Roughness and the mean free path of surface polaritons in tunnel-junction structures

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(Received 17 December 1979)

We calculate the mean free path of surface polaritons in tunnel junctions, in the presence of roughness on the surface of the outermost film. When these results are combined with our earlier theory of light emission from roughened junctions subject to a dc bias voltage, we discuss the saturation of the intensity of emitted light with increased roughness amplitude. Also, the probability that the surface polariton emits a photon into the vacuum during its lifetime is estimated and found to be very small for waves in the near infrared and visible frequency range.

I. INTRODUCTION

In the recent literature, Lambe and McCarthy have reported[1] light emission by tunnel junctions ($Au:Al_2O_3:Al$ and $Ag:Al_2O_3:Al$) biased by a dc voltage, and which have roughened surfaces. In an earlier paper, we have presented[2] a theory of this roughness-induced radiation, and have found in addition a contribution of appreciable intensity that comes from direct coupling between current fluctuations and the radiation field outside the junction. This latter contribution is present for a junction with perfectly smooth surface.

In our theory, which is valid in the limit of small-amplitude roughness, the roughness-induced radiation arises from a second-order process, as suggested by Lambe and McCarthy.[1] An electron creates a surface polariton as it tunnels through the oxide barrier, and in the second step, the roughness induces the surface polariton to radiate. The intensity of this portion of the radiation scales as δ^2, the square of the root-mean-square (rms) roughness amplitude δ. It is also proportional to the mean free path $l(\omega)$ of the surface polariton excited by the current fluctuations, which subsequently radiates into the vacuum by virtue of the roughness on the surface. The mean free path of the surface polariton may be regarded as the coherence length which controls the coupling between the surface polariton and the emitted photon.[3]

In our earlier paper,[2] where the roughness amplitude was assumed small, $l(\omega)$ was finite by virtue of the dissipation in the tunnel-junction structure produced by the surface polariton fields. We argued there that as δ^2 increases, in fact $l(\omega)$ must drop below the value $l^{(0)}(\omega)$ characteristic of the smooth surface, with the consequence that the radiation intensity must increase with δ more slowly than δ^2, for roughness amplitudes large enough for $l(\omega)$ to be significantly shorter than $l^{(0)}(\omega)$. In fact, if we assume perturbation theory adequately describes the damping of the surface polariton by roughness, then $l(\omega) \sim \delta^{-2}$ in the limit that roughness and not substrate dissipation controls the mean free path. The intensity of the emitted radiation should thus saturate as δ increases, within this simple picture. Such a saturation effect was reported prior to our work by Lambe and McCarthy.[4] These authors also comment that the saturation effect may have its origin in the fact that, as roughness increases in amplitude, the surface polariton fails to remain a well-defined elementary excitation of the structure.

The purpose of the present paper is to develop the theory of the influence of roughness on the mean free path of the surface polaritons that enter the theory of light emission from these structures. The analysis requires a nontrivial extension of an earlier calculation,[5] even though here, as before, we limit our attention to a perturbation theoretic treatment of the roughness. The reason is that the low-frequency (i.e., optical frequency) waves of interest have fields that extend throughout the structure. The oxide layer, though very thin, plays an essential role in binding the wave to the structure. We have thus carried out a new analysis of the interaction of surface polaritons with surface roughness, with the discussion framed in language that is rather general. The results of the numerical calculations, when combined with the analysis in the earlier paper, suggest that we can indeed obtain saturation of the radiation intensity for values of δ we believe are physically reasonable. We must append one qualification to this statement. In the saturation regime, the values we require for δ are likely attained in real junctions but they are uncomfortably large for use with our perturbation theoretic treatment of the roughness. It is difficult to carry through a more complete analysis, for these complex structures. It is our view that the calculations offer support for our earlier picture of the saturation mechanism, though the quantitative features of the results may be modified by a more complete theory, for the largest values of δ we use.

The calculation also provides insight into the physical picture that underlies the phenomenon of roughness-induced emission stimulated by the dc tunnel current. We write the surface polariton mean free path as a sum of three terms, which we denote here as

$$\frac{1}{l(\omega)}=\frac{1}{l_0(\omega)}+\frac{1}{l_{sp}(\omega)}+\frac{1}{l_R(\omega)} \quad . \tag{1.1}$$

The piece $l_0(\omega)^{-1}$, called $2Q_{\parallel}^{(1)}$ previously,[2] comes from dissipation in the substrate. Figure 2 of our earlier paper provides calculations of $Q_{\parallel}^{(1)}$, for a junction with smooth surfaces. (In the present paper, the mean free path is defined to be the distance for the energy density to drop to $1/e$ of its initial value, while $Q_{\parallel}^{(1)}$ is the inverse of the distance for the field amplitude to drop by the amount.)

Roughness limits the mean free path of the surface polariton in two ways.[5] First of all, a surface polariton of wave vector $\vec{k}_{\parallel}^{(0)}$ is elastically scattered into a surface polariton state of wave vector $\vec{k}_{\parallel} \neq \vec{k}_{\parallel}^{(0)}$. This contribution to the mean free path is $l_{sp}(\omega)$. Then the wave may radiate its energy into the vacuum above the crystal, to give rise to $l_R(\omega)$. We find here that $l_{sp}(\omega)$ and $l_0(\omega)$ can be the same order of magnitude for reasonable descriptions of the roughness. However, $l_R(\omega)$ is many orders of magnitude larger than $l_{sp}(\omega)$ or $l_0(\omega)$. Thus, the probability that the surface wave emits a photon is very small before its fields decay, or before its coupling to a particular radiative photon mode is interrupted by an elastic scattering. The tunnel junctions are, in fact, rather weak light sources, and it is this small radiative decay probability that is the source of the problem, since the fluctuations in tunneling current couple quite strongly to the surface mode. The fact that the roughness is an inefficient means of decoupling a surface polariton from a surface, on that portion of the surface-wave dispersion curve uninfluenced by retardation (where $c\vec{k}_{\parallel}^{(0)} >> \omega$, with c the vacuum velocity of light and ω the frequency of the surface polariton) has been pointed out in the earlier literature,[5,6] though the geometries explored are very different than the one of present interest.

Section II of this paper presents the theoretical discussion of the influence of roughness on the surface polariton mean free path, and our numerical calculations are discussed in Sec. III.

