphi_0] \times [(1 + |r| \cos \varphi_0)^2 + |r|^2 \sin^2 \varphi_0]^{-1}. \quad (54)$$

Thus, combining Eqs. (54) and (35), we shall write the final expression for the coefficient in front of the linear term

$$A = A' + iA'',$$

where

$$\left. \begin{aligned} A' &= \frac{1}{4} \omega_+ \left\{ \int_{-\infty}^{\infty} \left( \frac{1}{\varepsilon(z)} - \varepsilon(z) \right) dz - \frac{4\pi a |r| \sin \varphi_0}{R} \right\}, \\ A'' &= \frac{\pi a \omega_+}{2R} (1 - |r|^2), \quad R = (1 + |r| \cos \varphi_0)^2 + |r|^2 \sin^2 \varphi_0. \end{aligned} \right\} \quad (55)$$

We can see that the imaginary correction to the plasmon spectrum is associated with the coefficient representing the reflection of plasmons by the boundary. If the reflection is elastic, i.e., if  $|r|=1$ , the imaginary part of Eq. (54) vanishes and there is no plasmon damping. The inelastic nature of the plasmon scattering at the boundary results in finite damping. The coefficient of inelasticity should, as mentioned above, be found either experimentally or by a microscopic calculation.

It should be stressed that the final expression (55) for the imaginary part of the coefficient  $A$  is meaningful only within the framework of our model, but we shall assume that the conclusion about the relationship between the imaginary part  $A$  and the decay processes near the surface remains valid also in the kinetic approach, to which we hope to return.

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<sup>1</sup>A similar analysis of the region of a turning point is given by Golant and Piliya<sup>6</sup> in connection with the problem of linear wave transformation in a plasma. The corresponding expression (36) near the turning point has been investigated by Wasow.<sup>7</sup> The asymptotes of the solutions of interest to us can be obtained from these two papers, but for our purpose it is more convenient to obtain the solutions anew.

<sup>1</sup>V. Celli, in: Surface Science (Lectures presented at Intern. Course, Trieste, 1974, ed. by A. Salam), International Atomic Energy Agency, Vienna, 1975, p. 393.

<sup>2</sup>S. Lundqvist, *ibid.*, p. 331.

<sup>3</sup>C. Kittel, Quantum Theory of Solids, Wiley, New York, 1963 (Russ. Transl., Nauka, M., 1967).

<sup>4</sup>V. M. Agranovich, Usp. Fiz. Nauk 115, 199 (1975) [Sov. Phys. Usp. 18, 99 (1975)].

<sup>5</sup>V. I. Pakhomov and K. N. Stepanov, Zh. Tekh. Fiz. 37, 1393 (1967) [Sov. Phys. Tech. Phys. 12, 1011 (1968)].

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<sup>7</sup>W. R. Wasow, Ann. Math. 52, 350 (1950).

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