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Observation and explanation of light-emission spectra from statistically rough Cu, Ag, and Au tunnel junctions

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A detailed description of the experimentally observed light output from statistically rough Al-Al_2O_3-M (M=Ag,Au,Cu) tunnel junctions is presented. These data include a comprehensive description of the polarization and angular distribution of the light emitted from Al-Al_2O_3-Au junctions as well as spectra from reverse-biased Al-Al_2O_3-Ag junctions. It is argued, principally on the grounds of an examination of surface-plasmon—polariton (SPP) damping, that the bulk of the output from statistically rough tunnel junctions is due to the *fast*-SPP mode. The idea of fast-SPP mediation is found, in many respects, to be much more consistent with currently available experimental results than that of slow- (or junction) SPP mediation. Extant theoretical models hold *slow*-SPP mediation to be the dominant means of visible-regime emission. The view of the emission mechanism presented in this paper suggests that the statistically rough tunnel junction could emit light more efficiently (if the scale of the surface roughness were altered) and that it has potential as a spectroscopic tool.

I. BACKGROUND

A. Survey of light-emitting tunnel junctions

Tunnel junctions may emit light provided they exhibit surface roughness on a microscopic scale. The exact scale and geometry of the roughness considerably influences the form of the spectral output; it is thus possible to characterize light-emitting tunnel junctions (LETJ's) by the type of roughness introduced in their fabrication. We distinguish three types of LETJ's: those incorporating small, discrete, metal particles; those grown on diffraction-grating substrates; and those which are statistically rough. Table I summarizes the principal features and currently accepted understanding of each. The roughness-induced scattering of a different plasmon mode leads in each case to a characteristic radiative output.

Tunnel junctions which incorporate small, discrete, metal particles as an integral part of the top electrode structure[1–4] yield a broadband, excess p-polarized output (i.e., p-s intensity) which is well described in the *localized*-plasmon model of Scalapino and co-workers.[5–8]

Equally, many aspects if the "intense," quasimonochromatic, p-polarized light emitted by grating LETJ's

TABLE I. Summary of principal features and currently accepted understanding of LETJ's. (a denotes the grating periodicity; σ denotes the transverse correlation length.)

	Metal-particle LETJ	Diffraction-grating LETJ	Statistically rough LETJ
Nature of surface roughness	Discrete metal particles of 10—100 nm diameter	Large scale (a=500—1000 nm) periodic "roughness"	Small scale ($\sigma\sim5$ nm) statistical roughness
Plasmon mode scattered	Localized plasmons	Fast-SPP mode at top electrode-vacuum interface	Slow or junction SPP mode
Light output	Broadband, substantially p-polarized	Sharp, intense, angle-tunable, p-polarized peaks against a weak unpolarized background	Broadband, essentially unpolarized
References to experimental work	1—4	9—14	1, 4, and 16—26
References to theoretical work	5—8	8 and 15	8 and 27—30

(Refs. 8–14) are displayed in the theory of Laks and Mills[15] (LM). Indeed, the grating LETJ affords the most conclusive evidence for the involvement of surface-plasmon polaritons (SPP's) in the light-emission process in tunnel junctions. As a result of the well-defined nature of the surface roughness and the precise energy and angle dependence of the p-polarized peaks, it is possible to construct a dispersion curve for the plasmon mode responsible for the light output[9,12] (cf. Ref. 31). Such measurements conclusively establish that the *fast*-SPP mode mediates the emission of light from grating LETJ's.

In addition to the characteristic, p-polarized emission from metal-particle LETJ's and grating LETJ's, there is an unpolarized output. It has generally been equated[4,9,14] to that from statistically rough LETJ's and attributed to the same processes as in these junctions.

Interestingly, despite the earlier experimental development of the statistically rough LETJ, a clear understanding of its emission processes has been less readily forthcoming than for the other devices. The gap in understanding is evidenced by significant disagreements[4,22,26] between experimental observations and the current theoretical descriptions of the output from statistically rough devices; we develop this matter in some detail below. Behind these disagreements the central issue requiring resolution is, as we see it, whether the light emission is predominantly slow SPP mediated or fast SPP mediated.

