Phenomenological description of the dielectric properties of surfaces: Surface waves

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For processes of macroscopic electrodynamics such as the reflection and refraction of waves or the propagation of a surface wave the difference between the properties of some arbitrary thin surface layer and those of the rest of the material can be described phenomenologically by introducing a surface permittivity. The properties of the surface permittivity and its effect on surface waves are studied. The results show that the existence of a surface layer with properties different from those of the rest of the material gives rise to surface waves which would not occur at a pure surface. The dispersion relation for such waves is studied for the case of an anisotropic surface layer.

1. INTRODUCTION

In many situations the properties of a surface layer are quite different from those of the material in the interior of a material. As examples we might cite the reconstruction of the surface of a single crystal and the layer of adsorbed atoms on a surface. The difference between the properties of a surface and the bulk properties of a material also influences macroscopic electrodynamic processes such as the reflection of light or the propagation of surface electromagnetic waves.

In certain cases it is possible to carry out a systematic analysis of surface properties at the microscopic level.[1] The complexity of that approach, however, necessitates the use of additional assumptions, which narrow the range of applicability of the results. Sivukhin[2] has shown for several simple models that in the reflection of light the presence of a thin transition layer can be dealt with by introducing corrections in the boundary conditions.

For processes of macroscopic electrodynamics, it is not necessary to take microscopic approach to the description of the surface. We would expect that a phenomenological description of the dielectric properties of a surface, like a phenomenological description of bulk properties, by means of a permittivity would prove useful in macroscopic electrodynamics. For a thin surface layer with a thickness δ small in comparison with the wavelength, all the specific surface properties may be related to a surface current. The coefficient of the proportionality between this current and the field is a phenomenological characteristic of the dielectric properties of the surface of a material. Below we develop this approach for describing a surface, and we study the effect of a surface permittivity on the properties of surface electromagnetic waves.

2. SURFACE CURRENT

An interface between two nonmagnetic media with permittivities $\varepsilon_{is}^{(1)}(\omega)$ and $\varepsilon_{is}^{(2)}(\omega)$ lies in the $z=0$ plane. A field induces a microcurrent of volume density

$$j_i(\mathbf{R},z,\omega)=\frac{\omega}{4\pi i}\{\varepsilon_{is}^{(1)}(\omega)\theta(z)+\varepsilon_{is}^{(2)}(\omega)\theta(-z)-\delta_{is}\}\times E_s(\mathbf{R},z,\omega), \tag{1}$$

in the material, where $\theta(z)=1$ at $z>0$, $\theta(z)=0$ at $z<0$, $\mathbf{R}=\mathbf{r}-\mathbf{n}(\mathbf{nr})$, and $\mathbf{n}$ is the unit normal to the surface.

If a transition layer has a finite width, the position of the interface is fixed unambiguously by the choice of conditions

$$V_1+V_2=V,\quad n_e^{(1)}V_1+n_e^{(2)}V_2=N_e, \tag{2}$$

where V_1, V_2, and V are the volume of the first medium, the volume of the second medium, and the total volume; $n_e^{(1)}$ and $n_e^{(2)}$ are the electron number densities in the first and second media; and N_e is the total number of electrons in the system.

Relation (1) presupposes that the properties of the material remain absolutely constant all the way to the interface. If there are variations in the properties of the material near the interface, the actual microcurrent density will deviate from (1) by an amount which we denote by $\delta\mathbf{j}$. This quantity obviously vanishes far from the interface.

The surface current $\mathbf{J}(\mathbf{R},\omega)$ is related to $\delta\mathbf{j}(\mathbf{R},z,\omega)$ by

$$\mathbf{J}(\mathbf{R},\omega)=\int dz\langle\delta\mathbf{j}(\mathbf{R},z,\omega)\rangle, \tag{3}$$

where the angle brackets mean an average over an area which physically is infinitely small in the $z=0$ plane. Note that the surface current $\mathbf{J}$ does not depend on z; in particular, it does not change when the two media are interchanged.

The existence of a surface layer has the consequence that this current and the associated surface charge should be incorporated in the boundary conditions at the $z=0$ interface:

$$Q(\mathbf{R},\omega)=\frac{1}{i\omega}\int dz\langle\operatorname{div}\delta\mathbf{j}(\mathbf{R},z,\omega)\rangle=\frac{1}{i\omega}\left(\frac{\partial\mathbf{J}^t(\mathbf{R},\omega)}{\partial\mathbf{R}}\right). \tag{4}$$

The surface charge is related exclusively to the tangential component of $\mathbf{J}$, since $\delta\mathbf{j}(\mathbf{R},z,\omega)$ vanishes outside the surface layer. As a result, the normal component of the surface current does not appear in the boundary conditions:

$$[\mathbf{n},\mathbf{H}_1-\mathbf{H}_2]=\frac{4\pi}{c}\mathbf{J}^t,\quad [\mathbf{n},\mathbf{E}_1-\mathbf{E}_2]=0,$$
$$(\mathbf{n},\mathbf{D}_1-\mathbf{D}_2)=4\pi Q,\ (\mathbf{n},\mathbf{H}_1-\mathbf{H}_2)=0 \tag{5}$$

(either directly or through the surface current).