II. THEORETICAL ANALYSIS

The geometry we consider is the one employed in our earlier paper, and illustrated there in Fig. 1. We consider a semi-infinite substrate with dielectric $\epsilon_3(\omega)$. The substrate is overlaid with an oxide layer of thickness d, and this by a film of nominal thickness L. The dielectric constants of the oxide layer and of the film are $\epsilon_2(\omega)$ and $\epsilon_1(\omega)$, respectively. The coordinate system is chosen such that the z axis is normal to the surface of the substrate. The surface of the outermost layer is supposed to be roughened, in such a way that the location of a point on the surface is given by the relation $z = L + d + \zeta(\vec{x}_{\parallel})$ with $\vec{x}_{\parallel} = x\hat{x} + y\hat{y}$. The function $\zeta(\vec{x}_{\parallel})$ is defined so that its average value $\langle \zeta(\vec{x}_{\parallel}) \rangle$ vanishes. The root-mean-square value δ of the deviation of the outer surface from perfect flatness is defined as $\delta = [\langle \zeta^2(\vec{x}_{\parallel}) \rangle]^{1/2}$

To begin the discussion of the mean free path in the presence of roughness, we begin by looking for solutions of Maxwell's equations which vary harmonically with time

$$\vec{E}(\vec{x},t) = \vec{E}(\vec{x},\omega)e^{-i\omega t} \quad , \tag{2.1}$$

where the Fourier coefficient of the electric field $E(\vec{x},\omega)$ obeys

$$\vec{\nabla} \times [\vec{\nabla} \times \vec{E}(\vec{x},\omega)] - (\omega^2/c^2)\epsilon(\vec{x},\omega)\vec{E}(\vec{x},\omega) = 0 \quad . \tag{2.2}$$

The dielectric function $\epsilon(\vec{x},\omega)$ may be split into two parts

$$\epsilon(\vec{x},\omega) = \epsilon^{(0)}(z,\omega) + \Delta\epsilon(\vec{x},\omega) \quad . \tag{2.3}$$

The first term on the right-hand side is the dielectric function for the three-layer structure with smooth surfaces, therefore only a function of z, and the second term is the change in the dielectric function produced by roughness.

The equation for the Fourier component of the electric field can be written in the form

$$\vec{\nabla} \times [\vec{\nabla} \times \vec{E}(\vec{x},\omega)] - (\omega^2/c^2)\epsilon^{(0)}(z,\omega)\vec{E}(\vec{x},\omega) = (\omega^2/c^2)\Delta\epsilon(\vec{x},\omega)\vec{E}(\vec{x},\omega) \quad . \tag{2.4}$$

This can be converted into an integral equation through use of the Green's-function matrix $D_{\mu\lambda}^{(0)}(\vec{x},\vec{x}';\omega)$ constructed from Maxwell's equations

$$E_{\mu}(\vec{x},\omega) = E_{\mu}^{(0)}(\vec{x},\omega) - \frac{\omega^2}{4\pi c^2}\sum_{\lambda}\int d^3x' D_{\mu\lambda}^{(0)}(\vec{x},\vec{x}';\omega) \times \Delta\epsilon(\vec{x}',\omega)E_{\lambda}(\vec{x}',\omega) \quad . \tag{2.5}$$

Here $E_{\mu}^{(0)}(x,\omega)$ is a solution of Eq. (2.4) with $\Delta\epsilon(\vec{x},\omega)$ zero, and $D_{\mu\lambda}^{(0)}(\vec{x},\vec{x}';\omega)$ are the Green's functions for the structure with perfectly smooth surfaces. These are constructed as the solution of the equations

$$\sum_{\mu}\left(\frac{\omega^2}{c^2}\epsilon^{(0)}(z,\omega)\delta_{\lambda\mu} - \frac{\partial^2}{\partial x_{\lambda}\partial x_{\mu}} + \delta_{\lambda\mu}\nabla^2\right)D_{\mu\nu}^{(0)}(\vec{x},\vec{x}';\omega) = 4\pi\delta_{\lambda\nu}\delta(\vec{x}-\vec{x}') \quad . \tag{2.6}$$

We proceed by calculating the scattered electric field, produced by the second term on the right in Eq. (2.5), at point $\vec{x}$ in the vacuum above the sample. Upon taking into account the electromagnetic boundary conditions and then taking the limit of small amplitude roughness as described in I, one can find the following form for the scattered electric field:

$$E_{\mu}^{(s)}(\vec{x},\omega) = -\frac{\omega^2}{4\pi c^2}[\epsilon_1(\omega)-1]\sum_{\lambda}\int d^3x_{\parallel}' D_{\mu\lambda}^{(0)}(\vec{x}_{\parallel}z,\vec{x}_{\parallel}'(L+d)+;\omega)\zeta(\vec{x}_{\parallel}')E_{\lambda}(\vec{x}_{\parallel}',(L+d)-;\omega) \quad . \tag{2.7}$$

If we replace the electric field on the right-hand side of Eq. (2.7) by that of the surface polariton in the limit all surfaces are presumed smooth, then we obtain the amplitude of the scattered electric field to first order in roughness. We presume here the incident surface polariton propagates parallel to the $\hat{x}$ axis, so the expression for the scattered field assumes the form

$$E_{\mu}^{(s)}(\vec{x},\omega) = -\frac{\omega^2}{4\pi c^2}[\epsilon_1(\omega)-1]E^{(0)}\int d^2x_{\parallel}'\left[D_{\mu x}^{(0)}(\vec{x}_{\parallel}z,\vec{x}_{\parallel}'(L+d)+;\omega) + i\frac{k_{\parallel}^{(0)}}{\beta_0\epsilon_1(\omega)}D_{\mu z}^{(0)}(\vec{x}_{\parallel}z,\vec{x}_{\parallel}'(L+d)+;\omega)\right]e^{i\vec{k}_{\parallel}^{(0)}\cdot\vec{x}_{\parallel}'}\zeta(\vec{x}_{\parallel}') \quad . \tag{2.8}$$

In this expression, $E^{(0)}$ is the amplitude of the $\hat{x}$ component of the electric field in the surface polariton, evaluated just inside the surface of the upper film. We have, with $\vec{k}_{\parallel}^{(0)}$ the wave vector of the surface polariton parallel to the surface,

$$\beta_0 = (k_{\parallel}^{(0)2} - \omega^2/c^2)^{1/2} \quad , \tag{2.9}$$

where on the surface polariton dispersion curve, $k_{\parallel}^{(0)2} > \omega^2/c^2$ always.

