

Attenuation of surface polaritons by surface roughness*

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This paper presents a theoretical description of the attenuation of surface polaritons by roughness on the surface. In the presence of surface roughness, and in a frequency region where the dielectric constant is negative, the surface polariton is attenuated by two processes. It may lose energy by radiating into the vacuum, or by scattering into other surface-polariton states. Through application of a formalism developed recently to describe roughness-induced scattering and absorption of a plane electromagnetic wave incident on a surface, we obtain expressions for the contribution to the attenuation rate of the surface polariton from the two processes described above. We examine the relative importance of the two processes for surface polaritons on semiconductor surfaces, and on a nearly-free-electron metal at infrared frequencies.

I. INTRODUCTION

In a variety of materials, there are frequency regions where the dielectric constant is negative. This is the case in any insulating crystal with an infrared-active transverse-optical phonon, in the frequency region between the transverse- and the longitudinal-optical phonon at zero wave vector. Another example is provided by the nearly-free-electron metal, where the imaginary part of ϵ is small and the real part negative, for frequencies below the bulk-plasma frequency, but well above the conduction-electron relaxation frequency.

When the dielectric constant ϵ is real and negative, it is by now well known that surface electromagnetic waves (surface polaritons) may propagate down the surface.¹ The electromagnetic field of the surface polariton decays to zero exponentially with distance, as one moves away from the surface either into the vacuum above the material or into the material itself.

In the infrared frequency region, a number of experimental studies of surface polaritons have been reported in the recent literature. By means of a method such as the attenuated-internal-reflection technique, the dispersion relation and line-width of the surface polariton may be studied.² Also, the mean free path of infrared frequency surface polaritons on metal surfaces can be very long. As a consequence it is possible by use of a prism coupler to launch a surface polariton, and detect it after it has propagated along the surface a distance the order of a centimeter.³

As the polariton propagates down a perfectly smooth surface, it is attenuated by the dissipative processes present in the bulk of the material. An expression for the attenuation length may be obtained by inserting the complex dielectric constant into the dispersion relation, and extracting from it the imaginary part of the wave vector k_{\parallel} of the surface polariton.³ The attenuation lengths obtained by this means are in accord with the data.³

One may inquire about the possible importance of scattering processes and dissipation mechanisms specific to the surface region of the crystal in limiting the mean free path of surface polaritons. The purpose of the present paper is to describe a theoretical study of the contribution to the line-width and attenuation length from one such process, the attenuation produced by surface roughness. Although samples used in experiments in the optical and infrared range of the spectrum may have carefully prepared surfaces, many preparation techniques leave residual roughness on the surface, on the scale of a few hundred angstroms. Thus, it will be useful to assess the effect of this residual roughness in a quantitative manner.

If one considers an isotropic dielectric material with real dielectric constant ϵ , then the surface polariton may be attenuated by two processes, in the presence of roughness. The wave may radiate energy into the vacuum, or it may be scattered by the roughness into other surface-polariton states. In this paper, we obtain expressions for the contribution to the mean free path from each mechanism, and we examine their relative importance. We do this for surface polaritons in the infrared on semiconductor surfaces, and for surface polaritons on a nearly-free-electron metal in the infrared.

In this work, we employ a modification of a method developed recently⁴ to describe the roughness-induced scattering and absorption of a plane electromagnetic wave incident on a semi-infinite sample. At first glance, the present calculation seems a straightforward application of the formalism developed in Ref. 4. (Hereafter, we refer to Ref. 4 as Paper I.) However, there is one modification in the approach that must be made, if the theory is to be applied to a situation where the surface-polariton mean free path is very long. We comment briefly on this point here.

In I, the roughness-induced scattering and absorption of a plane electromagnetic wave incident

on the surface of a semi-infinite crystal with complex dielectric constant was examined. The surface of the crystal was presumed in the xy plane, and the incident wave illuminated a rectangular area on the surface with dimension $L_x \times L_y$. At non-normal incidence, the surface roughness produced a flow of energy parallel to the surface and localized to its near vicinity, in the x direction, where the xz plane is the plane of incidence. A certain fraction f_x of the incident flux is stored in this energy flow. If the real part $\epsilon^{(1)}$ of the dielectric constant is presumed negative, and the imaginary part $\epsilon^{(2)}$ small, then the dominant contribution to this energy flow has its origin in roughness-induced coupling of the incident radiation to surface polaritons. In I, we found f_x inversely proportional to L_x , but also inversely proportional to $\epsilon^{(2)}$ as $\epsilon^{(2)} \rightarrow 0$. Thus, the method used in I cannot be applied to the case where $\epsilon^{(2)} \rightarrow 0$ without producing an unphysically large energy flow parallel to the surface. Unfortunately, in the present paper, we wish to consider the case $\epsilon^{(2)} \equiv 0$, since particularly simple expressions obtain in this case, and $\epsilon^{(2)}$ is very small compared to $\epsilon^{(1)}$ in many instances of interest in the infrared region of the spectrum.

In I, it was pointed out that the expression for f_x obtained there is proportional to $l_{sp}(\omega)/L_x$, where $l_{sp}(\omega)$ is the mean free path of the surface polariton with frequency ω , the frequency of the incident radiation. We argued in I that the roughness-induced interaction between the incident radiation and the surface polariton had the nature of a phase-matched interaction, where the surface roughness upshifts the wave-vector component of the incident radiation parallel to the surface to match that of the surface polariton. Then for finite $\epsilon^{(2)}$, the mean free path of the surface polariton plays the role of the coherence length, and f_x is proportional to $l_{sp}(\omega)$ as a consequence. As argued in I, it is then clear that the result for f_x is correct only when $\epsilon^{(2)}$ is large enough that $l_{sp}(\omega) < L_x$. When $l_{sp}(\omega) > L_x$, L_x itself becomes the coherence length, and in the expression for f_x , the factor of $l_{sp}(\omega)/L_x$ should be replaced by a factor the order of unity.

The discussion in I centered on surface-roughness effects in the ultraviolet region of the spectrum, where the inequality $l_{sp}(\omega) < L_x$ is appropriate in the usual experimental situation. However, as remarked above, we wish to consider the limit $\epsilon^{(2)} = 0$ here, where $l_{sp}(\omega)$ becomes infinite on the perfectly smooth surface. While we use here the general formalism developed in I, a different method of evaluating the Poynting vector from the scattered field is required. In the discussion presented below, we use a method suitable for the limit $\epsilon^{(2)} = 0$.

With this method, we have derived a result for the quantity f_x defined in I valid when $\epsilon^{(2)} \equiv 0$ [and

$l_{sp}(\omega)$ is infinite], and we find that when $l_{sp}(\omega) \gg L_x$, the factor of $l_{sp}(\omega)/L_x$ is to be replaced by $\frac{1}{2}$. Thus, the conjecture in I about the result for f_x in the limit $\epsilon^{(2)} \rightarrow 0$ is found to be correct.

II. DERIVATION OF THE EXPRESSION FOR THE ATTENUATION LENGTH OF SURFACE POLARITONS IN THE PRESENCE OF SURFACE ROUGHNESS

In this section, we apply the approach of I to the derivation of the mean free path of surface polaritons in the presence of surface roughness. The basic formalism we use has been developed and described in I. As a consequence, we focus here only on those features of the derivation unique to the present problem. The notation we employ is identical to that in I, and we shall refer the reader there for explicit expressions for certain quantities which enter the discussion below.

We consider an isotropic dielectric material with surface parallel to the xy plane, and which lies in the region $z < 0$. The dielectric constant $\epsilon = \epsilon^{(1)} + i\epsilon^{(2)}$ is frequency dependent. We have in mind a frequency region where $\epsilon^{(1)}$ is negative and $\epsilon^{(2)}$ very small, as remarked in the Introduction. In fact, by the time we arrive at our final expressions, we shall have taken the limit $\epsilon^{(2)} \rightarrow 0$.

The surface of the dielectric is rough, and the height z of a point on the surface above the xy plane is given by the relation $z = \zeta(x, y)$. We presume the average value of ζ vanishes, and the root-mean-square value of $\zeta(x, y)$ will be denoted by δ^2 , i. e., $\delta^2 = \langle \zeta^2(x, y) \rangle$, where the angular brackets denote an average over the surface.