It follows that for a phenomenological description of the dielectric properties of a surface it is sufficient to introduce the coefficient of proportionality between the tangential component of the surface current and the field.

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3. SURFACE PERMITTIVITY IN THE CASE OF AN ISOTROPIC SURFACE LAYER

We begin with the case in which the surface layer is isotropic. In this case we can introduce the relation

$$\mathbf{J}(\mathbf{R},t)=\int_0^\infty d\tau f(\tau)\mathbf{E}(\mathbf{R},t-\tau). \qquad (6)$$

This relation reflects the causal relationship between the field and the current, and it presupposes that this relationship is local. Taking Fourier transforms, we write

$$\mathbf{J}(\mathbf{R},\omega)=\frac{\omega}{4\pi i}\xi(\omega)\mathbf{E}(\mathbf{R},\omega), \qquad (7)$$

where

$$\xi(\omega)=\frac{4\pi i}{\omega}\int_0^\infty d\tau f(\tau)\exp(i\omega\tau) \qquad (8)$$

is an analog of the permittivity which we might call the "surface permittivity."

We wish to stress that there is a fundamental distinction between this quantity and the surface impedance of a metal, which is a concept used in the optics of metals.[3] A surface impedance is determined entirely by the bulk characteristics of the material, while the surface permittivity is introduced in order to describe certain other properties of the material, which are not described by bulk characteristics. A surface permittivity is independent of the bulk permittivities of the two media. The surface permittivity should be thought of as a phenomenological characteristic of a material, which is to be determined in an independent experiment (or calculated from a microscopic theory).

We will analyze the properties of $\xi(\omega)$ by analogy with the analysis of Landau and Lifshitz[3] of the properties of the permittivity $\varepsilon(\omega)$. It follows from (8) that $\xi(\omega)$ is a complex function of the frequency:

$$\xi(\omega)=\xi'(\omega)+i\xi''(\omega), \qquad (9)$$

$$\xi'(\omega)=\xi'(-\omega)=-\frac{4\pi}{\omega}\int_0^\infty d\tau f(\tau)\sin\omega\tau, \qquad (10)$$

$$\xi''(\omega)=-\xi''(-\omega)=\frac{4\pi}{\omega}\int_0^\infty d\tau f(\tau)\cos\omega\tau. \qquad (11)$$

The properties of the static permittivity are related to the convergence of the integral

$$\varkappa=\int_0^\infty d\tau f(\tau). \qquad (12)$$

If $\varkappa$ vanishes the surface layer is nonconducting, while if $\varkappa$ is equal to a constant we would be dealing with a conducting surface layer.

Let us examine $\xi(\omega)$ as a function of the complex argument $\omega=\omega'+i\omega''$. It follows from (8) that in the case $\omega''>0$ the integral (8) always converges (except at the point $\omega=0$ if $\varkappa$ is nonzero). This statement means that $\xi(\omega)$ has no singular points in the upper ω half-plane (except for a pole at $\omega=0$ if $\varkappa$ is nonzero). At frequencies higher than atomic frequencies, on the other hand, where the field interacts with bound electrons as it would with free electrons, we have $\xi(\omega)\sim\omega^{-2}$.

Repeating the arguments which Landau and Lifshitz present[3] in the derivation of dispersion relations for the bulk permittivity, we easily find dispersion relations for $\xi(\omega)$:

$$\xi'(\omega_0)=\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{d\omega}{\omega-\omega_0}\xi''(\omega), \qquad (13)$$

$$\xi''(\omega_0)=-\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{d\omega}{\omega-\omega_0}\xi'(\omega)+\frac{\varkappa}{\omega}. \qquad (14)$$

It follows that the function $\xi(\omega)$ may not be real at all frequencies, but only in a certain frequency interval.

For example, let us assume $\xi''(\omega)=0$ on the interval $\omega_1<\omega<\omega_2$. We then find from (13) and (14)

$$\xi'(\omega)=\frac{1}{\pi}\int_{-\infty}^{0}\frac{du}{u-\omega}\xi''(u)+\frac{1}{\pi}\int_0^\infty\frac{du}{u-\omega}\xi''(\omega)$$
$$=\frac{2}{\pi}\int_0^\infty\frac{u\,du}{u^2-\omega^2}\xi''(u). \qquad (15)$$

We have omitted the principal-value symbol here since the point $u=\omega$ lies in the interval in which we have $\xi''(u)=0$ outside the integration region. If the region of real values of $\xi(\omega)$ is quite wide, then in this region, in the interval

$$\omega_1\ll\omega\ll\omega_2,$$

we can assume

$$\xi(\omega)=\frac{2}{\pi}\int_{\omega_2}^{\infty}\frac{du}{u}\xi''(u)-\frac{2}{\pi\omega^2}\int_0^{\omega_1}u\,du\xi''(u)=\xi_0[1-(\omega_s/\omega)^2], \qquad (16)$$

where

$$\xi_0=\frac{2}{\pi}\int_{\omega_2}^{\infty}\frac{du}{u}\xi''(u),\qquad \xi_0\omega_s^2=\frac{2}{\pi}\int_0^{\omega_1}u\,du\xi''(u). \qquad (17)$$

It follows that if the frequency ω_s defined in (17) falls in the interval $\omega_1\ll\omega_s\ll\omega_2$ than $\xi(\omega)$ goes through zero at $\omega=\omega_s$.