Following the procedure employed in I, we take advantage of translational invariance in the two directions in the surface, for the smooth structure, by writing

$$D_{\mu\nu}^{(0)}(\vec{x},\vec{x}';\omega) = \int \frac{d^2k_{\parallel}}{(2\pi)^2}e^{i\vec{k}_{\parallel}\cdot(\vec{x}_{\parallel}-\vec{x}_{\parallel}')} \times d_{\mu\nu}^{(0)}(\vec{k}_{\parallel}\omega|zz') \quad . \tag{2.10}$$

The functions $d_{\mu\nu}^{(0)}(\vec{k}_{\parallel}\omega|zz')$ are given explicitly in the Appendix of I, for the structure employed in the present paper. Moreover, if we look at the scattered electric field in the limit $z > L+d$, then it is possible to introduce a new matrix $\gamma_{\mu\nu}(\vec{k}_{\parallel},\omega)$ such that

$$d_{\mu\nu}^{(0)}(\vec{k}_{\parallel}\omega|z(L+d)+) = e^{ik_z z}\gamma_{\mu\nu}(\vec{k}_{\parallel},\omega) \quad , \tag{2.11}$$

so when these results are assembled, we find Eq. (2.8) in the form

$$E_{\mu}^{(s)}(\vec{x},\omega) = -\frac{\omega^2[\epsilon_1(\omega)-1]}{16\pi^3c^2}E^{(0)}\int d^2k_{\parallel}e^{i\vec{k}\cdot\vec{x}}\zeta(\vec{k}_{\parallel}-\vec{k}_{\parallel}^{(0)})\left[\gamma_{\mu x}(\vec{k}_{\parallel},\omega)+i\frac{k_{\parallel}^{(0)}}{\epsilon_1(\omega)\beta_0}\gamma_{\mu z}(\vec{k}_{\parallel},\omega)\right] \quad , \tag{2.12}$$

where in Eq. (2.12), $\vec{k} = \vec{k}_{\parallel} + \hat{z}k_z$ and

$$k_z = (\omega^2/c^2 - k_{\parallel}^2)^{1/2}, \quad \mathrm{Im}(k_z) > 0 \quad , \tag{2.13}$$

and $\zeta(\vec{k}_{\parallel})$ is the Fourier transform of $\zeta(\vec{x}_{\parallel})$, defined by the same convention as that used in I.

The integral over $\vec{k}_{\parallel}$ covers all possible values of the two-dimensional wave vector. We confine our attention to two distinct regions of the $k_{\parallel}$ plane. In the region where $k_{\parallel} < \omega/c$, the quantity k_z is real (and chosen positive). This portion of the integral describes radiation emitted into the vacuum above the substrate. In the remainder of the $k_{\parallel}$ plane, where $k_{\parallel} > \omega/c$, k_z is purely imaginary with positive imaginary part. The contribution of this region describes electromagnetic fields localized near the surface of the tunnel-junction structure. This portion of the scattered field comes from roughness-induced elastic scattering of the incident surface polariton into a final surface polariton state with wave vector $\vec{k}_{\parallel} \pm \vec{k}_{\parallel}^{(0)}$. The former part of the integral contributes to $1/l^{(R)}$ in Eq. (1.1), while the second part contributes to $1/l^{(\mathrm{sp})}$. We consider each region separately.

A. Region $k_{\parallel} < \omega/c$

If we imagine the roughness is confined to a finite area of the surface, then in the limit $|\vec{x}| \to \infty$, the method of stationary phase may be used to evaluate the integral in Eq. (2.12). This method is described in I, and gives us the following expression for the scattered electric field in the vacuum:

$$E_{\mu}^{(R)}(\vec{x},\omega) = +i\frac{\omega^3(\epsilon_1(\omega)-1)}{8\pi^2c^3|\vec{x}|}E^{(0)}e^{i(\omega/c)|\vec{x}|}\zeta(\vec{k}_{\parallel}^{(s)}-\vec{k}_{\parallel}^{(0)})\cos\theta_s\left[\gamma_{\mu x}(\vec{k}_{\parallel}^{(s)},\omega)+\frac{k_{\parallel}^{(0)}}{\epsilon_1(\omega)\beta_0}\gamma_{\mu z}(\vec{k}_{\parallel}^{(s)},\omega)\right] \quad . \tag{2.14}$$

Here $\vec{k}_{\parallel}^{(s)} = (\omega/c)\sin\theta_s[\hat{x}\cos\phi_s + \hat{y}\sin\phi_s]$ is the projection of the wave vector of the scattered radiation onto a plane parallel to the surface, while θ_s and ϕ_s are the standard polar angles that describe the direction of the outgoing wave.

The next step in forming an expression of $1/l^{(R)}$ is to calculate the energy per unit time per unit solid angle carried away by the radiation field. This can be done by evaluating the time-averaged Poynting vector with the radiation field in Eq. (2.14), then multiplying by $|\vec{x}|^2$. In this way we obtain, with $(d^2W^{(R)}/d\Omega\, dt)\, d\Omega$ the energy per unit time radiated into solid angle $d\Omega$,

$$\frac{d^2W^{(R)}}{d\Omega\, dt} = \frac{\omega^4|\epsilon_1(\omega)-1|^2}{32\pi^3c^3}\cos^2\theta_s|\zeta(\vec{k}_{\parallel}^{(s)} - \vec{k}_{\parallel}^{(0)})|^2|E^{(0)}|^2 \times[\,|\cos\phi_s + i(k_{\parallel}^{(0)}/\epsilon_1\beta_0)\tan\theta_s|^2(|r_x(\vec{k}_{\parallel}^{(s)},\omega)|^2\cos^2\theta_s + |r_z(\vec{k}_{\parallel}^{(s)},\omega)|^2\sin^2\theta_s) + |r_y(\vec{k}_{\parallel}^{(s)},\omega)|^2\sec^2\theta_s\sin^2\phi_s\,]\,, \tag{2.15}$$

where the functions $r_\alpha(\vec{k}_{\parallel}^{(s)},\omega)$ are the same as those defined in I.

The calculation of the total energy per unit time radiated off the surface requires one integral over solid angle to be performed. The integral over the azimuthal angle ϕ_s can be performed analytically if it is noted that the functions $|r_\alpha(\vec{k}_{\parallel}^{(s)},\omega)|^2$ depend on only θ_s, and if we describe the surface roughness by the Gaussian distribution function commonly employed in theories of surface roughness phenomena

$$\langle\zeta(\vec{x}_{\parallel})\zeta(\vec{0})\rangle = \delta^2\exp(-x_{\parallel}^2/a^2)\,, \tag{2.16}$$

where δ is the rms amplitude of the roughness, and a is the transverse correlation length, which is a measure of the distance between adjacent peaks on the rough surface. The use of Eq. (2.16) combined with the definition of $|\zeta(\vec{k}_{\parallel}^{(s)} - \vec{k}_{\parallel}^{(0)})|^2$, with A the area of the rough patch on the surface, gives for the total energy per unit time radiated by the junction,

$$\frac{dW^{(R)}}{dt} = \frac{A\delta^2a^2\omega^4|\epsilon_1(\omega)-1|^2}{16\pi c^3}|E^{(0)}|^2\exp(-\tfrac{1}{4}a^2k_{\parallel}^{(0)2}) \times\int_0^{\pi/2} d\theta_s\sin\theta_s\cos^2\theta_s\exp\left(-\frac{a^2\omega^2}{4c^2}\sin^2\theta_s\right) \times\left[[\,|r_x(k_{\parallel}^{(s)},\omega)|^2\cos^2\theta_s + |r_z(k_{\parallel}^{(s)},\omega)|^2\sin^2\theta_s]\left[I_0(z) - \frac{I_1(z)}{z} + \left|\frac{k_{\parallel}^{(0)}}{\epsilon_1\beta_0}\right|^2\tan^2\theta_s I_0(z)\right] + \frac{|r_y(k_{\parallel}^{(s)},\omega)|^2 I_1(z)}{z}\right], \tag{2.17}$$

where $z = a^2\omega k_{\parallel}^{(0)}\sin\theta_s/2c$, and we are assuming $k_{\parallel}^{(0)}$ as well as $\epsilon_1(\omega)$ is purely real, an approximation that introduces little error.