We shall presume that $\zeta(x, y)$ is nonzero only within a rectangular region of the surface with linear dimensions L_x and L_y . The reason why we allow $\zeta(x, y)$ to be nonzero only over a finite region of the surface is that we wish to limit the time any particular portion of the incident surface polariton samples the roughness. This will ensure that as $\epsilon^{(2)} \rightarrow 0$, all scattering cross sections remain finite. We take the incident surface polariton to propagate parallel to the x axis, with wave vector $\vec{k}_{||}^{(0)}$ and frequency ω . The frequency and wave vector of the surface polariton are related through the dispersion relation¹

$$\frac{c^2 k_{||}^{(0)2}}{\omega^2} = \frac{\epsilon}{\epsilon + 1}. \quad (2.1)$$

The geometry is illustrated in Fig. 1.

The formalism developed in I provides an expression for the contribution to the amplitude of the scattered electric field $\vec{E}^{(s)}$ first order in the roughness amplitude $\zeta(x, y)$. This is the electromagnetic analog of the first Born approximation of quantum-mechanical scattering theory.

We first consider the form of the scattered field in the vacuum outside the crystal. The formalism

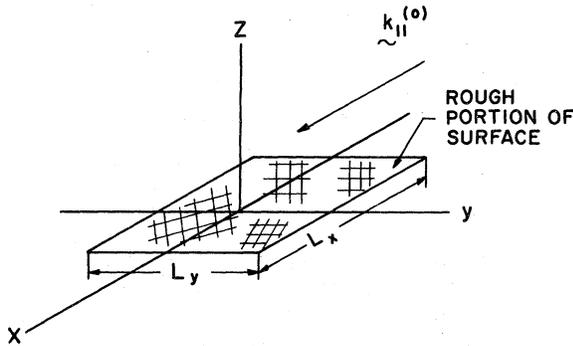


FIG. 1. Geometry employed in the surface-polariton attenuation-length calculation. The wave vector $\vec{k}_{\parallel}^{(0)}$ of the incident wave is parallel to the x axis.

in I provides an expression for the scattered electric field at point \vec{x} in the vacuum above the crystal that may be written

$$\vec{E}^{(s)}(\vec{x}, \omega) = -\frac{\omega^2}{16\pi^3 c^2} (\epsilon - 1) \int d^2 k_{\parallel} e^{i\vec{k} \cdot \vec{x}} \times \xi(\vec{k}_{\parallel} - \vec{k}_{\parallel}^{(0)}) \vec{\lambda}(\vec{k}_{\parallel} \vec{k}_{\parallel}^{(0)}, \omega). \quad (2.2)$$

The scattered electric field has the same frequency as the incident surface polariton. The quantity ϵ is the (complex) dielectric constant of the material, \vec{k}_{\parallel} is a two-dimensional wave vector in the plane of the surface, and

$$\vec{k} = \vec{k}_{\parallel} + \hat{z} k_z, \quad (2.3)$$

where

$$k_z = \left(\frac{\omega^2}{c^2} - k_{\parallel}^2 \right)^{1/2}. \quad (2.4)$$

We also have

$$\xi(\vec{Q}_{\parallel}) = \int d^2 x_{\parallel} e^{-i\vec{Q}_{\parallel} \cdot \vec{x}_{\parallel}} \xi(\vec{x}_{\parallel}). \quad (2.5)$$

The vector quantity $\vec{\lambda}(\vec{k}_{\parallel} \vec{k}_{\parallel}^{(0)}, \omega)$ is defined as follows. We define a matrix $S(\vec{k}_{\parallel})$ given by

$$S(\vec{k}_{\parallel}) = \frac{1}{k_{\parallel}} \begin{pmatrix} k_x & k_y & 0 \\ -k_y & k_x & 0 \\ 0 & 0 & k_{\parallel} \end{pmatrix}. \quad (2.6)$$

Then we have

$$\vec{\lambda}_{\mu}(\vec{k}_{\parallel} \vec{k}_{\parallel}^{(0)}, \omega) = \sum_{\mu'} \lambda_{\mu'}(\vec{k}_{\parallel} \vec{k}_{\parallel}^{(0)}, \omega) S_{\mu \mu'}(\vec{k}_{\parallel}). \quad (2.7)$$

The discussion in I employed an expression for $\lambda_{\mu}(\vec{k}_{\parallel} \vec{k}_{\parallel}^{(0)}, \omega)$ which in terms of the quantities defined there may be written

$$\lambda_{\mu}(\vec{k}_{\parallel} \vec{k}_{\parallel}^{(0)}, \omega) = \frac{1}{2} \sum_{\nu} [\hat{g}_{\mu\nu}(k_{\parallel} \omega | +) \mathcal{E}_{\nu}^{(0)}(\vec{k}_{\parallel} \vec{k}_{\parallel}^{(0)} \omega | +) + \hat{g}_{\mu\nu}(k_{\parallel} \omega | -) \mathcal{E}_{\nu}^{(0)}(k_{\parallel} k_{\parallel}^{(0)} \omega | -)], \quad (2.8)$$

where the field amplitudes $\mathcal{E}_{\nu}^{(0)}(\vec{k}_{\parallel} \vec{k}_{\parallel}^{(0)} \omega | \pm)$ are related to the amplitudes $E_{\nu}^{(0)}(\vec{k}_{\parallel}^{(0)} \omega | \pm)$ just above (+) and just below (-) the surface:

$$\mathcal{E}_{\nu}^{(0)}(\vec{k}_{\parallel} \vec{k}_{\parallel}^{(0)} \omega | \pm) = \sum_{\mu} S_{\mu\nu}(\vec{k}_{\parallel}) E_{\mu}^{(0)}(\vec{k}_{\parallel}^{(0)} \omega | \pm). \quad (2.9)$$

In the present paper, we utilize an expression for $\lambda_{\mu}(\vec{k}_{\parallel} \vec{k}_{\parallel}^{(0)}, \omega)$ which differs from that used in I, and displayed in Eq. (2.8) above. We shall use instead of the expression in Eq. (2.8) the form

$$\lambda_{\mu}(\vec{k}_{\parallel} \vec{k}_{\parallel}^{(0)}, \omega) = \sum_{\nu} \hat{g}_{\mu\nu}(k_{\parallel} \omega | +) \mathcal{E}_{\nu}^{(0)}(k_{\parallel} k_{\parallel}^{(0)} \omega | -). \quad (2.10)$$

Before we proceed, we explain the reason for this choice.

The derivation in I involved one tricky mathematical point. We evaluated the scattered fields by expanding the dielectric constant

$$\epsilon(x, y, z) = \theta(z - \zeta(x, y)) + \epsilon \theta(\zeta(x, y) - z) \quad (2.11a)$$

of the system in powers of $\zeta(x, y)$, with only the first term retained in the expansion:

$$\epsilon(x, y, z) = \theta(z) + \epsilon \theta(-z) + \zeta(x, y) \delta(z) (\epsilon - 1). \quad (2.11b)$$

The term proportional to $\zeta(x, y)$ was treated as a small perturbation in Maxwell's equations.

Through use of appropriate Green's functions, Maxwell's equations were rewritten in integral form, and the scattered field was calculated to first order in $\zeta(x, y)$. This leads to the evaluation of integrals of the form

$$\int dz' d_{\mu\nu}(k_{\parallel} \omega | z z') \delta(z') E_{\nu}^{(0)}(k_{\parallel}^{(0)} \omega | z'), \quad (2.12)$$

where $E_{\nu}^{(0)}(k_{\parallel}^{(0)} \omega | z')$ is the ν th Cartesian component of the incident electromagnetic field and $d_{\mu\nu}(k_{\parallel} \omega | z z')$ an element of a Green's-function array which, when considered a function of z' , obeys the same boundary conditions as $E_{\nu}^{(0)}(k_{\parallel}^{(0)} \omega | z')$.