4. SURFACE PERMITTIVITY FOR AN ANISOTROPIC SURFACE LAYER

In general, the surface current will depend on the values of the fields in the two media at the interface between them $(z=0)$. It is convenient to treat the surface current as a function not of the fields but of linear combinations of the fields:

$$\mathbf{F}_1=\mathbf{E}_1+\mathbf{n}(\mathbf{n}\mathbf{D}_1-\mathbf{n}\mathbf{E}_1),\quad \mathbf{F}_2=\mathbf{E}_2+\mathbf{n}(\mathbf{n}\mathbf{D}_2-\mathbf{n}\mathbf{E}_2). \qquad (18)$$

Without any loss of generality we can assume $\mathbf{J}=\mathbf{J}(\mathbf{F}_1+\mathbf{F}_2;\mathbf{F}_1-\mathbf{F}_2)$. As we mentioned earlier, the surface current does not change if the two media are interchanged. Consequently, $\mathbf{J}$ can depend on only $(F_1-F_2)^2$. It follows that to first order in the field $\mathbf{J}$ depends on only $\mathbf{F}_1+\mathbf{F}_2$. The field dependence of the tangential component of the surface current can be written in its most general form as follows:

$$J_i^t(\mathbf{R},\omega)=\frac{\omega}{8\pi i}\xi_{is}(\omega)\{F_{1s}(\mathbf{R},\omega)+F_{2s}(\mathbf{R},\omega)\}, \qquad (19)$$

where $\xi_{is}(\omega)$ is the surface permittivity. Here the Latin indices are to be understood as running over the values x, y, z;

while the Greek indices run over the values x, y. For the $z=0$ interface we can thus put (19) in the form

$$J_\alpha(\mathbf{R},\omega)=\frac{\omega}{4\pi i}\xi_{\alpha\beta}(\omega)E_\beta(\mathbf{R},\omega)+\frac{\omega}{8\pi i}\xi_{\alpha z}(\omega)[D_{1z}(\mathbf{R},\omega)+D_{2z}(\mathbf{R},\omega)]. \quad (20)$$

The components of the permittivity tensor, $\xi_{\alpha z}$, describe the rather exotic case in which the normal component of the field excites a tangential surface current in a thin surface layer.

It is convenient to direct the x and y axes along the principal axes of the two-dimensional tensor $\xi_{\alpha\beta}$. In this coordinate system, the tensor $\xi_{\alpha\beta}$ is diagonal. We denote its diagonal components by

$$\xi_{xx}=\xi+\eta,\quad \xi_{yy}=\xi-\eta. \quad (21)$$

The boundary conditions (5) then become

$$E_{1x}(\mathbf{R},\omega)=E_{2x}(\mathbf{R},\omega),\quad E_{1y}(\mathbf{R},\omega)=E_{2y}(\mathbf{R},\omega),$$

$$H_{1z}(\mathbf{R},\omega)=H_{2z}(\mathbf{R},\omega), \quad (22)$$

$$\varepsilon_1\left(1+\frac{i\omega}{2}\xi_{\alpha z}\frac{\partial}{\partial x_\alpha}\right)E_{1z}(\mathbf{R},\omega)+\varepsilon_2\left(1-\frac{i\omega}{2}\xi_{\alpha z}\frac{\partial}{\partial x_\alpha}\right)E_{2z}(\mathbf{R},\omega)=-(\xi+\eta)\frac{\partial E_{2x}}{\partial x}(\mathbf{R},\omega)-(\xi-\eta)\frac{\partial E_{2y}}{\partial y}(\mathbf{R},\omega), \quad (23)$$

$$H_{1x}(\mathbf{R},\omega)-H_{2x}(\mathbf{R},\omega)=i(\eta-\xi)\frac{\omega}{c}E_{2y}(\mathbf{R},\omega)-i\frac{\omega}{2c}\xi_{yz}(D_{1z}(\mathbf{R},\omega)+D_{2z}(\mathbf{R},\omega)), \quad (24)$$

$$H_{1y}(\mathbf{R},\omega)-H_{2y}(\mathbf{R},\omega)=i(\xi+\eta)\frac{\omega}{c}E_{2x}(\mathbf{R},\omega)+i\frac{\omega}{2c}\xi_{xz}(D_{1z}(\mathbf{R},\omega)+D_{2z}(\mathbf{R},\omega)). \quad (25)$$