To form an expression for the contribution $1/l^{(R)}$ to the mean free path of the surface polariton from radiation into the vacuum, we note the factor of $|E^{(0)}|^2$ renders $(dW^{(R)}/dt)$ is proportional to the energy per unit time $(dW^{(0)}/dt)$ in the surface polariton which flows over the rough patch on the surface. Let the patch be a rectangle with sides L_x and L_y, with the side L_y oriented perpendicular to $\hat{x}$, the propagation direction of the incident prolariton. Then $(dW^{(0)}/dt)$ is proportional to L_y, so Eq. (2.17) may be rewritten

$$\frac{dW^{(R)}}{dt} = \frac{L_x}{l_R(\omega)}\frac{dW^{(0)}}{dt}\,, \tag{2.18}$$

where $1/l_R(\omega)$ is the distance required for the energy stored in the surface polariton to decay to $1/e$ of its initial value, from roughness-induced radiation of its energy density into free space.

Thus, we require a form for $dW^{(0)}/dt$. This involves integrating the Poynting vector of the incident surface polariton from $z=-\infty$ to $+\infty$. The calculation is straightforward, but tedious, so we only quote the result here. We write it in the form

$$\frac{dW^{(0)}}{dt} = \frac{\omega k_{\parallel}^{(0)}L_y}{8\pi\beta_0^3}(1+f)|E^{(0)}|^2\,, \tag{2.19}$$

where the term proportional to f describes the fraction of the energy in the incident wave stored within the tunnel-junction structure, and remainder is the fraction which resides in the vacuum above the structure. The expression for f is quite complicated in form, and is reproduced in the Appendix. Recall that $|E^{(0)}|^2$ is the $\hat{x}$ component of the electric field of the incident polariton, evaluated at the outer surface $z = L + d$ of the tunnel-junction structure.

When these results are combined, for $1/l^{(R)}$ we find

$$\frac{1}{l_R(\omega)} = \frac{\delta^2 a^2 \omega^3 \beta_0^3 |\epsilon_1(\omega)-1|^2}{2c^3 k_{\parallel}^{(0)}(1+f)} \exp(-\tfrac{1}{4}a^2 k_{\parallel}^{(0)2}) \times \int_0^{\pi/2} d\theta_s \sin\theta_s \cos^2\theta_s \exp\left[-\frac{a^2\omega^2}{4c^2}\sin^2\theta_s\right] \times \left\{[|r_x(k_{\parallel}^{(s)},\omega)|^2\cos^2\theta_s + |r_z(k_{\parallel}^{(s)},\omega)|^2\sin^2\theta_s]\left[\left(1+\left|\frac{k_{\parallel}^{(0)}}{\epsilon_1\beta_0}\right|^2\tan^2\theta_s\right)I_0(z) - \frac{I_1(z)}{z}\right] + \frac{|r_y(k_{\parallel}^{(s)},\omega)|^2 I_1(z)}{z}\right\} . \tag{2.20}$$

In an earlier paper on the interaction of surface polaritons with surface roughness,[5] where the surface polariton propagated on the interface between a semi-infinite dielectric and vacuum, extensive use of the dispersion relation of the surface wave was used to simplify the expressions for the two contributions to the mean free path. This is difficult to do here, since the dispersion relation is given by a rather complicated equation displayed in I, which must be solved numerically save in certain special limits.

We turn next to evaluation of the contribution to Eq. (2.12) from the region $k_{\parallel} > \omega/c$.

B. Region $k_{\parallel} > \omega/c$

We again use the method of stationary phase, as outlined in I and in earlier work, to evaluate the contribution to the scattered field from the region $k_{\parallel} > \omega/c$. Here it is the limit $|\vec{x}_{\parallel}| \to \infty$, for fixed $z (z > L+d)$ that is of interest, where $\vec{x}_{\parallel}$ is the projection of $\vec{x}$ onto the plane parallel to the surface. After evaluating the expression for the scattered field in this manner, we take the limit $c \to \infty$, so retardation effects are ignored here. In I, and also in the earlier work by Davis,[7] it was pointed out that retardation has been negligible influence on the properties of surface polaritons in tunnel junctions, since the dimensions of the various elements are very small compared to the wavelength of light.

If $\vec{k}_{\parallel}^{(s)}$ is the wave vector of the scatter polariton (the magnitude of $\vec{K}_{\parallel}^{(s)}$ equals $\vec{k}_{\parallel}^{(0)}$, the wave vector of the incident surface polariton), the procedure above gives for the scattered field, in the region $z > L+d$,

$$\vec{E}^{(s)}(\vec{x},\omega) = \left(\hat{x}\frac{k_x^{(s)}}{k_{\parallel}^{(s)}} + \hat{y}\frac{k_y^{(s)}}{k_{\parallel}^{(s)}} + i\hat{z}\right)\frac{e^{ik_{\parallel}^{(0)}x}}{x^{1/2}} \exp[-k_{\parallel}^{(0)}(z-L-d)]E^{(s)} , \tag{2.21}$$

where

$$E^{(s)} = -\frac{2\sqrt{2}[\epsilon_1(\omega)-1]k_{\parallel}^{(0)}}{(i\pi)^{1/2}(\partial D/\partial k_{\parallel})_0}\left(if_x\cos\phi_s - i\frac{f_z}{\epsilon_1(\omega)}\right)E^{(0)}\zeta(\vec{k}_{\parallel}^{(s)} - \hat{x}\vec{k}_{\parallel}^{(0)}) . \tag{2.22}$$