As long as ν in Eq. (2.12) refers to x or y (the two directions parallel to the surface), the product of functions which multiply $\delta(z')$ is continuous across the boundary $z'=0$, and the integral may be evaluated in an unambiguous manner. However, when $\nu=z$, the function $d_{\mu\nu}(k_{\parallel} \omega | z z') E_{\nu}^{(0)}(k_{\parallel}^{(0)} \omega | z')$ suffers a jump discontinuity at $z'=0$, where the argument of $\delta(z')$ vanishes. In I, the integral was evaluated as

$$\frac{1}{2} [d_{\mu\nu}(\vec{k}_{\parallel} \omega | z 0+) E_{\nu}^{(0)}(k_{\parallel}^{(0)} \omega | 0+) + d_{\mu\nu}(\vec{k}_{\parallel} \omega | z 0-) E_{\nu}^{(0)}(\vec{k}_{\parallel}^{(0)} \omega | 0-)], \quad (2.13a)$$

a prescription readily derived by regarding the δ function in Eq. (2.12) as the limit of the appropriate Gaussian. This procedure leads to Eq. (2.8) for $\lambda_{\mu}(\vec{k}_{\parallel} \vec{k}_{\parallel}^{(0)}, \omega)$.

It has been pointed out^{5,6} that when one compares the prediction of the method in I with results ob-

tained by other methods, there are differences for one case, the scattering of p -polarized radiation at non-normal incidence into final states of p polarization. The discrepancy has its origin in the terms in Eq. (2.12) with $\nu = z$, where the discontinuity occurs in the prefactor of $\delta(z)$. In the formulas for pp scattering, where the method in I produces the term $\frac{1}{2}(1 + \epsilon^2) \sin\theta_s \sin\theta_0$ [see Eq. (2.72) of I], the other approaches lead to the term $\epsilon \sin\theta_s \sin\theta_0$.

The perturbation method of I calculates the scattered fields as if they were produced by certain perturbed surface currents in the surface region. Juranek⁷ has examined the question of how such surface currents may be placed and adjusted in strength so the radiated fields produced agree with those computed by matching boundary conditions across the perturbed surface.^{6,8,9} Juranek argues that (in the notation of I) the perturbed surface currents should be placed in infinitesimal distance above the crystal in the vacuum, and that the ν th Cartesian component of the current should have a strength proportional to $\zeta(x, y)(\epsilon - 1) \times E_\nu^{(0)}(k_\parallel^{(0)} \omega | -)$.

The prescription of Juranek may be built into the method of I by taking for the integral in Eq. (2.12) the value

$$d_{\mu\nu}(\vec{k}_\parallel \omega | z0+) E_\nu^{(0)}(k_\parallel^{(0)} \omega | 0-). \quad (2.13b)$$

When this is done, it is straightforward to see the factor of $\frac{1}{2}(1 + \epsilon^2)$ is replaced everywhere by ϵ , and the results for pp scattering produced by the method of I then agree with those of the boundary-matching method.

We adopt the prescription of Juranek here. When this is done, the expression for $\lambda_\mu(\vec{k}_\parallel, \vec{k}_\parallel^{(0)}, \omega)$ becomes

$$\lambda_\mu(\vec{k}_\parallel, \vec{k}_\parallel^{(0)}, \omega) = \sum_\nu \hat{g}_{\mu\nu}(k_\parallel \omega | +) \mathcal{G}_\nu^{(0)}(\vec{k}_\parallel, \vec{k}_\parallel^{(0)} \omega | -). \quad (2.14)$$

The results obtained below employ this form.

In I, the expression for $\vec{E}^{(s)}(\vec{x}, \omega)$ was derived for the case where the incident wave is a plane electromagnetic wave incident on the sample from above. The results can be applied equally well to the case where the incident wave is a surface polariton simply by inserting into Eq. (2.9) appropriate expressions for $E_\nu^{(0)}(\vec{k}_\parallel^{(0)} \omega | -)$.

In I, we formed an expression for the Poynting vector directly from Eq. (2.2), and then obtained expressions for the various cross sections of interest. Here we shall use a different procedure. We evaluate the expression for the scattered field directly from Eq. (2.2), in the asymptotic region $|\vec{x}| \rightarrow \infty$. Then from the scattered fields, we obtain the Poynting vector.

Before we proceed, one must note that two distinct domains of \vec{k}_\parallel exist. When $k_\parallel < \omega/c$, the quan-

tity k_z is real. This portion of the k_\parallel integral in Eq. (2.2) describes the field radiated away from the surface into the vacuum. When $k_\parallel > \omega/c$, k_z is pure imaginary. As described in I, one chooses the root for which $\text{Im}(k_z) > 0$. This portion of the k_\parallel integration describes electromagnetic fields localized to the surface, and which propagate parallel to it. It is these fields which give rise to the energy flow parallel to the surface discussed in Sec. I. To evaluate the asymptotic behavior of the field far from the region where $\zeta(x, y) \neq 0$, we consider each of these contributions separately.

(a) The region $k_\parallel < \omega/c$: In this region, \vec{k} is real, and we write

$$\begin{aligned} k_x &= k \sin\theta \cos\varphi, \\ k_y &= k \sin\theta \sin\varphi, \\ k_z &= k \cos\theta, \end{aligned}$$

while the orientation of the vector \vec{x} is described by the angles θ_s and φ_s of a spherical coordinate system.

When $k_\parallel < \omega/c$, the integral over d^2k_\parallel may be replaced by one over θ and φ through use of the relation¹⁰

$$d^2k_\parallel = (\omega/c)^2 \cos\theta \sin\theta d\theta d\varphi. \quad (2.15)$$

Crudely speaking, the coefficient of $e^{i\vec{k} \cdot \vec{x}}$ in Eq. (2.2) measures the amplitude of the plane wave of wave vector \vec{k} which radiates away from the surface. A detector placed far from the region where $\zeta(x, y) \neq 0$ should detect only the contribution to $\vec{E}^{(s)}(\vec{x}, \omega)$ directed from this region toward the detector. Formally, one may see this is so by evaluating the integral in Eq. (2.2) by the method of steepest descents in the limit $|\vec{x}| \rightarrow \infty$, after the transformation in Eq. (2.15). The basis for this is the observation that if we let $\Phi = \vec{k} \cdot \vec{x}$, then for fixed θ_s and φ_s , we have

$$\frac{\partial \Phi}{\partial \theta} \Big|_{\theta_s, \varphi_s} = \frac{\partial \Phi}{\partial \varphi} \Big|_{\theta_s, \varphi_s} = 0. \quad (2.16)$$

Then for θ near θ_s and φ near φ_s , we have

$$\Phi = kx - \frac{1}{2}kx [(\varphi_s - \varphi)^2 \sin^2\theta_s + (\theta_s - \theta)^2]. \quad (2.17)$$

In the limit $|\vec{x}| \rightarrow \infty$, we may evaluate the integral by using Eq. (2.17) in Eq. (2.2), and then removing all factors which vary smoothly with θ and φ from the integral, after they are evaluated at θ_s and φ_s . Then (again as $|\vec{x}| \rightarrow \infty$), the limits on the θ and φ integration may be extended to $\pm\infty$.

Upon noting that

$$\int_{-\infty}^{+\infty} d\varphi e^{i\alpha(\varphi - \varphi_s)^2} = (i\pi/\alpha)^{1/2}, \quad (2.18)$$

for the scattered electric field in the vacuum we find

$$\vec{E}^{(s)}(\vec{x}, \omega) = -i \frac{\omega^3(\epsilon - 1)}{8\pi^2 c^3} \vec{\lambda}(\vec{k}_\parallel, \vec{k}_\parallel^{(0)}, \omega) \times \zeta(\vec{k}_\parallel - \vec{k}_\parallel^{(0)}) \cos\theta_s \frac{e^{i\mathbf{k}\mathbf{x}}}{x}. \quad (2.19)$$

In Eq. (2.13), the vector \vec{k}_\parallel is to be evaluated on the presumption that \vec{k} is directed toward the point of observation \vec{x} .