5. EFFECT OF THE SURFACE PERMITTIVITY ON SURFACE ELECTROMAGNETIC WAVES

Let us examine surface electromagnetic waves at the $z=0$ interface between a nonmagnetic isotropic medium and a nonmagnetic cubic crystal. A surface wave can propagate along such an interface. We seek the field in this wave in the usual form [here and in (27), the upper sign corresponds to the first medium, and the lower to the second]

$$\mathbf{E}_{1(2)}\exp\{iq\mathbf{R}(\mp)\gamma_{1(2)}z-i\omega t\},\quad \mathbf{H}_{1(2)}\exp\{iq\mathbf{R}(\mp)\gamma_{1(2)}z-i\omega t\}.$$

Substitution of the fields, written in this form, into the wave equations for the first and second media leads to

$$\gamma_{1(2)}=[q_x^2+q_y^2-(\omega/c)^2\varepsilon_{1(2)}(\omega)]^{1/2}, \quad (26)$$

where $\gamma_{1(2)}$ are positive real quantities. Consequently the following equality must hold:

$$\mathrm{Im}\, q^2=\mathrm{Im}\,\omega^2\varepsilon_1/c^2=\mathrm{Im}\,\omega^2\varepsilon_2/c^2.$$

This equality is satisfied for real values of ε_1 and ε_2 at arbitrary frequencies: if ε_1 and ε_2 are complex, it is satisfied only at certain values of the frequencies. Since the fields in the two media are transverse, we can write

$$iq_xE_{1(2)x}+iq_yE_{1(2)y}=\pm\gamma_{1(2)}E_{1(2)z},$$

$$iq_xH_{1(2)x}+iq_yH_{1(2)y}=\pm\gamma_{1(2)}E_{1(2)z}. \quad (27)$$

We restrict the discussion to the case $\xi_{\alpha z}=0$,

For fields of this type, the boundary conditions (22)–(25) become

$$E_{1x}=E_{2x},\quad E_{1y}=E_{2y},\quad H_{1z}=H_{2z}, \quad (28)$$

$$\varepsilon_1E_{1z}-\varepsilon_2E_{2z}=-i(\xi+\eta)q_xE_{2x}-i(\xi-\eta)q_yE_{2y}, \quad (29)$$

$$H_{1x}-H_{2x}=-i(\xi-\eta)(\omega/c)E_{2y}, \quad (30)$$

$$H_{1y}-H_{2y}=i(\xi+\eta)(\omega/c)E_{2x}. \quad (31)$$

From (27) and (28) we find

$$\gamma_1E_{1z}+\gamma_2E_{2z}=0. \quad (32)$$

Using Maxwell's equation

$$q_xE_{2y}-q_yE_{2x}=\frac{\omega}{c}H_{2z}$$

and (27), we find

$$(q_x^2+q_y^2)E_{2x}=i\gamma_2q_xE_{2z}-q_y(\omega/c)H_{2z},$$

$$(q_x^2+q_y^2)E_{2y}=i\gamma_2q_yE_{2z}-q_x(\omega/c)H_{2z}. \quad (33)$$

Substituting (32) and (33) into (29), we can express all the quantities in the latter equation in terms of E_{2z} and H_{2z}:

$$\{\varepsilon_1\gamma_2+\varepsilon_2\gamma_1+\xi\gamma_1\gamma_2-\eta\gamma_1\gamma_2\cos 2\theta\}E_{2z}=iH_{2z}\eta\gamma_1\frac{\omega}{c}\sin 2\theta, \quad (34)$$

where θ is the angle which lies between the vector q and the y axis in the x, y plane, so that we have $q_x=q\sin\theta$, and $q_y=q\cos\theta$. Multiplying (30) by iq_x, multiplying (31) by iq_y, adding the results, and using (27), (28) and (33), we easily find

$$\left\{\gamma_1+\gamma_2-\xi\frac{\omega^2}{c^2}-\eta\gamma_1\gamma_2\cos 2\theta\right\}H_{2z}=-iE_{2z}\eta\gamma_2\frac{\omega}{c}\sin 2\theta. \quad (35)$$

The condition under which system of equations (34), (35) can be solved is the condition that the determinant of this system vanishes:

$$(\varepsilon_1\gamma_2+\varepsilon_2\gamma_1+\xi\gamma_1\gamma_2-\eta\gamma_1\gamma_2\cos 2\theta)[\gamma_1+\gamma_2-\xi(\omega/c)^2-\eta\gamma_1\gamma_2\cos 2\theta]=\eta^2\gamma_1\gamma_2(\omega/c)^2\sin^2 2\theta. \quad (36)$$

This equation relates q and ω; i.e., it is a dispersion relation for surface waves in the case in which there is a thin, anisotropic, surface transition layer.

6. ANALYSIS OF THE DISPERSION RELATION

A characteristic feature of the dispersion relation (36) is its dependence on the angle θ, i.e., on the wave propagation direction. By analogy with the optics of crystals, waves with direction-dependent dispersion relations might naturally be called "extraordinary surface waves."