The quantities f_x and f_z are given by

$$f_x = \exp[-2k_{\parallel}^{(0)}(L+d)]\left[\cosh(k_{\parallel}^{(0)}d)\cosh(k_{\parallel}^{(0)}L) + \frac{\epsilon_2(\omega)}{\epsilon_1(\omega)}\cosh(k_{\parallel}^{(0)}d)\sinh(k_{\parallel}^{(0)}L) + \frac{\epsilon_3(\omega)}{\epsilon_2(\omega)}\cosh(k_{\parallel}^{(0)}L)\sinh(k_{\parallel}^{(0)}L) + \frac{\epsilon_2(\omega)}{\epsilon_1(\omega)}\sinh(k_{\parallel}^{(0)}d)\sinh(k_{\parallel}^{(0)}L)\right] \tag{2.23a}$$

and

$$f_z = \exp[-2k_{\parallel}^{(0)}(L+d)]\{\epsilon_3(\omega)\cosh(k_{\parallel}^{(0)}d)\cosh(k_{\parallel}^{(0)}L) + \epsilon_1(\omega)\cosh(k_{\parallel}^{(0)}d)\sinh(k_{\parallel}^{(0)}L) + \epsilon_2(\omega)\sinh(k_{\parallel}^{(0)}d)\cosh(k_{\parallel}^{(0)}L) + [\epsilon_1(\omega)\epsilon_3(\omega)/\epsilon_2(\omega)]\sinh(k_{\parallel}^{(0)}d)\sinh(k_{\parallel}^{(0)}L)\} , \tag{2.23b}$$

while the function $D(k_{\parallel},\omega)$ whose derivative appears in the demoninator of Eq. (2.22) is

$$D(k_{\parallel},\omega)=[1+\epsilon_1(\omega)]\left[1+\frac{\epsilon_2(\omega)}{\epsilon_1(\omega)}\right]\left[1+\frac{\epsilon_3(\omega)}{\epsilon_2(\omega)}\right]+[1+\epsilon_1(\omega)]\left[1+\frac{\epsilon_2(\omega)}{\epsilon_1(\omega)}\right]\left[1-\frac{\epsilon_3(\omega)}{\epsilon_2(\omega)}\right]\exp(-2k_{\parallel}d)$$
$$+[1-\epsilon_1(\omega)]\left[1+\frac{\epsilon_3(\omega)}{\epsilon_2(\omega)}\right]\left[1+\frac{\epsilon_2(\omega)}{\epsilon_1(\omega)}\right]\exp(-2k_{\parallel}L)$$
$$+[1-\epsilon_1(\omega)]\left[1+\frac{\epsilon_2(\omega)}{\epsilon_1(\omega)}\right]\left[1-\frac{\epsilon_3(\omega)}{\epsilon_2(\omega)}\right]\exp[-2k_{\parallel}(L+d)]\ . \tag{2.24}$$

The dispersion relation of the surface polariton is given, in the absence of retardation, by the zeros of $D(k_{\parallel},\omega)$. To evaluate the scattered field in Eq. (2.21), we have used a "pole approximation" and extracted only the contribution from the surface polariton pole in the integration over the magnitude of $k_{\parallel}$ which remains after the method of stationary phase is used to perform the integration over the direction of $\vec{k}_{\parallel}$ in Eq. (2.12). (See Ref. 2 for an expanded description of this procedure.) In Eq. (2.22), it is the derivative of $D(k_{\parallel},\omega)$ evaluated at the surface polariton pole that appears in the denominator.

To form an expression for $1/l_{sp}(\omega)$, we proceed as follows. We can calculate the total energy per unit time fed into the surface polaritons noncolinear with the incident wave by forming the Poynting vector fom Eq. (2.21), then finding the total energy per unit time that flows through the walls of a cylinder of large radius erected with axis parallel to the surface normal. If we calculate this for the region $z > L+d$, the contribution from the region $z < L+d$ is found by simply multiplying the above by the same factor f that enters Eq. (2.19). The above procedure gives the total energy per unit time transported away from the rough patch on the surface by energy carried in the form of surface polaritons bound to the surface. Upon dividing by $dW^{(0)}/dt$, we obtain an expression similar to Eq. (2.18), which leads us to an explicit form for $1/l_{sp}(\omega)$

$$\frac{1}{l_{sp}(\omega)}=\frac{4\beta_0^2 k_{\parallel}^{(0)}}{\pi A}\frac{|\epsilon_1(\omega)-1|^2}{(\partial D/\partial k_{\parallel}|_0)^2}\int_0^{2\pi}d\phi_s|\zeta(\vec{k}_{\parallel}-\vec{k}_{\parallel}^{(0)})|^2\left|f_x\cos\phi_s-\frac{k_{\parallel}^{(0)2}}{\epsilon_1\beta_0^2}f_z\right|^2\ . \tag{2.25}$$

Upon recalling that $|\zeta(\vec{k}_{\parallel}-\vec{k}_{\parallel}^{(0)})|^2$ is proportional to the area A of the roughened region of the surface, we see that $l_{sp}(\omega)$ is in fact independent of the area A.

If the Gaussian form of the surface roughness correlation function $\langle\zeta(\vec{x}_{\parallel})\zeta(0)\rangle$ is used to evaluate the integral on ϕ_s in Eq. (2.21), we then find a closed analytic expression for $l_{sp}(\omega)^{-1}$

$$\frac{1}{l_{sp}(\omega)}=\frac{8\pi\delta^2a^2[\epsilon_1(\omega)-1]^2k_{\parallel}^{(0)}\beta_0^2}{(\partial D/\partial k_{\parallel}|_0)^2}\left[f_x^2[I_0(z)+I_z(z)]-\frac{4k_{\parallel}^{(0)2}}{\epsilon_1(\omega)\beta_0}f_xf_zI_1(z)+2\frac{k_{\parallel}^{(0)4}}{\epsilon_1^2\beta_0^4}f_z^2I_0(z)\right]\ . \tag{2.26}$$

where we have $z=\frac{1}{2}a^2k_{\parallel}^{(0)2}$.

The results displayed in Eqs. (2.15) and (2.20)–(2.22) are the principal results of the present paper. Section III is devoted to discussion of numerical calculations of $l_R(\omega)$ and $l_{sp}(\omega)$, and the implications of these calculations.

III. RESULTS AND DISCUSSION

In this section, we describe calculations of $1/l_{sp}(\omega)$, and $1/l_R(\omega)$ for a junction fabricated from an Al substrate and an Ag overlayer. In all the calculations, the thickness L of the Ag film was taken to be 200 Å, that of the Al_2O_3 oxide layer 30 Å, and the Gaussian correlation function in Eq. (2.16) was used to characterize the roughness on the surface.

In Fig. 1, we show $1/l_{sp}(\omega)$ calculated for the case where the rms amplitude of the roughness is 35 Å. For the transverse correlation length a, we have chosen $a=35$, 60, and 100 Å, respectively.