It is a short exercise to show the Poynting vector is given by

$$\vec{S} = \frac{\omega^5 |\epsilon - 1|^2}{512\pi^5 c^4} \vec{k} \left| \vec{\lambda}(\vec{k}_\parallel, \vec{k}_\parallel^{(0)}, \omega) \right|^2 \times \left| \zeta(\vec{k}_\parallel - \vec{k}_\parallel^{(0)}) \right|^2 \frac{\cos^2\theta_s}{x^2}. \quad (2.20)$$

Let $(d^2 E^{(R)}/d\Omega dt)d\Omega$ be the energy per unit time radiated into the vacuum, into the solid-angle range $d\Omega$. From the expression for the Poynting

vector, we find

$$\frac{d^2 E^{(R)}}{d\Omega dt} = \frac{\omega^6 |\epsilon - 1|^2}{512\pi^5 c^5} \times \left| \vec{\lambda}(\vec{k}_\parallel, \vec{k}_\parallel^{(0)}, \omega) \right|^2 \left| \zeta(\vec{k}_\parallel - \vec{k}_\parallel^{(0)}) \right|^2 \cos^2\theta_s. \quad (2.21)$$

This result may be used to reproduce the results displayed in Eqs. (2.70)–(2.72) of I. Here we cast it in a form that will prove suitable to the discussion of the attenuation length of surface polaritons by evaluating $\lambda_\mu(\vec{k}_\parallel, \vec{k}_\parallel^{(0)}, \omega)$ for the case where the quantities $E_x^{(0)}(\vec{k}_\parallel^{(0)}\omega|-)$ and $E_z^{(0)}(\vec{k}_\parallel^{(0)}\omega|-)$ are nonzero, but $E_y^{(0)}(\vec{k}_\parallel^{(0)}\omega|-) \equiv 0$. This corresponds to the incident-wave configuration illustrated in Fig. 1. If we evaluate $\lambda_\mu(\vec{k}_\parallel, \vec{k}_\parallel^{(0)}, \omega)$ explicitly for the case where the dielectric constant is real and negative, and insert the result into Eq. (2.14), then after some algebra along lines very similar to that in I, we find

$$\frac{d^2 E^{(R)}}{d\Omega dt} = \frac{\omega^4(1 + |\epsilon|)}{32\pi^3 c^3} \cos^2\theta_s \left| \zeta(\vec{k}_\parallel - \vec{k}_\parallel^{(0)}) \right|^2 \left(\frac{|i(|\epsilon| + \sin^2\theta_s)^{1/2} \cos\varphi_s E_x^{(0)}(k_\parallel^{(0)}\omega|-) + |\epsilon| \sin\theta_s E_z^{(0)}(k_\parallel^{(0)}\omega|-)|^2}{|\epsilon| \cos^2\theta_s + \sin^2\theta_s} + \sin^2\varphi_s |E_x^{(0)}(k_\parallel^{(0)}\omega|-)|^2 \right). \quad (2.22)$$

In Eq. (2.22), the term proportional to $\sin^2\varphi_s$ has its origin in roughness-induced radiation to a final state of s polarization, and the other term in the large parentheses in radiation to a final state of p polarization.

Before we obtain the contribution to the attenuation length of the surface polariton by roughness-induced radiation into the vacuum, we turn to an evaluation of the asymptotic form of the scattered field in the region $k_\parallel > \omega/c$, where the scattered field is localized to the near vicinity of the surface.

(b) The regime $k_\parallel > \omega/c$: In this region, k_z is pure imaginary, and the scattered fields are localized near the surface. Outside the crystal, we choose the square root in Eq. (2.4) so that

$$k_z = i\beta_0, \quad (2.23)$$

with $\beta_0 > 0$. Then the expression for the scattered field becomes

$$\vec{E}^{(s)}(\vec{x}, \omega) = -\frac{\omega^2(\epsilon - 1)}{16\pi^3 c^2} \int d^2 k_\parallel e^{i\vec{k}_\parallel \cdot \vec{x}_\parallel} e^{-\beta_0 z} \times \lambda_\mu(\vec{k}_\parallel, \vec{k}_\parallel^{(0)}, \omega) \zeta(\vec{k}_\parallel - \vec{k}_\parallel^{(0)}). \quad (2.24)$$

We shall again use the method of steepest descent to evaluate the integral. However, we proceed differently than before. We suppose z fixed, and examine the limit $|x_\parallel| \rightarrow \infty$. For a given value

of k_\parallel , the quantity $e^{i\vec{k}_\parallel \cdot \vec{x}_\parallel - \beta_0 z} \vec{f}(\vec{k}_\parallel)$ describes a surface wave which propagates in the direction \hat{k}_\parallel parallel to the surface. A detector placed in the direction \hat{x}_\parallel very far from the region where $\zeta(x, y) \neq 0$ will collect waves with \vec{k}_\parallel in the direction of \hat{x}_\parallel . We obtain the contribution of these waves to the scattered field in the limit $|\vec{x}_\parallel| \rightarrow \infty$ by the method of steepest descents, after the replacement of $\vec{k}_\parallel \cdot \vec{x}_\parallel$ by $k_\parallel x_\parallel (1 - \theta^2/2)$, where as $|\vec{x}_\parallel| \rightarrow \infty$, only very small values of θ are important. An integration over θ may then be carried out by the use of Eq. (2.18). By this means we find as $|x_\parallel| \rightarrow \infty$ that

$$\vec{E}^{(s)}(\vec{x}, \omega) = \frac{\omega^2(\epsilon - 1)}{8\sqrt{2}\pi^{5/2} c^2} \frac{1}{(ix_\parallel)^{1/2}} \times \int_{\omega/c}^{\infty} dk_\parallel k_\parallel^{1/2} e^{-\beta_0 z} \vec{\lambda}(\vec{k}_\parallel, \vec{k}_\parallel^{(0)}, \omega) \zeta(\vec{k}_\parallel - \vec{k}_\parallel^{(0)}) e^{ik_\parallel x_\parallel}. \quad (2.25)$$

In Eq. (2.25), it is to be understood that \vec{k}_\parallel is directed toward the direction of \vec{x}_\parallel , i. e., $k_\parallel = k_\parallel \hat{x}_\parallel$.

It is a short exercise to obtain the form of $\vec{\lambda}(\vec{k}_\parallel, \vec{k}_\parallel^{(0)}, \omega)$. When $\vec{\lambda}(\vec{k}_\parallel, \vec{k}_\parallel^{(0)}, \omega)$ is written out explicitly, there are contributions with the quantity $k_1 - \epsilon k_z$ in the denominator, where $k_1 = -(\epsilon \omega^2/c^2 - k_\parallel^2)^{1/2}$ with $\text{Im}(k_1) < 0$. It is these terms which are of interest here. The reason is that the quantity $(k_1 - \epsilon k_z)^{-1}$ has a pole at the wave vector of the

surface polariton, when considered a function of k_{\parallel} . One sees this by noting that

$$\frac{1}{k_1 - \epsilon k_x} = \frac{k_1 + \epsilon k_x}{k_1^2 - \epsilon^2 k_x^2} = \left(\frac{k_1 + \epsilon k_x}{\epsilon^2 - 1} \right) \left(k_{\parallel}^2 - \frac{\omega^2}{c^2} \frac{\epsilon}{\epsilon + 1} \right)^{-1}. \quad (2.26)$$