In the case $\eta=0$, i.e., in the case of a transition layer with an isotropic surface, the system of equations (34), (35) splits into two independent equations:

$$(\varepsilon_1\gamma_2+\varepsilon_2\gamma_1+\xi\gamma_1\gamma_2)E_{2z}=0, \quad (37)$$

$$(\gamma_1+\gamma_2-\xi\omega^2/c^2)H_{2z}=0. \quad (38)$$

For the case of a plane macroscopic transition layer between media, with a thickness shorter than the wavelength, it is again possible to derive equations analogous to (37), (38) (Ref. 4). In this case, however, the derivation leans heavily on the assumption that there are two parallel interfaces which bound the transition layer down to distances small in comparison with the wavelength. An additional assumption of this sort is not physically justified, and it seriously restricts the range of applicability of the analysis. In problems involving the reflection of light, taking this approach for very thin layers leads to results at odds with experiment.[5]

Equations (37), (38) determine surface waves in the case of a surface-isotropic transition layer for an arbitrary value of the surface permittivity. The qualitative effect of the transition layer in this case is that there can be surface waves for which the electric field is transverse with respect to the propagation direction:

$$E_{2z}=0, \quad \gamma_1+\gamma_2=\xi(\omega/c)^2. \tag{39}$$

Such waves would not be possible in the absence of a surface layer, because γ_1 and γ_2 would be positive. The existence of such waves in the particular case of a macroscopic thin transition layer with a permittivity

$$\varepsilon(\omega)=\varepsilon_\infty(\omega^2-\omega_{LO}^2)/(\omega^2-\omega_{TO}^2) \tag{40}$$

was observed in Ref. 6. It follows from (39) that such waves exist in the general case of an arbitrary thin transition layer.

If a surface wave is propagating along the principal axes of the tensor $\xi_{\alpha\beta}$, i.e., along the x or y axis, and we have $\theta=\pi/2$ or $\theta=0$, then the system (34), (35) again splits into two independent equations:

$$[\gamma_1+\gamma_2-\xi(\omega/c)^2\mp\eta\gamma_1\gamma_2]H_{2z}=0, \tag{41}$$

$$[\varepsilon_1\gamma_2+\varepsilon_2\gamma_1+\gamma_1\gamma_2(\xi\mp\eta)]E_{2z}=0 \tag{42}$$

(the upper sign corresponds to $\theta=0$, and the lower sign to $\theta=\pi/2$).

We now consider the case in which ξ and η can be assumed to be small quantities, i.e., the case in which the properties of the surface layer differ only slightly from the properties of the material in the interior. In this case we can restrict the discussion to the approximation linear in ξ and η.

We accordingly ignore the term proportional to η^2 on the right side of Eq. (36), finding

$$\{\gamma_1+\gamma_2-(\omega/c)^2\xi-\eta\gamma_1\gamma_2\cos 2\theta\}\{\varepsilon_1\gamma_2+\varepsilon_2\gamma_1+\xi\gamma_1\gamma_2-\eta\gamma_1\gamma_2\cos 2\theta\}=0. \tag{43}$$

This result means that at small values of ξ and η there are again two solutions. One of them corresponds to the case

$$\varepsilon_1\gamma_2+\varepsilon_2\gamma_1+\xi\gamma_1\gamma_2-\eta\gamma_1\gamma_2\cos 2\theta=0, \tag{44}$$

and for it we have $H_{2z}=0$ by virtue of (34). The second solution corresponds to the equation

$$\gamma_1+\gamma_2-(\omega/c)^2\xi-\eta\gamma_1\gamma_2\cos 2\theta=0, \tag{45}$$

and for it we have $E_{2z}=0$ by virtue of (35).

The properties of surface waves satisfying (45) are related to the structure of the surface layer. If this layer has properties differing from those of the bulk material, then such waves exist; if there is no difference in properties, such waves do not exist.

7. SURFACE WAVES IN A CASE OF AN INHOMOGENEOUS THIN SURFACE LAYER

The properties of a thin surface layer may vary along the surface. In that case the surface permittivity should depend on the coordinates of the point on the interface, so we should replace (1) by the equality (we are again putting the interface in the $z=0$ plane)

$$\mathbf{J}(\mathbf{R},\omega)=\frac{\omega}{4\pi i}\xi(\mathbf{R},\omega)\mathbf{E}^t(\mathbf{R},\omega). \tag{46}$$

Such behavior might be caused in particular by a situation in which a surface layer with altered properties has a varying thickness (which remains small in comparison with the wavelength of the field). A similar situation would arise in a case in which the distribution of atoms adsorbed on a surface varied along the surface. Another particular case of this behavior would arise if there were a nonplanar surface $z=z_0(x,y)$ between two homogeneous media and if the dimensions of excursions from the $z=0$ plane along the normal to the surface, $|z_0(x,y)|$, remained small in comparison with the length scale of the field variations in the direction normal to the surface. In this particular case we would have

$$\xi(\mathbf{R},\omega)=(\varepsilon_2-\varepsilon_1)z_0(x,y). \tag{47}$$

How would the properties of surface waves change in the case of a thin, inhomogeneous surface layer of a material with unchanged properties? If the surface properties depend on the x and y coordinates the problem is no longer uniform as a function of x and y. We can thus no longer assume that the field of the surface wave behaves as a plane wave along the x and y directions (this assumption was made in Sec. 5). Variation as a function of x and y bends the waves and converts a plane wave into a packet of waves. We therefore seek the electric field of the surface wave as a superposition of waves