The most striking feature of these curves is the prominant peak in the scattering rate in and near the visible, for the curves with $a=60$ and 100 Å. As the transverse correlation length increases, the peak shifts toward the infrared. If one consults the dispersion relation for the surface polariton calculated and displayed in Fig. 2 of I, then one sees the peak in $1/l_{sp}(\omega)$ occurs at that frequency where $k_{\parallel}^{(0)}a\approx 1$. At this frequency, the wavelength of the surface polariton approximately matches the distance between adjacent maxima on the rough surface, and the wave is scattered strongly by the roughness. At lower frequencies, where its wavelength is much longer, the wave sees the average profile of the surface and scatters weakly, and at high frequencies it tends to

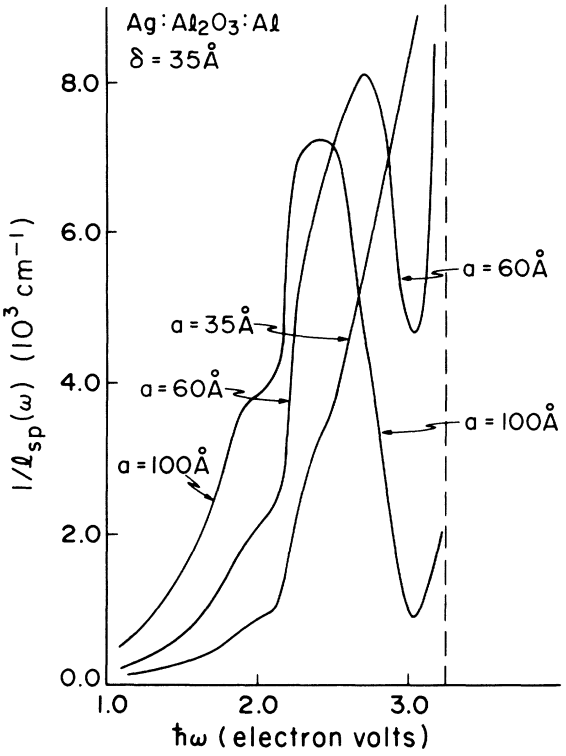

FIG. 1. Contribution $l_{sp}(\omega)$ to the surface polariton mean free path, for a Ag:Al_2O_3:Al junction. The thickness L of the Ag film has been taken to be 200 Å, that of the oxide layer is 30 Å, while $\delta = 200$ Å.

follow the contours on the surface adiabatically. The curve for $a = 35$ Å shows no maximum, since the condition $k_{\parallel}^{(0)} a \approx 1$ requires $k_{\parallel}^{(0)}$ to be sufficiently large that we are well out on the flat portion of the surface polariton dispersion curve close to the limiting frequency as $k_{\parallel}^{(0)} \to \infty$ determined by the requirement $\epsilon_1(\omega) + \epsilon_2(\omega) = 0$. For the Ag-$Al_2O_3$ interface, this limiting frequency is about 3.25 eV, for the parameters we have used.

The feature near 3.25 eV in $1/l_{sp}(\omega)$ visible in the curves with $a = 60$ and 100 Å occurs as one moves out on the flat portion of the dispersion curve. Mathematically, this upturn occurs because of the factor $(\partial D/\partial k_{\parallel})_0$ that appears squared in the denominator of Eq. (2.21). The origin of this factor may be understood as follows. Suppose instead of using the present Green's-function method, we calculated $1/l_{sp}(\omega)$ from a quantum-theoretic description by first calculating the scattering rate from the Golden Rule, then dividing by the group velocity $v_G(\omega)$ to form $1/l_{sp}(\omega)$. The scattering rate is proportional to the density of final states of the modes, which is proportional to $v_G(\omega)^{-1}$. Thus, $l_{sp}(\omega)^{-1}$ is proportional to $v_G(\omega)^{-2}$, and $v_G(\omega) = \partial\omega/\partial k_{\parallel}$ vanishes as $k_{\parallel} \to \infty$. In fact, we may also write $v_G(\omega) = (\partial D/\partial k_{\parallel})_0/(\partial D/\partial\omega)_0$, so we can appreciate the origin of the factor $(\partial D/\partial k_{\parallel})_0^2$ in the denominator of Eq. (2.21). [As ω approaches the limiting frequency of 3.25 eV, $(\partial D/\partial\omega)_0$ remains finite.] In the curve with $a = 35$ Å in Fig. 1, in effect the maximum where $k_{\parallel}^{(0)} a \approx 1$ has blended with the feature just described, to give $1/l_{sp}(\omega)$ a monotonic dependence on frequency.

We shall see shortly that $1/l_{sp}(\omega) \gg 1/l_R(\omega)$, as remarked in Sec. I. Thus, we may pause here to comment on the mechanism for saturation discussed in I, and also in Sec. I of the present paper. We can see that to produce saturation efficiently in the visible range of frequencies, we require roughness with a transverse scale in the range of 50–100 Å, which is the range which yielded spectra as calculated in I which are in qualitative accord with the data. As remarked in I, optical studies of surfaces of films prepared on CaF_2 substrates suggest the transverse correlation length is the order of 400 Å.[8] Such a long correlation length leads to emission spectra, calculated in I, that cutoff in the near infrared and give very little visible radiation. Thus, with a the order of 400 or 500 Å, our analysis is unable to account for emission spectra such as those reported by Lambe and McCarthy, and the proposed saturation mechanism will not operate, in the visible. This leads us to conclude here as in I that the junction surfaces must contain features with transverse scale in the range of 50 Å. We may imagine a surface that undulates slowly on the scale of 500 Å, as the optical experiments suggest, but with steps or terraces on a length scale an order of magnitude smaller.

We remark that the curves in Fig. 1 lean heavily on the Gaussian ansatz in Eq. (2.16), and thus may not accurately represent the statistical distribution of roughness on a real surface. We do believe, however, that for any picture that builds in a characteristic length to characterize the transverse scale in the roughness, one will find a maximum in $l_{sp}(\omega)^{-1}$ when $k_{\parallel}^{(0)} a \approx 1$.

One qualification must be added to the calculation in Fig. 1, in regard to its quantitative validity for a real junction structure. We have assumed the outer surface only is rough, while in reality each interface, including the Ag:Al_2O_3 and Al_2O_3:Al interface will also not be flat. We thus have in reality a complex problem in which each interface can be expected to be rough on the scale of a few tens of angstroms. To analyze this would be quite complex, and one would be led to a theory with numerous parameters few of which can be pinned down with reliability; the characteristics of the roughness on each interface will depend on the history of its preparation and chemistry, with that on the Al_2O_3:Al interface possibly rather different than that on the Ag vacuum interface.

One detailed analysis of the influence of roughness at an oxide overlayer on Al on the optical reflectivity of the structure has explored what happens when

both interfaces become rough.[9] The result depends on how the roughness on the inner interface is *correlated* with that on the outer interface. If the inner interface has a surface parallel to the oxide-vacuum surface, the result is a modest enhancement of the effect calculated with the Al:Al_2O_3 interface smooth, but the Al_2O_3: vacuum interface rough, If, however, the oxide is "lumpy", with thickness that varies with position, there is a dramatic one order of magnitude enhancement of the influence of roughness with given magnitude on the optical reflectivity. In the tunnel junctions, the oxide overlayer likely replicates roughness on the substrate, but the outer film evaporated later most probably has an outer surface with small scale features (steps, terraces) uncorrelated with those on the oxide. In this circumstance, the present calculation very likely underestimates the influence of roughness on the surface polariton mean free path, and provides a picture of the saturation mechanism that is unduly pessimistic, though the qualitative picture of the frequency variation we believe would be unaffected if all interfaces have roughness described by approximately the same transverse correlation length.