The remaining terms in $\vec{\lambda}(\vec{k}_{\parallel}, \vec{k}_{\parallel}^{(0)}, \omega)$ are proportional to $(k_1 - k_x)^{-1}$. There are no poles of this denominator in the k_{\parallel} plane. We retain only the terms in $\vec{\lambda}(\vec{k}_{\parallel}, \vec{k}_{\parallel}^{(0)}, \omega)$ proportional to $(k_1 - \epsilon k_x)^{-1}$. From the arguments below, it will be clear that the neglected terms do not contribute to the scattered field, as $|\vec{x}_{\parallel}| \rightarrow \infty$. We then find

$$\bar{\lambda}_x(\vec{k}_{\parallel}, \vec{k}_{\parallel}^{(0)}, \omega) = (k_x/k_{\parallel}) \bar{\lambda}, \quad (2.27a)$$

$$\bar{\lambda}_y(\vec{k}_{\parallel}, \vec{k}_{\parallel}^{(0)}, \omega) = (k_y/k_{\parallel}) \bar{\lambda}, \quad (2.27b)$$

$$\bar{\lambda}_z(\vec{k}_{\parallel}, \vec{k}_{\parallel}^{(0)}, \omega) = i(k_{\parallel}/\beta_0) \bar{\lambda}, \quad (2.27c)$$

where

$$\bar{\lambda} = \frac{4\pi c^2}{\omega^2} \frac{k_{\parallel} \beta_0}{k_1 - i\epsilon\beta_0} \left(\frac{k_1 k_x}{k_{\parallel}^2} E_x^{(0)}(k_{\parallel}^{(0)} \omega | -) + \epsilon E_x^{(0)}(k_{\parallel}^{(0)} \omega | -) \right). \quad (2.28)$$

The integral over k_{\parallel} which appears in Eq. (2.25) may be evaluated through use of the contour in Fig. 2. The integral we seek is along the real axis in the k_{\parallel} plane, from ω/c to infinity. In Fig. 2, there is no contribution from the circular arc, so the integral in Eq. (2.25) is given by the contribution from the surface-polariton pole, along with that along the vertical segment from $k_{\parallel} = \omega/c$ to $k_{\parallel} = \omega/c + i\infty$. In the limit $\epsilon^{(2)} \rightarrow 0$, the contribution from the surface-polariton pole has the form $e^{ik_{\parallel}x_{\parallel}}/x_{\parallel}^{1/2}$, which is a radiation field in two dimensions. The contribution from the vertical segment falls off rapidly with x_{\parallel} (proportional to x_{\parallel}^{-3} for large x_{\parallel}) and does not contribute to the radiation field. Thus, we retain only the contribution from the surface-polar-

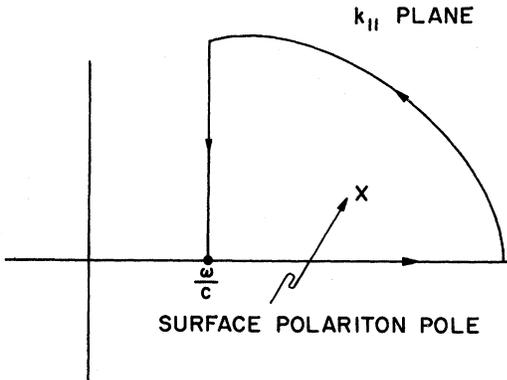


FIG. 2. Contour employed to evaluate the contribution to the scattered field from surface polaritons.

iton pole. When this is done, and we allow the dielectric constant to become real and negative, we find

$$\vec{E}^{(s)}(\vec{x}, \omega) = \left(\hat{x} \frac{k_x}{k_{\parallel}} + \hat{y} \frac{k_y}{k_{\parallel}} + i\hat{z} \frac{k_{\parallel}}{\beta_0} \right) \frac{E^{(s)}}{x_{\parallel}^{1/2}} e^{ik_{\parallel}x_{\parallel}} e^{-\beta_0 z}, \quad (2.29)$$

where

$$E^{(s)} = \frac{1}{(2\pi i)^{1/2}} \zeta(\vec{k}_{\parallel} - \vec{k}_{\parallel}^{(0)}) \frac{\beta_0 \beta_1 k_{\parallel}^{1/2}}{|\epsilon| - 1} \times \left(i \frac{\beta_1 k_x}{k_{\parallel}^2} E_x^{(0)}(\vec{k}_{\parallel}^{(0)} \omega | -) + |\epsilon| E_x^{(0)}(\vec{k}_{\parallel}^{(0)} \omega | -) \right). \quad (2.30)$$

In Eq. (2.30), we have written $k_1 = -i\beta_1$. The wave vector \vec{k}_{\parallel} is in the direction of the observation direction \vec{x}_{\parallel} ($\vec{k}_{\parallel} = \hat{x}_{\parallel} k_{\parallel}$), and its magnitude is found from Eq. (2.1); i. e., \vec{k}_{\parallel} is the wave vector of the surface polariton created in the scattering process.

Now let the total energy per unit time carried by the surface-polariton field in the vacuum above the crystal in the angular range between φ_s and $\varphi_s + d\varphi_s$ be denoted by $(d^2 E^{(sp)})/d\varphi_s dt d\varphi_s$. The time average of the Poynting vector in the vacuum above the crystal is parallel to \vec{k}_{\parallel} . If we denote its magnitude by s_{\parallel} , then

$$\frac{d^2 E^{(sp)}}{d\varphi_s dt} = x_{\parallel} \int_0^{\infty} dz s_{\parallel}. \quad (2.31)$$

We find

$$\frac{d^2 E^{(sp)}}{d\varphi_s dt} = \frac{\omega}{32\pi^2 \beta_0} \frac{k_{\parallel}^2 \beta_1^2}{(|\epsilon| - 1)^2} |\zeta(\vec{k}_{\parallel} - \vec{k}_{\parallel}^{(0)})|^2 \times \left[i \frac{\beta_1}{k_{\parallel}} \cos \varphi_s E_x^{(0)}(\vec{k}_{\parallel}^{(0)} \omega | -) + |\epsilon| E_x^{(0)}(\vec{k}_{\parallel}^{(0)} \omega | -) \right]^2. \quad (2.32)$$

In Eq. (2.32), we have the energy flow stored in the portion of the surface-polariton field which extends into the vacuum above the crystal. There is also a contribution from the portion of the surface-polariton field which extends into the medium. This second contribution is readily evaluated by the methods described above, and we do not present the details here. Indeed, the electric field in the medium may be obtained directly from Eq. (2.29) by noting that tangential components of E and normal components of D are conserved across the surface. In the medium, when the dielectric constant is negative, the Poynting vector is antiparallel to \vec{k}_{\parallel} .¹ One has

$$\frac{d^2 E^{(sp)}}{d\varphi_s dt} = -\frac{1}{|\epsilon|^2} \frac{d^2 E^{(sp)}}{d\varphi_s dt}. \quad (2.33)$$

Thus, the total rate at which energy flows in the surface-polariton field is

$$\frac{d^2 E^{(sp)}}{d\varphi_s dt} = \left(1 - \frac{1}{|\epsilon|^2}\right) \frac{d^2 E^{(sp)}}{d\varphi_s dt}. \quad (2.34)$$

After some rearrangement, the result reads

$$\begin{aligned} \frac{d^2 E^{(sp)}}{d\varphi_s dt} &= \frac{\omega^4}{32\pi^2 c^3} \frac{|\epsilon|(|\epsilon|+1)}{(|\epsilon|-1)^{5/2}} |\zeta(\vec{k}_{||} - \vec{k}_{||}^{(0)})|^2 \\ &\times |i|\epsilon|^{1/2} \cos\varphi_s E_x^{(0)}(\vec{k}_{||}^{(0)}\omega| -) \\ &+ |\epsilon| E_z^{(0)}(\vec{k}_{||}^{(0)}\omega| -)|. \end{aligned} \quad (2.35)$$

The expressions in Eqs. (2.22) and (2.35) are the principal results of the present section. They may be applied either to the case where the incident wave is a surface polariton, or to the case of a p -polarized plane wave incident at non-normal incidence.

As remarked in Sec. I, we have rederived the quantity $f_x^{(s)}$ discussed in paper I by the method of the present section. The calculation (carried out for s -polarized radiation at non-normal incidence) is straightforward and we do not present the details here. When we compare the result with Eq. (4.17) of I, we find that when ϵ is real, the factor of $l_{sp}(\omega)/L_x$ is replaced by $\frac{1}{2}$, as remarked in Sec. I.