$$\mathbf{E}_{1(2)}(\mathbf{r},t)=\int d^2q\mathbf{E}_{1(2)}(\mathbf{q})\exp\{i\mathbf{qR}\mp\gamma_{1(2)}(q)z-i\omega t\}, \tag{48}$$

$$\mathbf{H}_{1(2)}(\mathbf{r},t)=\int d^2q\mathbf{H}_{1(2)}\mathbf{q}\exp\{i\mathbf{qR}\mp\gamma_{1(2)}(q)z-i\omega t\}. \tag{49}$$

Substituting fields of this type into the boundary conditions (5), we find boundary conditions on the expansion coefficients $\mathbf{E}_{1(2)}(q)$ and $\mathbf{H}_{1(2)}(q)$:

$$H_{1z}(\mathbf{q})-H_{2z}(\mathbf{q})=0, \tag{50}$$

$$\varepsilon_1E_{1z}(\mathbf{q})-\varepsilon_2E_{2z}(\mathbf{q})=-i\int d^2l\xi(\mathbf{l},\omega)\mathbf{E}_2(\mathbf{q}-\mathbf{l})\mathbf{q}, \tag{51}$$

$$[\mathbf{n},\mathbf{E}_1(\mathbf{q})-\mathbf{E}_2(\mathbf{q})]=0, \tag{52}$$

$$[\mathbf{n},\mathbf{H}_1(\mathbf{q})-\mathbf{H}_2(\mathbf{q})]=-i\frac{\omega}{c}\int d^2l\xi(\mathbf{l},\omega)E_2^t(\mathbf{q}-\mathbf{l}), \tag{53}$$

where

$$\xi(\mathbf{l},\omega)=(2\pi)^{-2}\int d^2R\xi(\mathbf{R},\omega)\exp(-i\mathbf{lR}). \tag{54}$$

It is not difficult to see that for the field expansion coefficients $\mathbf{E}_{1(2)}(\mathbf{q})$ and $\mathbf{H}_{1(2)}(\mathbf{q})$ the transversality conditions (27) and (33) hold [Eqs. (33) express the tangential components of the electric field in terms of the normal components of **E** and **H**].

Taking the vector product of (52) and $i\mathbf{q}$ and using (27), we easily find

$$\gamma_1(q)E_{1z}(\mathbf{q})+\gamma_2(q)E_{2z}(\mathbf{q})=0, \tag{55}$$

which we can use to eliminate $E_{1z}(\mathbf{q})$ from (51). Replacing $\mathbf{q}$ by $\mathbf{q}-\mathbf{l}$, we can express $E_{2x}(\mathbf{q}-\mathbf{l})$ and $E_{2y}(\mathbf{q}-\mathbf{l})$ in (33) in terms of $H_{2z}(\mathbf{q}-\mathbf{l})$ and $E_{2z}(\mathbf{q}-\mathbf{l})$. As a result we can replace (51) by

$$[\varepsilon_1\gamma_2(q)+\varepsilon_2\gamma_1(q)]E_{2z}(\mathbf{q})+\gamma_1(q)$$

$$\times\int d^2l\xi(\mathbf{l})\gamma_2(\mathbf{q}-\mathbf{l})E_{2z}(\mathbf{q}-\mathbf{l})\mathbf{q}(\mathbf{q}-\mathbf{l})/(\mathbf{q}-\mathbf{l})^2$$

$$=-i(\omega/c)\int d^2l\xi(\mathbf{l})\gamma_2(\mathbf{q}-\mathbf{l})E_{2z}(\mathbf{q}-\mathbf{l})(q_xl_y-q_yl_x)(\mathbf{q}-\mathbf{l})^{-2}. \tag{56}$$

Taking the vector product of (53) and $i\mathbf{q}$ and then taking the scalar product with $\mathbf{n}$, we find, using (27),

$$\gamma_1(q)H_{1z}(\mathbf{q})+\gamma_2(q)H_{2z}(\mathbf{q})=(\omega/c)^2\int d^2l\xi(\mathbf{l})H_{2z}(\mathbf{q}-\mathbf{l})$$
$$+(\omega/c)\int d^2l\xi(\mathbf{l})\{l_xE_{2y}(\mathbf{q}-\mathbf{l})-l_yE_{2x}(\mathbf{q}-\mathbf{l})\}.$$

Now eliminating H_{1z} with the help of (50), and eliminating E_{2x} and E_{2y} with the help of (33), we find

$$[\gamma_1(q)+\gamma_2(q)]H_{2z}(\mathbf{q})$$

$$-(\omega/c)^2\int d^2l\,H_{2z}(\mathbf{q}-\mathbf{l})\xi(\mathbf{l})\mathbf{q}(\mathbf{q}-\mathbf{l})/(\mathbf{q}-\mathbf{l})^2$$

$$=-i(\omega/c)\int d^2l\,\gamma_2(\mathbf{q}-\mathbf{l})\xi(\mathbf{l})E_{2z}(\mathbf{q}-\mathbf{l})(q_xl_y-q_yl_x)(\mathbf{q}-\mathbf{l})^{-2}. \tag{57}$$

Equations (56) and (57) can be used to find the functional dependences $H_{2z}(\mathbf{q})$ and $E_{2z}(\mathbf{q})$. The other field components can be found easily from the results, with the help of (33), the corresponding relations for H_{2x} and H_{2y}, and the boundary conditions (50)–(54).