In Fig. 2(a), we show the variation of the emission intensity at 2.5 eV with rms roughness amplitude δ, for the model junction considered here. The saturation effect is clearly visible, though we have yet to achieve full saturation at $\delta = 200$ Å. We believe this calculation establishes the plausibility of the proposed saturation mechanism, particularly in view of the remarks above on the role of roughness on more than one interface.

One might expect a substantial frequency variation of the saturation effect, since from Fig. 1 we see that as one moves from visible frequencies into the infrared, $l_{sp}(\omega)$ lengthens appreciably. However, at the same time $l_0(\omega)$ increases also, and the ratio $r(\omega) = l_{sp}(\omega)/[l_0(\omega) + l_{sp}(\omega)]$ that controls the saturation effect does not display a strong frequency dependence as a consequence. [One is to multiply Eq. (3.4) of our earlier paper[2] by $r(\omega)$ to include the effect of roughness on the mean free path in the expression for the emission intensity.] We illustrate this point in Fig. 2(b), where we plot $r(\omega)$ as a function of photon energy for $a = 100$ Å and $\delta = 200$ Å. This ratio does depend on frequency, but not in a dramatic or particularly striking way. This suggests that a study of the saturation phenomenon as a function of frequency from the infrared through the visible should show no great dependence of this phenomenon on frequency.

We conclude with a discussion of our calculation of $1/l_R(\omega)$, the mean free path from radiation into the vacuum. For $\delta = 35$ Å, we display our calculations of $1/l_R(\omega)$ in Fig. 3. From the numbers and a comparison with Fig. 1, it is evident that $1/l_R(\omega)$ is smaller than $1/l_{sp}(\omega)$ and also $1/l_0(\omega)$ by roughly five orders

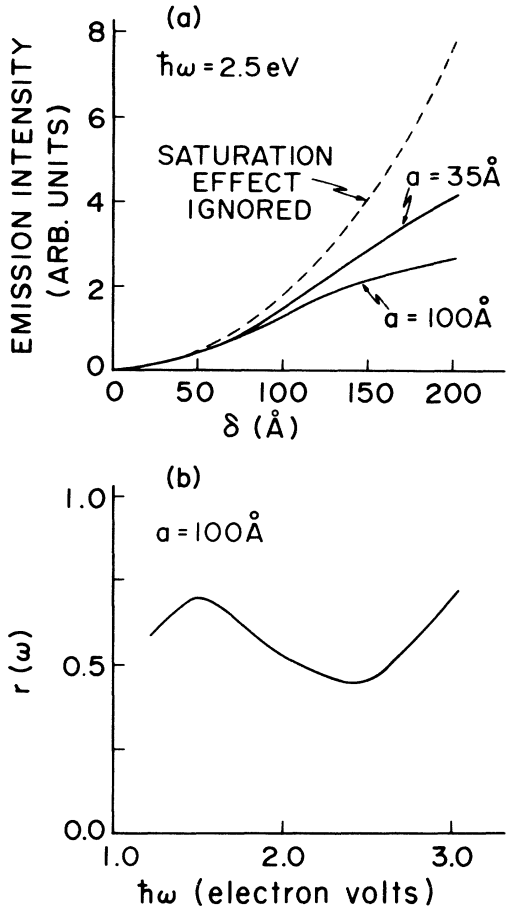

FIG. 2. (a) For $\hbar\omega$ and the junction considered in Fig. 1, we plot the variation of the emission intensity at 2.5 eV with rms amplitude δ. (b) For $\delta = 200$ Å and $a = 200$ Å, we plot as a function of photon frequency the ratio $r(\omega) = l_{sp}(\omega)/[l_0(\omega) + l_{sp}(\omega)]$ that controls the roughness-induced saturation effect.

of magnitude. This says not only can we neglect $1/l_R(\omega)$ in our discussion of the saturation effect, but also it is extremely improbable that the surface polariton will emit a photon into the vacuum, before its energy is dissipated by the currents induced in the substrate.

Upon elaborating on the preceding remark, we may see that the reason why the tunnel junctions are weak light emitters is that roughness is an inefficient means of decoupling energy stored in the surface polariton from the substrate. Indeed, a very short calculation allows us to understand the quantum efficiencies calculated in I.

An electron incident on a metal from the outside with velocity v_0 normal to the surface has a probability $P = \pi e^2/2\hbar v_0$ of exciting a surface plasmon.[10] Since the surface polariton fields inside the metal are quite comparable in strength with those outside, an

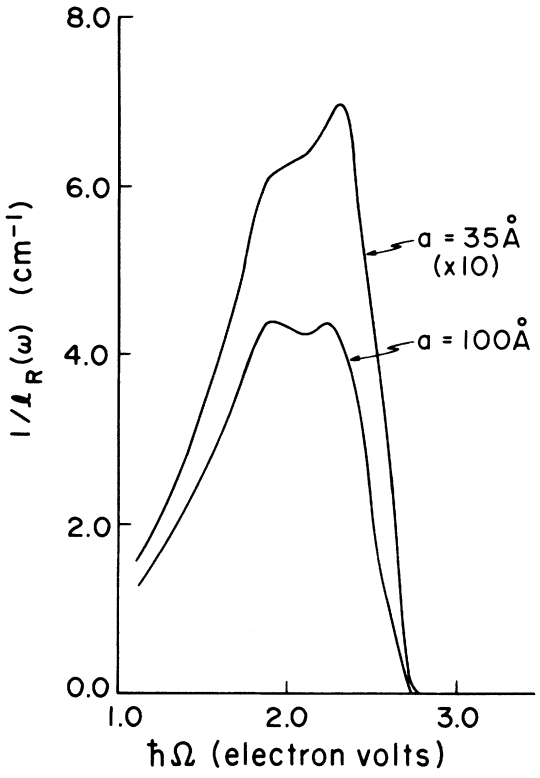

FIG. 3. For two values of a, $\delta = 35$ Å, and parameters used in the earlier calculations, we plot $1/l_R(\omega)$ as a function of photon energy. The curve for $a = 35$ Å has been multiplied by a factor of 10.

electron that tunnels through the barrier excites a surface polariton with about this probability. With $v_0 = 3 \times 10^8$ cm/sec characteristic of electrons at the Fermi surface of a metal, we have $P \approx 1$; i.e., a tunneling electron couples very strongly to the surface wave, and excites it with a probability rather close to unity.

Once the surface plasmon is excited, it must radiate a photon in the vacuum. The probability that this happens before the energy stored in the surface polaritons is dissipated by losses associated with the currents induced in the substrates is given by $l_0(\omega)/l_R(\omega)$, in the limit that the probability of photon emission is small. From the numbers displayed in Fig. 3, we estimate (for $\delta = 35$ Å and $a = 100$ Å) that in the visible range of frequencies $l_0(\omega)/l_R(\omega) \approx 4 \times 10^{-5}$, which with $P \approx 1$ gives an overall quantum efficienty quite close to that calculated in I.