We now use Eqs. (2.22) and (2.35) to deduce an expression for the mean free path of a surface polariton, in the presence of surface roughness. For the incident polariton, one has

$$E_z^{(0)}(\vec{k}_{||}^{(0)}\omega| -) = -\frac{i}{|\epsilon|^{1/2}} E_x^{(0)}(\vec{k}_{||}^{(0)}\omega| -), \quad (2.36)$$

where the direction of $\vec{k}_{||}^{(0)}$ is illustrated in Fig. 1. It is a short exercise to show that the energy per unit time stored in the incident wave is

$$\frac{dE_0}{dt} = \frac{L_y c^2}{16\pi \omega} \frac{(|\epsilon|-1)^2(|\epsilon|+1)}{|\epsilon|^{3/2}} |E_x^{(0)}(\vec{k}_{||}^{(0)}\omega| -)|^2. \quad (2.37)$$

Finally, we need the form of $|\zeta(\vec{Q}_{||})|^2$ for the case where random roughness is present on the surface. If we denote an ensemble average by angular brackets, then

$$\langle |\zeta(\vec{Q}_{||})|^2 \rangle = \int d^2 x_{||} d^2 r_{||} e^{i\vec{Q}_{||} \cdot \vec{r}_{||}} \langle \zeta(\vec{x}_{||} + \vec{r}_{||}) \zeta(\vec{x}_{||}) \rangle. \quad (2.38)$$

If the area $L_x L_y$ is large, $\langle \zeta(\vec{x}_{||} + \vec{r}_{||}) \zeta(\vec{x}_{||}) \rangle$ is independent of $\vec{x}_{||}$ save for a small fraction of the area close

to the boundary of the rough portion of the surface. We make little error by replacing $\langle \zeta(\vec{x}_{||} + \vec{r}_{||}) \zeta(\vec{x}_{||}) \rangle$ by $\langle \zeta(\vec{r}_{||}) \zeta(0) \rangle$. We then write

$$\int d^2 r_{||} e^{i\vec{Q}_{||} \cdot \vec{r}_{||}} \langle \zeta(\vec{r}_{||}) \zeta(0) \rangle = \delta^2 g(\vec{Q}_{||}), \quad (2.39)$$

where $g(\vec{Q}_{||})$ is normalized so

$$\int \frac{d^2 Q_{||}}{4\pi^2} g(\vec{Q}_{||}) = 1, \quad (2.40)$$

and $\delta = \langle \zeta^2 \rangle^{1/2}$ is the root-mean-square height of the surface roughness. Then we have

$$\langle |\zeta(\vec{Q}_{||})|^2 \rangle = L_x L_y \delta^2 g(\vec{Q}_{||}). \quad (2.41)$$

We now have the ingredients to form an expression for the attenuation length of the surface polariton. Upon integrating Eq. (2.22) over the appropriate portion of solid angle (φ_s ranges from $-\pi$ to $+\pi$ while θ_s ranges from 0 to $\pi/2$), and Eq. (2.35) over φ_s from $-\pi$ to $+\pi$, we obtain an expression for the total energy per unit time, $dE^{(T)}/dt$, radiated by the surface polariton as it passes by the rough patch on the surface. Upon use of Eqs. (2.36)–(2.38), one obtains the relation

$$\frac{dE^{(T)}}{dt} = \frac{L_x}{l} \frac{dE^{(0)}}{dt}. \quad (2.42)$$

From Fig. 1, one sees that L_x is the distance traveled by the surface polariton as it passes over the rough region of the surface. The ratio $(dE^{(T)}/dt)/L_x(dE^{(0)}/dt)$ is the energy lost per unit of distance traveled by the surface polariton. This is the inverse of the mean free path of the wave. From Eq. (2.37), one sees that the quantity l is the mean free path of the surface polariton. We write

$$\frac{1}{l} = \frac{1}{l^{(R)}} + \frac{1}{l^{(sp)}}, \quad (2.43)$$

where $1/l^{(R)}$ is the contribution to the attenuation length from roughness-induced radiation into the vacuum, and $1/l^{(sp)}$ is that from roughness-induced scattering of the surface polariton into other surface-polariton states.

Upon computing $1/l$ by means of the prescription just provided, we find

$$\frac{1}{l^{(R)}} = \frac{\delta^2 \omega^5}{2\pi^2 c^5} \frac{|\epsilon|^{3/2}}{(|\epsilon|-1)^2} \int_{-\pi}^{+\pi} d\varphi_s \int_0^{\pi/2} d\theta_s \cos^2 \theta_s \sin \theta_s g(\vec{k}_{||} - \vec{k}_{||}^{(0)}) \left(\sin^2 \varphi_s + \frac{(|\epsilon| + \sin^2 \theta_s)^{1/2} \cos \varphi_s - |\epsilon|^{1/2} \sin \theta_s}{|\epsilon| \cos^2 \theta_s + \sin^2 \theta_s} \right)^2 \quad (2.44)$$

and

$$\frac{1}{l^{(sp)}} = \frac{2}{\pi^2} \frac{\delta^2 \omega^5}{c^5} \frac{|\epsilon|^{7/2}}{(|\epsilon|-1)^{9/2}} \int_{-\pi}^{+\pi} d\varphi_s$$

$$\times g(\vec{k}_{||} - \vec{k}_{||}^{(0)}) \sin^4 \left(\frac{1}{2} \varphi_s\right). \quad (2.45)$$

These are the final results of this section. The analysis in Sec. III is based on the expressions in Eq. (2.44) and Eq. (2.45).

III. DISCUSSION OF THE RESULTS

In this section, we examine some consequences of the results obtained in Sec. II. We have in mind the infrared frequency region, where surface polaritons may be launched and propagated over macroscopic distances,³ or studied by either Raman scattering¹¹ or by the attenuated total reflection (ATR) method.

The two cases of interest are the properties of surface polaritons on the nearly-free-electron metal, which is described by the dielectric constant

$$\epsilon(\omega) = \epsilon_0 - \frac{\omega_p^2}{\omega^2} \quad (3.1)$$

provided $\omega\tau_e \gg 1$, where τ_e is the conduction-electron relaxation time, and surface polaritons on the surface of a semiconductor characterized by the dielectric constant

$$\epsilon(\omega) = \epsilon_0 + \frac{\Omega_p^2}{\omega_{TO}^2 - \omega^2}. \quad (3.2)$$

In Eq. (3.1), ω_p is the electron-plasma frequency, and in the infrared, $\omega \ll \omega_p$. In Eq. (3.2), ω_{TO} is the phonon frequency, and $\Omega_p^2 = 4\pi n e^{*2}/\mu$, where e^* is the transverse effective charge, n the number

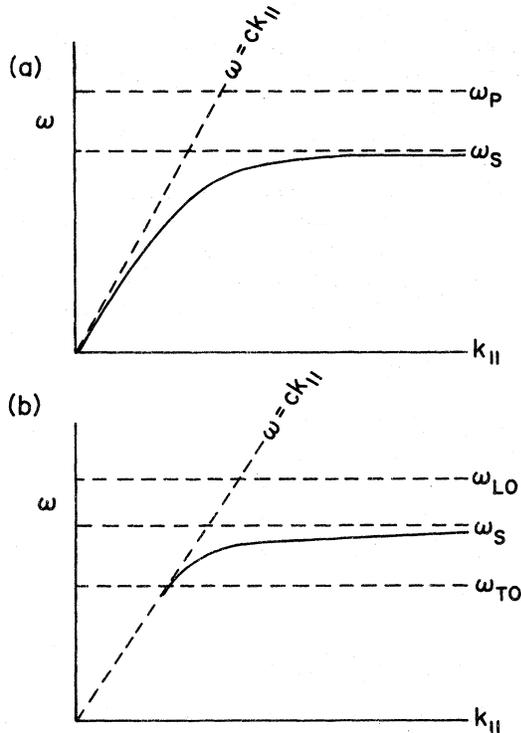


FIG. 3. The dispersion relation for (a) surface polaritons on the surface of a semi-infinite nearly-free-electron metal, and (b) surface polaritons in the surface of a semiconductor with a single infrared-active TO phonon. In each case, the frequency ω_s is found from the condition $\epsilon(\omega_s) = -1$.

of unit cells per unit volume, and μ a reduced mass of the ions.

The dispersion relation for the two cases is illustrated in Fig. 3. The frequency ω_s in each graph is found from the condition $\epsilon(\omega_s) = -1$ in each case, as one sees from Eq. (2.1).