B. Preliminary discussion of the emission mechanism in statistically rough tunnel junctions

The spatial variations of the electric fields associated with both the fast- and slow-SPP modes for a typical Al-Al_2O_3-Ag tunnel-junction structure have been presented by Mills *et al.*[8] The electric fields of the slow-SPP mode are highly localized to the junction-interface region at visible frequencies. By contrast, the fields associated wth the fast mode decay slowly on the vacuum side of the Ag-vacuum interface. Consequently, a large proportion of the fast-mode energy resides in the vacuum outside the metal-film structure at visible wavelengths. It is for this reason that tunneling electrons couple to the fast mode very inefficiently.[8] On the other hand, the energy-density distribution of the fast mode means that it is much less subject, than the slow mode, to the strong internal damping mechanisms of the metal-film structure. The less severe the internal damping of a SPP mode, the greater are its chances of undergoing radiative decay.[29] Thus, the very feature which mitigates against efficient excitation of the fast mode favors its decay into photons, and conversely for the slow mode. The relative excitation and radiative-decay efficiencies of the fast- and slow-SPP modes determine the relative contribution of each to the final radiative output of a tunnel junction. Commonly,[15,16,27,28] it has been thought that the excitation of the fast mode in the statistically rough LETJ proceeds so inefficiently that its contribution to the radiative output is negligible. [In a recent theory of Arya and Zeyher[30] (AZ), the role of the fast mode is reconsidered, but even here it is found that the bulk of the visible output is slow SPP mediated.] Kroó *et al.*[20] have taken the opposite view, namely that the slow mode is so much more severely damped than the fast mode that the emitted light must be fast mode mediated. However, the experimental evidence which Kroó *et al.* present in favor of fast-mode mediation is not, as Kirtley *et al.*[10] point out, open to unambiguous interpretation.

In previous publications[24–26] we have presented findings which strongly favor the view that the emission from statistically rough LETJ's is fast SPP mediated. Our approach to the problem is somewhat different from that of the theorists and from that of Kroó *et al.* Here, we bring together our previous deliberations into a coherent whole and present further results and arguments in support of fast SPP mediation.

The remainder of this paper is arranged as follows. Section II describes the experiments, and Sec. III contains the core of the evidence and argument for fast-mode mediation, a view which is consolidated with various corroborative data given in Sec. IV. Sec. V presents a critical discussion of the surface roughness employed in LETJ fabrication and also attempts to place the LETJ in the context of related experimental work. Finally, Sec. VI summarizes our conclusions.

TABLE II. Details concerning the fabrication of LETJ's.

Material	Evaporation source	Pressure (Torr)	Rate of evaporation (nm/min)	Final film thickness (nm)
CaF_2	Molybdenum boat	Rising to $\sim 2\times 10^{-4}$	20–30	100–140
Al	Tungsten filament	$\sim 10^{-6}$	30–40	50–70
Al_2O_3	Al exposed to oxygen glow discharge at pressure of ~ 6 Pa with current of 10 mA for 5 min			2–3
M (Ag, Au, or Cu)	Tungsten filament	10^{-6}–10^{-5}	15–25	25–40

II. EXPERIMENTAL PROCEDURE

A. Junction fabrication

We have examined the light output from statistically rough tunnel junctions of the type Al-Al_2O_3-M where M=Ag, Au, or Cu. The various layers of the structure were laid down in the order and under the conditions described in Table II. The necessary roughness was produced by an underlayer of CaF_2 evaporated directly onto the glass slide; a thickness of ~ 120 nm was chosen in order to achieve an optimum output[1] from the devices. Each of the metal films is ~ 7 mm wide, resulting in a large junction area of ~ 0.5 cm^2. The necessary electrical contacts are formed by narrow (~ 1 mm) evaporated Al strips, thus avoiding the problem of wires obstructing the path of the emitted light; this consideration is important for the angle-resolved measurements. A further useful feature is that the glass side is supported from its "top" edge only—electrical connection of the Al contact strips with the external circuitry is thereby also simply achieved. This arrangement has the advantage that light from the rear (substrate) side of the structure may be observed.

B. Optical-detection system

A schematic representation of the optical and electronic system employed in the investigation of the emitted-light spectra is shown in Fig. 1. The final product of the system is a graph of uncalibrated, recorded intensity versus wavelength or spectrum of the light emitted from a tunnel junction. Included in this spectrum, however, are the spectral responses of the monochromator and photomultiplier. The spectra response of the monochromator to p-polarized light relative to that of s-polarized light was ascertained using a tungsten-halogen lamp. The manufacturer's test data relating the response of the particular grating, in the Littrow configuration, to s-polarized light, were modified, according to the treatment of Whitaker,[32] to take account of reflections at the monochromator mirrors. The modified s-response curve was combined with experimental data to yield a p-response curve (and thence also a response curve for unpolarized light) for the monochromator. These, together with manufacturer's data for the photomultiplier tube, allowed curves to be obtained describing the spectral response of the optical system to s-polarized, p-polarized, and unpolarized light. Thence the recorded spectra could be calibrated.