We thus see very clearly from this example that the tunnel junctions are rather weak light emitters because the energy stored in the surface polariton fields near the surface is decoupled out ineffectively by surface roughness. The basic reason for this is that in the relevant range of frequencies, these modes have $ck_{\parallel}^{(0)} >> \omega$, and are thus really electrostatic normal modes of the structure, influenced only very slightly by retardation. If we adopt, in a heuristic sense here, language introduced with more precision elsewhere,[11] the "photon strength" of these modes is very small, with the consequence that they couple weakly with the radiation field, even if the perturbation they encounter is large (i.e., roughness with $k_{\parallel}^{(0)}a \approx 1$ and amplitude δ large enough that δ/L is not small). Such perturbations scatter the wave strongly [i.e., $l_{sp}(\omega)$ can become short and comparable to $l_0(\omega)$, thus leading to saturation of the emission], but they are unable to "trigger" the surface polariton, and induce it to radiate an appreciable fraction of its energy into the vacuum before its fields decay by other processes.

The above reasoning leads us to a pessimistic view of the possibility of fabricating junction structures with quantum efficiency substantially larger than the figures reported to date.[1] An interesting possibility, not considered here in detail, would be absorption of large molecules with size in the range of 50–100 Å onto the outer surface. Within the emission bands of the molecule, it is possible intensities substantially larger than those we estimate may be achieved.

ACKNOWLEDGMENTS

This research was supported by the Air Force Office of Scientific Research under Contract No. F49620-78-C-0019. One of us (B. L.) is partially supported by CNPq (Conselho Nacional de Desenvolvimento Cientifico e Tecnologico fron Brasil).

APPENDIX: EXPRESSION FOR THE ENERGY DENSITY OF THE SURFACE POLARITON STORED IN THE TUNNEL-JUNCTION STRUCTURE

Here we quote the expression for the quantity f that enters the main text, in Eq. (2.19) and Eq. (2.20). As remarked in the text, the derivation of this quantity is straightforward but tedious, so we quote only the final result, and we consider only the case where the three dielectric constants $\epsilon_1(\omega)$, $\epsilon_2(\omega)$, and $\epsilon_3(\omega)$ are all real.

We introduce, as in the text, $\beta_0 = (k_{\parallel}^{(0)2} - \omega^2/c^2)^{1/2}$, and $\beta_i = [k_{\parallel}^{(0)2} - \omega^2\epsilon_i(\omega)/c^2]^{1/2}$, where $i = 1$, 2, or 3. Then we let

$$\Lambda_{i0}^{(\pm)} = \epsilon_i\beta_0 \pm \beta_i \tag{A1}$$

and

$$\Lambda_{ij}^{(\pm)} = \epsilon_i\beta_j \pm \epsilon_j\beta_i \ . \tag{A2}$$

Then

$$f = (\beta_0/4\epsilon_1\beta_1^2)D \tag{A3}$$

and

$$
\begin{aligned}
D = {} & (1/\beta_1)\Lambda_{10}^{(-)2}[1-\exp(-2\beta_1 L)] + (1/\beta_1)\Lambda_{10}^{(+)2}[\exp(2\beta_1 L)-1] - 4L\,\Lambda_{10}^{(+)}\Lambda_{10}^{(-)} \\
& + (1/4\epsilon_1\epsilon_2\beta_2^3)\{[\Lambda_{21}^{(+)2}\Lambda_{10}^{(-)2}\exp(-2\beta_1 L) + \Lambda_{21}^{(-)2}\Lambda_{10}^{(+)2}\exp(2\beta_1 L) + 2\Lambda_{21}^{(+)}\Lambda_{21}^{(-)}\Lambda_{10}^{(+)}\Lambda_{10}^{(-)}][1-\exp(-2\beta_2 d)] \\
& \quad + [\Lambda_{21}^{(-)2}\Lambda_{10}^{(-)2}\exp(-2\beta_1 L) + \Lambda_{21}^{(+)2}\Lambda_{10}^{(-)2}\exp(2\beta_1 L) + 2\Lambda_{21}^{(-)}\Lambda_{21}^{(+)}\Lambda_{10}^{(+)}\Lambda_{10}^{(-)}][\exp(2\beta_2 d)-1]\} \\
& - (d/\epsilon_1\epsilon_2\beta_2^2)[\Lambda_{21}^{(+)}\Lambda_{21}^{(-)}\Lambda_{10}^{(-)2}\exp(-2\beta_1 L) + \Lambda_{21}^{(+)}\Lambda_{21}^{(-)}\Lambda_{10}^{(+)2}\exp(2\beta_1 L) + (\Lambda_{21}^{(+)2}+\Lambda_{21}^{(-)2})\Lambda_{10}^{(+)}\Lambda_{10}^{(-)}] \\
& + (\epsilon_3/4\epsilon_1\epsilon_2\beta_3^2)\{[\Lambda_{21}^{(+)}\Lambda_{10}^{(-)2}\exp(-2\beta_1 L) + \Lambda_{21}^{(-)2}\Lambda_{10}^{(+)2}\exp(+2\beta_1 L) + 2\Lambda_{21}^{(+)}\Lambda_{21}^{(-)}\Lambda_{10}^{(+)}\Lambda_{10}^{(-)}]\exp(-2\beta_2 d) \\
& \quad + [\Lambda_{21}^{(-)2}\Lambda_{10}^{(-)2}\exp(-2\beta_1 L) + \Lambda_{21}^{(+)2}\Lambda_{10}^{(+)2}\exp(+2\beta_1 L) + 2\Lambda_{21}^{(+)}\Lambda_{21}^{(-)}\Lambda_{10}^{(+)}\Lambda_{10}^{(-)}]\exp(+2\beta_2 d) \\
& \quad + 2[\Lambda_{21}^{(+)}\Lambda_{21}^{(-)}\Lambda_{10}^{(-)2}\exp(-2\beta_1 L) + \Lambda_{21}^{(+)}\Lambda_{21}^{(-)}\Lambda_{10}^{(+)2}\exp(2\beta_1 L) + (\Lambda_{21}^{(+)2}+\Lambda_{21}^{(-)2})\Lambda_{10}^{(+)}\Lambda_{10}^{(-)}]\} \ . \qquad \text{(A4)}
\end{aligned}
$$

In the limit $L \to 0$ and $d \to 0$, the quantity reduces to

$$f = \epsilon_3\beta_0^3/\beta_3^2 \ , \qquad \text{(A5)}$$

which, upon further use of the dispersion relation of a surface polariton which propagates on a semi-infinite dielectric of dielectric constant $\epsilon_3(\omega)$ reduces to the frequently quoted result[3,11]

$$f = -1/|\epsilon_3(\omega)|^2 \ . \qquad \text{(A6)}$$

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