There are two quantities of physical interest in the present discussion. The attenuation length l obtained in Sec. II is useful for estimating the effect of roughness on propagation-length studies such as the one reported by Shoenwald *et al.*³ As one sees from Fig. 3, near ω_s the surface dispersion curve is flat, and the group velocity $\partial\omega/\partial k_||$ of the mode small. Here the propagation length is too short for direct observation by virtue of the small group velocity. In this regime, however, one may study the roughness-induced linewidth of the surface-polariton mode either by Raman scattering or by the ATR method. The roughness-induced linewidth may be characterized by the dimensionless quantity Q , the number of oscillations of the mode before its energy density decays to e^{-1} of its initial value. This quantity is given by

$$Q = \frac{\omega}{2\pi} \tau = \frac{l}{2\pi v_g}, \quad (3.3)$$

where l is the attenuation length computed in Sec. II, and v_g the group velocity of the mode.

To compute the attenuation lengths $l^{(sp)}$ and $l^{(R)}$ obtained in Sec. II requires the form of the surface-roughness "structure factor" $g(\vec{Q}_||)$ defined in Eq. (2.39). If we make the ansatz that $\langle \xi(\vec{r}_||)\xi(0) \rangle$, has the Gaussian form

$$\langle \xi(\vec{r}_||)\xi(0) \rangle = \delta^2 e^{-r_||^2/\alpha^2}, \quad (3.4)$$

then $g(\vec{Q}_||)$ is given by

$$g(\vec{Q}_||) = \pi\alpha^2 e^{-(\alpha^2/4)Q_||^2}. \quad (3.5)$$

The length α is called the transverse correlation length. While δ is the rms height of the roughness, α is a measure of the average distance between successive "peaks" or "valleys."

With the Gaussian form for $g(\vec{Q}_||)$, the integration over φ_s is readily performed in Eqs. (2.44) and (2.45). For example, for $1/l^{(sp)}$ one finds

$$\frac{1}{l^{(sp)}} = \frac{\delta^2 \alpha^2 \omega^5}{c^5} \frac{|\epsilon|^{7/2}}{(|\epsilon| - 1)^{9/2}} e^{-t} \times \left[\frac{3}{2} I_0(\xi) - 2 I_1(\xi) + \frac{1}{2} I_2(\xi) \right], \quad (3.6)$$

where

$$\xi = \frac{1}{2} \frac{\omega^2 \alpha^2}{c^2} \frac{|\epsilon|}{(|\epsilon| - 1)} = \frac{1}{2} \alpha^2 k_||^2, \quad (3.7)$$

and $I_n(\xi)$ is the modified Bessel function of order n , and $k_||$ the wave vector of the surface polariton of frequency ω . If the Gaussian form of $g(\vec{Q}_||)$ is substituted into Eq. (2.44), the integral on φ_s may

also be performed, but the integral on θ_s must be evaluated numerically.

In the infrared frequency range, however, a simple approximation is valid in many circumstances. The wavelength of the surface polariton will frequently be large compared to the transverse correlation length α . Then in Eqs. (2.44) and (2.45), one will have a $|\vec{k}_\parallel - \vec{k}_\parallel^{(0)}| \ll 1$, and $g(\vec{k}_\parallel - \vec{k}_\parallel^{(0)})$ may be replaced by $g(0)$. The angular integrations in both Eqs. (2.44) and (2.45) may then be evaluated in closed form.

With $g(\vec{k}_\parallel - \vec{k}_\parallel^{(0)})$ replaced by $g(0)$, for $1/l^{(sp)}$ we find

$$\frac{1}{l^{(sp)}} = \frac{3}{2\pi} \frac{\delta^2 \omega^5}{c^5} \frac{|\epsilon|^{7/2}}{(|\epsilon| - 1)^{9/2}} g(0). \quad (3.8)$$

We split $1/l^{(R)}$ into two parts, the contribution $1/l_s^{(R)}$ that comes from radiation into final states of s polarization, and $1/l_p^{(R)}$ that comes from radiation into final states of p polarization. Then

$$\frac{1}{l_s^{(R)}} = \frac{\delta^2 \omega^5}{6\pi c^5} \frac{|\epsilon|^{3/2}}{[|\epsilon| - 1]^2} g(0) \quad (3.9)$$

and

$$\begin{aligned} \frac{1}{l_p^{(R)}} &= \frac{\delta^2 \omega^5}{2\pi c^5} \frac{|\epsilon|^{3/2}}{[|\epsilon| - 1]^3} g(0) \\ &\times \left[-\frac{2}{3} |\epsilon| \left[-\frac{1}{3} + \frac{3|\epsilon|^2}{|\epsilon| - 1} \left(1 - \frac{\tan^{-1}[(|\epsilon| - 1)^{1/2}]}{(|\epsilon| - 1)^{1/2}} \right) \right] \right]. \end{aligned} \quad (3.10)$$

To gain an appreciation of the relative importance of the different radiation processes in the various characteristic frequency regimes, we consider the limiting form of the results displayed above in the two cases $|\epsilon| \gg 1$ and $|\epsilon|$ close to unity in value (then ϵ itself is close to -1 and the frequency of the surface polariton lies close to the asymptotic value ω_s in Fig. 3).

(a) The case $|\epsilon| \gg 1$: This limit applies to surface polaritons on the metal-vacuum interface when $\omega \ll \omega_p$, and as one sees from Fig. 3(a), one has $\omega \approx ck_\parallel$ here. It also applies to the semiconductor-vacuum interface when ω is close to ω_{TO} . Then we have in this limit

$$\frac{1}{l^{(sp)}} = \frac{3}{2\pi} \frac{\delta^2 \omega^5}{c^5} \frac{g(0)}{|\epsilon|}, \quad (3.11)$$

$$\frac{1}{l_s^{(R)}} = \frac{1}{6\pi} \frac{\delta^2 \omega^5}{c^5} \frac{g(0)}{|\epsilon|^{1/2}}, \quad (3.12)$$

and

$$\frac{1}{l_p^{(R)}} = \frac{7}{6\pi} \frac{\delta^2 \omega^5}{c^5} \frac{g(0)}{|\epsilon|^{1/2}}. \quad (3.13)$$

One sees that the most effective process for attenuating the surface polariton is the roughness-induced radiation into the vacuum above the substrate. The attenuation length in this regime is given by

$$l = \frac{3\pi}{4} \frac{c^5}{\delta^2 \omega^5} \frac{|\epsilon|^{1/2}}{g(0)} = \frac{3}{4} \frac{c^5}{\delta^2 \alpha^2 \omega^5} |\epsilon|^{1/2}, \quad (3.14)$$

where the last form follows upon using the Gaussian form for $g(0)$. For the quantity Q , one has, with $v_g \approx c$, the result

$$Q = \frac{3}{8\pi} \frac{c^4}{\delta^2 \alpha^2 \omega^4} |\epsilon|^{1/2}. \quad (3.15)$$

(b) The case where $|\epsilon|$ is near unity: When $|\epsilon|$ is near unity, as remarked above, one is in the regime where the frequency ω of the surface polariton lies close to the asymptotic frequency ω_s found from the condition $\epsilon(\omega_s) = -1$. In this regime, we have

$$\frac{1}{l^{(sp)}} = \frac{3}{2\pi} \frac{\delta^2 \omega^5}{c^5} \frac{g(0)}{(|\epsilon| - 1)^{9/2}}, \quad (3.16)$$

$$\frac{1}{l_s^{(R)}} = \frac{\delta^2 \omega^5}{6\pi c^5} \frac{g(0)}{(|\epsilon| - 1)^2}, \quad (3.17)$$

$$\frac{1}{l_p^{(R)}} = \frac{11\delta^2 \omega^5}{30\pi c^5} \frac{g(0)}{(|\epsilon| - 1)^2}. \quad (3.18)$$

From these results, one sees that when ϵ is near -1 , the surface polariton is damped predominantly by the scattering of the wave into other surface-polariton states, rather than roughness-induced radiation into the vacuum. Calculations based on the full formulas show that the surface scattering process dominates the radiation damping for $|\epsilon| \lesssim 7$.