8. SURFACE LAYER VARYING IN ONE DIRECTION

Let us consider the case in which the surface permittivity depends only on the single coordinate x (and is independent of y). In this case we can write

$$\xi(\mathbf{l})=\xi(l_x)\delta(l_y). \tag{58}$$

We consider a surface wave which is propagating along the x axis, so that in (48) and (49) we have

$$\mathbf{H}_{1(2)}(\mathbf{q})=\mathbf{H}_{1(2)}(q_x)\delta(q_y),$$
$$\mathbf{E}_{1(2)}(\mathbf{q})=\mathbf{E}_{1(2)}(q_x)\delta(q_y).$$

In this case Eqs. (56) and (57) can be written

$$(\varepsilon_1\gamma_2+\varepsilon_2\gamma_1)E_{2z}(q_x)$$
$$+\gamma_1(q_x)\int dl_x\xi(l_x)\gamma_2(q_x-l_x)E_{2z}(q_x-l_x)q_x(q_x-l_x)/(q_x-l_x)^2=0, \tag{59}$$

$$[\gamma_1(q_x)+\gamma_2(q_x)]H_{2z}(q_x)-(\omega/c)^2\int dl_x\,\xi(l_x)H_{2z}(q_x-l_x)=0; \tag{60}$$

i.e., Eqs. (56), (57), which constitute a system of coupled equations, split into two independent equations. It follows that in this case there exist two independent solutions, one with $E_{2z}=E_{1z}=0$ and one with $H_{2z}=H_{1z}=0$.

We now assume that $\xi(x,\omega)$ varies substantially over distances far greater than the wavelength. This assumption means that $\xi(l_x)$ is nonzero only at values of l_x much smaller than q_x. In this particular case we can expand $H_{2z}(q_x-l_x)$ in powers of l_x and retain only the first terms of the expansion.

Equation (60) then becomes

$$\varkappa dH_{2z}(q)/dq=\{(\gamma_1(q)+\gamma_2(q))-(\omega/c)^2\xi_0\}H_{2z}(q), \tag{61}$$

where

$$\xi_0=\int dl_x\xi(l_x),\qquad \varkappa=\int dl_xl_x\xi(l_x). \tag{62}$$

Integrating (61), we find $H_{2z}(q)$; then taking Fourier transforms we find

$$H_{2z}(\mathbf{r},t)=H_0\int dq\exp\{iqx-\Phi(q,z)-i\omega t\}, \tag{63}$$

where H_0 is a constant of integration, and

$$\Phi(q,z)=\gamma_2(q)|z|+\frac{1}{\varkappa}\int_{q_0}^{q}dq'(\gamma_1(q')+\gamma_2(q')-(\omega/c)^2\xi_0). \tag{64}$$

The integral is dominated by values of q for which Φ is a small quantity. The minimum of Φ corresponds to the condition

$$\varkappa^{-1}\left\{\gamma_1(q)+\gamma_2(q)-\left(\frac{\omega}{c}\right)^2\xi_0\right\}+|z|\frac{\partial\gamma_2(q)}{\partial q}=0, \tag{65}$$

which is the same as (39) at $z=0$.

Denoting the root of this equation by $z=Q(z)$, we see that (63) is dominated by values of q close to Q. Since large-scale variations alter q by only a small amount, we can assume that for the effective values of q the inequality $|q-Q|\ll Q$ holds. In this case we can expand $\Phi(q,z)$ in (63) in powers of $q-Q$:

$$\Phi(q,z)=\Phi(Q)+\frac{(q-Q)^2}{2\varkappa}Q\left(\frac{1}{\gamma_1(Q)}+\frac{1}{\gamma_2(Q)}\right). \tag{66}$$

An elementary integration of (63) using (66) leads to

$$H_{2z}(\mathbf{r},t)=H_0\left[\frac{2\pi\varkappa}{Q}\frac{\gamma_1(Q)\gamma_2(Q)}{\gamma_1(Q)+\gamma_2(Q)}\right]^{1/2}$$
$$\times\exp\{-\Phi(Q,z)+iQx-i\omega t\}. \tag{67}$$

In the case of a surface layer with large-scale variations in one dimension, a surface wave propagating along the x axis therefore constitutes a wave packet whose width depends on both the properties of the variations and the properties of the two media.