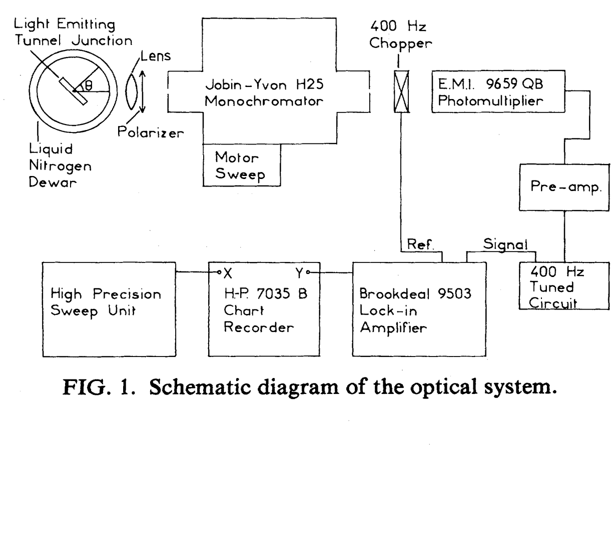

FIG. 1. Schematic diagram of the optical system.

III. SPECTRAL SHAPES AND THE EMISSION MECHANISM

A. Comparison of experimental and theoretical results

The corrected spectra of Fig. 2 are those of a Ag LETJ biased at different values of tunneling current; the emission angle θ is 0°. For each spectrum the maximum photon energy $\hbar\omega_{max}$ is within a few percent of that given by the simple quantum relation[16]

$$\hbar\omega_{max} = |eV_0| , \tag{1}$$

where V_0 is the value of the dc bias applied across the junction and e it the electronic charge. The general profiles and progression of the spectra (with increasing values of the tunneling current) are quite consistent with the visually observed changes in color from red, through orange and yellow, to blue. It is worth noting that the spectra of Fig. 2 are particularly simple in form (additional structure has sometimes been found superimposed[20,22]). The form of the spectra is in reasonably good agreement with the theoretical calculations of LM (Ref. 28) and Arya and Zeyher (AZ) (Ref. 30). However, since Ag is an optically well-behaved metal in the visible region, the Ag LETJ offers limited ground for a critical evaluation of the theoretical models. (However, see Sec. III F.)

More interesting is a typical series of corrected emission spectra from an Au LETJ as presented in Fig. 3 and from a Cu LETJ in Fig. 4. Both types of junction consistently show a marked drop in the intensity at higher energy (see also Refs. 1, 4, 16, and 17 for spectra from Au LETJ's). As yet, there is no theoretical description of the output

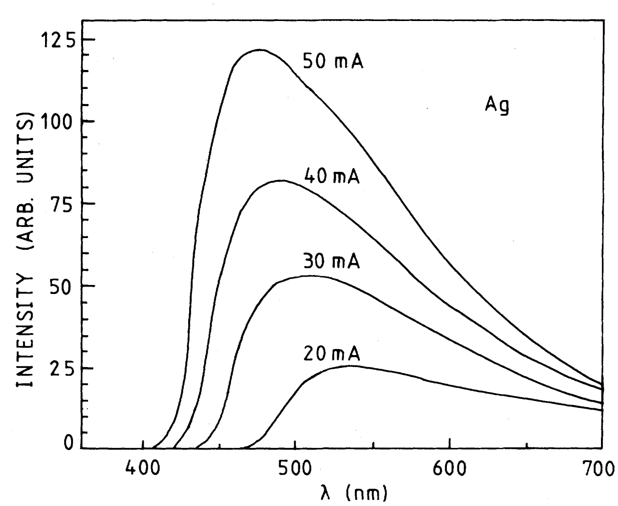

FIG. 2. Observed spectra of the light emitted by statistically rough Al-Al_2O_3-Ag junction biased with various values of tunneling current: i=20 mA (V_0=2.70 V), 30 mA (V_0=2.85 V), 40 mA (V_0=2.92 V), and 50 mA (V_0=3.00 V). θ=0°. Optical-system response corrections have been applied.