When ω is near ω_s , the attenuation length is necessarily short, simply because the group velocity is small and the surface polariton will not propagate far even if the scattering is relatively weak. A more useful measure is the quantity Q defined above. For $|\epsilon|$ near unity, and for surface polaritons on a substrate described by the dielectric function in Eq. (3.2), the group velocity v_g of the surface polariton is given by

$$v_g \cong \frac{\epsilon_s - \epsilon_0}{\epsilon_s + 1} c (|\epsilon| - 1)^{3/2} \quad (3.19)$$

and from Eq. (3.16) one has

$$Q \cong \frac{c^4}{3\omega^4} \frac{(|\epsilon| - 1)^3}{\delta^2 g(0)} \left(\frac{\epsilon_s + 1}{\epsilon_s - \epsilon_0} \right). \quad (3.20)$$

This result may also be written in the form

$$Q \cong \frac{\omega_s^2}{3c^2} \left(\frac{\epsilon_s + 1}{\epsilon_s - \epsilon_0} \right) \frac{1}{\delta^2 g(0)} \frac{1}{k_\parallel^6}, \quad (3.21)$$

where k_\parallel is the wave vector of the surface polariton.

There are two criteria for the validity of the result in Eq. (2.21). One must have $ck_\parallel \gg \omega$ (this ensures the condition $|\epsilon|$ near unity is satisfied), and one must also have $k_\parallel \alpha \ll 1$; i. e., the wavelength of the surface polariton must be long compared to the transverse correlation length. In this

wave-vector regime, the scattering rate is found to be a very strong function of the wave vector of the surface polariton.

As remarked earlier, on the basis of a numerical evaluation of the expressions above, we find the dominant contribution to the roughness-induced damping of the surface polariton comes from the scattering processes which contribute to $1/l^{(\text{sp})}$, so long as $|\epsilon| < 7$. In this regime, for the case where $g(\tilde{Q}_{||})$ may be taken to be Gaussian, Eqs. (3.6) and (3.7) provide expressions for $1/l^{(\text{sp})}$ valid for all values of $k_{||}\alpha$. [Note that $k_{||}^2 = \omega^2|\epsilon|/c^2(|\epsilon| - 1)$.] Then for $k_{||}\alpha \gg 1$, the Gaussian roughness model predicts that one should have

$$\frac{1}{l^{(\text{sp})}} = \frac{3}{4(2\pi)^{1/2}} \frac{\delta^2}{\alpha^3} \frac{|\epsilon|}{(|\epsilon| - 1)^2} \quad (3.22)$$

or

$$Q = \frac{4}{3(2\pi)^{3/2}} \frac{\omega_s}{c} \frac{\alpha^3}{\delta^2} \left(\frac{\epsilon_s + 1}{\epsilon_s - \epsilon_0} \right) (|\epsilon| - 1)^{1/2}. \quad (3.23)$$

If the result is written in terms of the wave vector of the surface polariton, then

$$Q = \frac{3}{3(2\pi)^{3/2}} \frac{\omega_s^2}{c^2} \frac{\alpha^3}{\delta^2 k_{||}} \left(\frac{\epsilon_s + 1}{\epsilon_s - \epsilon_0} \right). \quad (3.24)$$

We remind the reader that the result in Eq. (3.24) requires for its validity the conditions $ck_{||} \gg \omega_s$ and also $k_{||}\alpha \gg 1$ to be satisfied.

It is important to note that one expects the results in Eqs. (3.8)–(3.10) to be generally valid, as long as $k_{||}\alpha \ll 1$. This is independent of the assumption that $g(\tilde{Q}_{||})$ has the Gaussian form. Quite generally, when $ck_{||} \gg \omega$, if $g(\tilde{Q}_{||})$ has its maximum at $\tilde{Q}_{||} = 0$ one may show that for large $k_{||}$ the quantity Q becomes inversely proportional to $k_{||}$, as one sees in Eq. (3.24). However, the coefficient of the combination

$$\frac{\omega_s^2}{c^2} \frac{1}{\delta^2 k_{||}} \left(\frac{\epsilon_s + 1}{\epsilon_s - \epsilon_0} \right)$$

displayed in Eq. (3.24) depends on the assumption that $g(\tilde{Q}_{||})$ is a Gaussian.

We conclude with some comments about the implications of the results given above.

First consider the effect of surface roughness on the macroscopic attenuation lengths measured by Shoenwald, Burstein, and Elson.³ In these experiments, a CO₂ laser was used as a source of 10.6- μm radiation, and a surface polariton was launched on a metallic substrate through use of a prism coupler. Attenuation lengths the order of 1 cm were measured for the wave, and the measured attenuation length was found in semiquantitative agreement with that by computing the imaginary part of $k_{||}$ from Eq. (2.1), with the complex dielectric constant of the substrate inserted on the

right-hand side.

One can inquire how sensitive this very long attenuation length is to the presence of roughness on the surface. It is reasonable to describe the substrate by the model dielectric constant in Eq. (3.1), and under the experimental conditions $\omega \gg \omega_p$ one has $|\epsilon| \gg 1$. Thus the roughness contribution to the attenuation rate should have its origin primarily in roughness-induced radiation into the vacuum above the crystal. The contribution to the attenuation length from roughness is then given by Eq. (3.14), provided that $k_{||}\alpha \ll 1$ for the wave. For a well-polished surface, the condition $k_{||}\alpha \ll 1$ should be well satisfied at 10.6 μm . If we set $\alpha = \delta$, a reasonable approximation for grit-polished surfaces, the roughness-induced attenuation rate will be 1 cm^{-1} if $\delta \approx 2500 \text{ \AA}$. Thus, if the surfaces used in the propagation experiments are optically smooth, the attenuation length should not be affected seriously by the residual surface roughness. However, this estimate does suggest that it will be necessary to use surfaces of good optical quality for the work, since the attenuation constant can be degraded considerably by larger values of δ . Indeed, for all the substrates studied (Cu, Ag, and Au), the measured attenuation constant was greater than that calculated by the procedure described above by a factor that ranged from 20% to a factor of 2. The estimate above suggests that the discrepancy could readily arise from residual surface roughness.

In the recent literature, surface polaritons on semiconductor surfaces have been studied by the method of Raman scattering,¹¹ and by the ATR method.¹²

Of these two methods, the Raman technique affords the possibility of direct comparison with the results presented here.¹³ This method studies the properties of surface polaritons on a surface unperturbed by a grating or nearby prism coupler. A wide range of wave vectors may also be probed. In the study of surface polaritons by Raman scattering, one has a well-defined wave vector $\vec{k}_{||}$ associated with each scattering angle, and in principle only in a single mode contributes to the spectrum. While it has been demonstrated that in the backscattering geometry, the linewidth of the bulk TO and bulk LO phonons is sensitive to imperfections near the surface,¹⁴ such data are difficult to analyze quantitatively because the Raman signal contains contributions from bulk modes with a range of wave-vector components k_x normal to the surface.¹⁵ We hope that in the near future, Raman studies of the surface-polariton linewidth on surfaces with controlled amounts of roughness may be carried out. Numerical estimates based on Eq. (3.21) suggest that one can readily make the roughness-induced contribution to the linewidth quite appreciable.

The present results do not apply to the study of the width of the surface-polariton dip observed by the ATR method, since we ignore the effect of the prism coupler on the fields above the substrate. For example, in the presence of a prism with index of refraction n_p , the energy in the scattered waves

with $\omega/c < k_{||} < (\omega/c)n_p$ will be radiated through the prism coupler, while the description presented here ignores this possibility. This radiation has been studied in detail in a recent paper by Otto and Bodesheim,¹⁶ who have extracted information from it on the nature of $g(\vec{Q}_{||})$ for aluminum film.

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¹A recent brief review of the properties of surface polaritons may be found in Sec. X of the review article by D. L. Mills and E. Burstein, *Rep. Prog. Phys.* **37**, 817 (1974).

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¹²See in particular the review article by Otto cited in Ref. 2.

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¹⁵See the paper cited in Ref. 10.

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