Expression (67) holds in the case in which the conditions $q^2>\varepsilon_1\omega^2/c^2$ and $\varepsilon_2\omega^2/c^2$ hold for the effective components of the packet. The latter conditions are satisfied if the following inequalities hold:

$$Q^2(z)-2\varkappa\gamma_1(Q)\gamma_2(Q)/Q[\gamma_1(Q)+\gamma_2(Q)]>(\omega/c)^2\varepsilon_1,\ (\omega/c)^2\varepsilon_2. \tag{68}$$

If these inequalities do not hold, components with imaginary values of γ_1 and γ_2 will appear in the packet. Such components would correspond to internal waves propagating away from the surface. In other words, they would correspond to a leakage of energy from surface waves to internal waves and to a pronounced damping of the surface waves.

At small values of $\varkappa$, an expansion of $H_{2z}(q_x - l_x)$ in l_x which retains no terms higher than linear terms is inadequate; expansion terms quadratic in l_x would have to be taken into account.

9. DISCUSSION OF RESULTS

An advantage of describing the properties of a surface layer in terms of a surface permittivity is that there are no model-dependent assumptions. It is thus possible to unambiguously relate the results found in the study of a surface by different electrodynamic methods. We wish to stress that the introduction of a surface permittivity permits a rigorous description of surface waves at a frequency ω for which the relation $\varepsilon_1(\omega) = \varepsilon_2(\omega)$ holds. In this case, ordinary surface waves simply do not exist, and they could arise only by virtue of the presence of a surface layer with altered properties.

Such surface waves (which might be called "degenerate surface waves") are particularly convenient for studying a surface since they are very sensitive to the properties of the surface layer. For a surface-isotropic and homogeneous transition layer, the conditions for the existence of degenerate surface waves can be derived easily from (34) and (35):

$$(2\varepsilon+\gamma\xi)E_{2z}=0, \qquad \left(2\gamma-\left(\frac{\omega}{c}\right)^2\xi\right)H_{2z}=0. \tag{69}$$

In the case $\xi > 0$, $\varepsilon > 0$, there can be degenerate surface waves for which the following hold:

$$E_{2z}=0,\ q^2=(\omega/c)^2\{\varepsilon(\omega)+\xi^2\omega^2/4c^2\}. \tag{70}$$

In the case $\xi\varepsilon < 0$, there can be waves for which the following hold:

$$H_{2z}=0,\ q^2=\varepsilon(\omega/c)^2+4\varepsilon^2/\xi^2. \tag{71}$$

A surface permittivity could be used to describe the dielectric properties of a plane in which defects accumulate in a crystal. Crystals that form in nonstoichimetric systems of various materials usually have planar defects.[7] The dielectric properties of a crystal vary near such a plane. This variation can be described by an additional surface current and thus by a surface permittivity. Degenerate surface waves can propagate along such planar defects.

We conclude with an estimate of the magnitude of the surface permittivity. For frequencies higher than the atomic frequencies, the microcurrent volume density is known to be related to the field by

$$\mathbf{j}(\mathbf{r},\ \omega)=\mathbf{E}(\mathbf{r},\ \omega)(ien_0Z_0/m\omega),$$

where n_0 is the number of atoms per unit volume, and Z_0 is the atomic number. The number of electrons in a case in which an atom of the material is replaced by an impurity atom with a charge Z varies by an amount $\Delta Z = Z - Z_0$. For a monolayer of impurity atoms on a surface we would have

$$J_t(r_\perp,\ \omega)=(i\omega_p{}^2b\Delta Z/4\pi\omega Z_0),$$

where b is the interatomic distance, and

$$\omega_p{}^2=4\pi e^2n_0Z_0/m.$$

Hence

$$\xi(\omega)\sim-b\Delta Z(\omega_p/\omega)^2.$$

Near a resonance for an impurity atom, but far from a resonance for the host material, an additional factor of $(\omega/\Delta\omega)$ arises:

$$\xi\sim\omega_pb\Delta Z/\omega\Delta\omega$$

(the deviation from the resonant frequency, $\Delta\omega$, is greater than the linewidth).

We now note that the customary boundary conditions, (5), are found in the limit $(\delta/\lambda)\to 0$. From Maxwell's equation

$$\mathrm{curl}_z\,\mathbf{H}=(4\pi/c)\delta j_z-i(\omega/c)D_z$$

in the same approximation, we find $J_z = 0$, by analogy with (5). The physical meaning of the latter equation is obvious: If a nonzero J_z is to appear, there must be oscillations of the charge along the z axis. If we denote by a the amplitude of these oscillations, we have $\delta \gtrsim a$, and the limit $\delta\to 0$ means that we also have $a\to 0$; i.e., the oscillations disappear along with J_z. Accordingly, the normal component of the surface current can be taken into consideration only along with finite corrections on the order (δ/λ) in the boundary conditions. The boundary conditions constructed in Ref. 8, which explicitly contain J_z, are physically meaningless since in finding them the authors went outside the range of applicability of the model which they were using [for a surface layer of thickness d, an expression of the type $\mathbf{P}\delta(z)$ was used for the polarization in Ref. 8, but this expression is incorrect at $z \lesssim d$, and it is specifically this region which is important to the construction of the boundary conditions in Ref. 8].

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