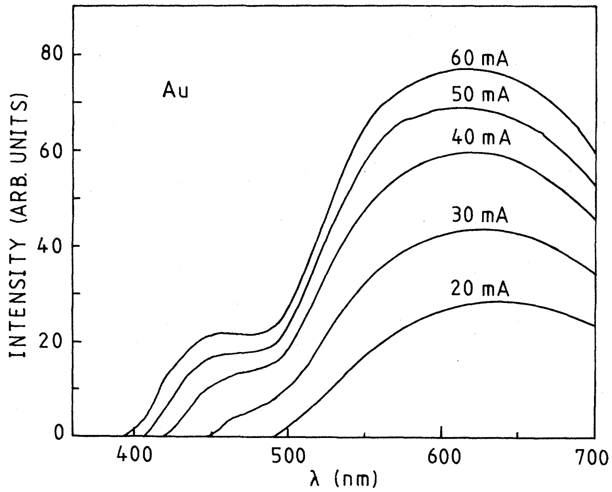

FIG. 3. Observed spectra of light emitted by statistically rough Al-Al_2O_3-Au junction biased with various values of tunneling current: $i=20$ mA ($V_0=2.56$ V), 30 mA ($V_0=2.77$ V), 40 mA ($V_0=2.86$ V), 50 mA ($V_0=2.96$ V), and 60 mA ($V_0=3.05$ V). $\theta=0°$. Optical-system response corrections have been applied.

from Cu LETJ's. In the case of the Au LETJ's the theoretical models of either LM (Ref. 28) or AZ (Ref. 30) have difficulty in describing the quite substantial output of light above the maximum energy (2.2 eV) of the slow-, or junction, SPP mode; both models hold the slow mode to be primarily responsible for mediating the visible output. In order to account for any output at higher energy (>2.2 eV) from such devices, the LM model points to a direct-emission process whereby fluctuations in the tunneling current couple directly to the radiation field. However, this process yields an intensity peak at 2.5 eV, where

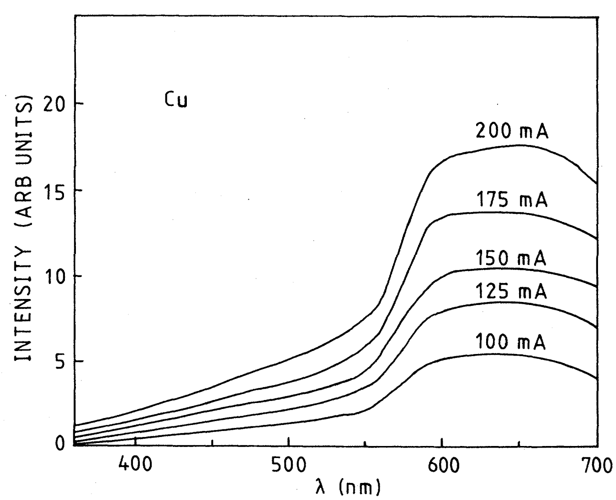

FIG. 4. Observed spectra of light emitted by statistically rough Al-Al_2O_3-Cu junction biased with various values of tunneling current: $i=100$ mA ($V_0=3.56$ V), 125 mA ($V_0=3.70$ V), 150 mA ($V_0=3.75$ V), 175 mA ($V_0=3.78$ V), and 200 mA ($V_0=3.80$ V). $\theta=0°$. Optical-system response corrections have been applied.

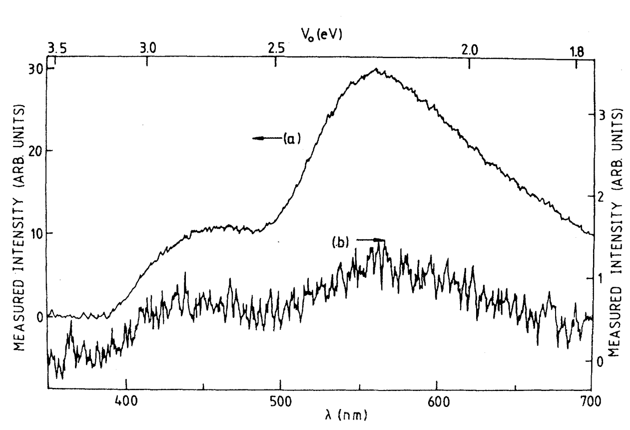

FIG. 5. Recorded (uncorrected) intensity signal vs wavelength plot of (a) forward ($\theta=0°$) and (b) rear ($\theta=180°$) light emitted from Al-Al_2O_3-Au junction biased with 50-mA current. Sensitivity is higher by a factor of 10 for curve (b).

we in fact observe a sharp dip. The AZ model adopts a more rigorous, nonperturbative treatment of the surface roughness in which multiple scattering of SPP's is included. The effect is to boost considerably (by a factor ~ 6) the intensity of the SPP-mediated emission. Consequently, the direct-emission intensity is insignificant by comparison. Thus, no account of significant emission above ~ 2.2 eV is given by the AZ model.

Certainly in the case of the Au LETJ, experiment and theory are at considerable variance. Rather than attempt a reconcilation (this exercise has been carried out by Parvin and Parker[22] in any case), we shall endeavor to construct a more straightforward and physically direct explanation of the spectral output from LETJ's.

B. Rear emission and the possibility of junction photons

It has been suggested by Adams and Hansma[4] that the dip in the spectra of Au LETJ's is due to the absorption of photons above 2.5 eV as they pass through the Au electrode (Au appears yellowish since is absorbs blue-green light). This view assumes, of course, that the SPP photon decay occurs in the junction region of the structure. However, we would argue against this idea on several grounds.

Our first argument concerns a feature of the LETJ which as yet has not been reported by other workers, namely the emission of light from the rear (or substrate) side of the device.[25] In both the case of the Au LETJ (Fig. 5) and the Ag LETJ (Fig. 16), the rear-emission spectrum is similar in structure to the forward-emission spectrum. This is an interesting observation in the case of the Au LETJ. The characteristic structure at 2.5 eV in the rear-emission spectrum is quite contrary to any expectations based on the postulate of light emission at the junction interface and the subsequent absorption of light in the base Al electrode. Furthermore, for the particular Au LETJ whose output is described by Fig. 5, the optical transmission of the Al base film was 0.2% at 500 nm, whereas that of the Au top film was 20%. Hence light generated at the oxide-metal interface would be weaker in the rear direction by a factor of ~ 100, not ~ 20 as observed.

Secondly, if the optical-absorption explanation is correct, then the structure at 2.5 eV in the spectra of Au LETJ's should reflect the variation in the imaginary part of the dielectric function ϵ for Au.[33] However, an inspection of ϵ_i for Au (Ref. 34) shows that it does not vary dramatically enough in a sufficiently narrow energy range to account satisfactorily for the observed spectral shape of Fig. 3. Similarly, the variation in the imaginary part of the dielectric function for Cu (Ref. 34) is too weak to account for the sharp decrease at 2.25 eV in the output from Cu LETJ's.

Thirdly, there are the results of other workers. Kroó *et al.* found[20] that the experimentally observed dependence of the emitted-light intensity on the top-electrode (Ag) thickness did not correlate with the optical attenuation length for Ag. Furthermore, Kirtley *et al.*, in their investigation of LETJ's grown on grating substrates, found that the rate of decay in the emitted intensity with top-electrode thickness was very much dependent on the detailed morphology of the top electrode.[10,11] Depending on the conditions of evaporation of the top electrode, the characteristic decay thickness could be made larger or smaller than the optical attenuation length.

Drawing together the various arguments, it is clear that the light emitted from statistically rough tunnel junctions does not originate at the oxide-metal interface.

C. Hot electrons and the volume-loss function

A further decisive step towards a simple physical understanding of statistically rough LETJ's occurred when it was found that the spectra from Au and Cu LETJ's appeared to image, very directly, the volume-loss function $\mathrm{Im}(-1/\epsilon)$.[24] Here, ϵ is the dielectric function of the top metal electrode. (See Figs. 6 and 7.) The volume-loss function exhibits a sharp increase in the very region where the light output drops off. In both cases the increase is attributed to an interband transition, which for Au (at 2.5 eV) has been described in the relativistic band-structure calculations of Christensen and Seraphin,[35]

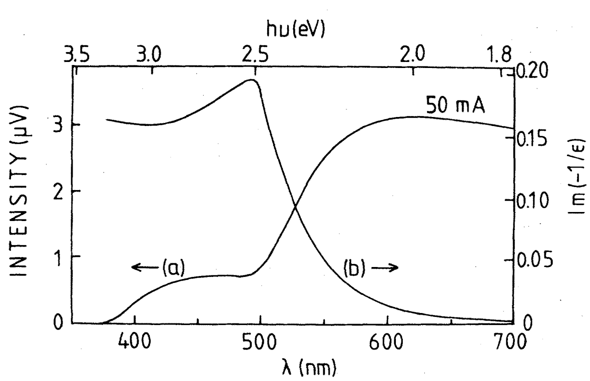

FIG. 6. (a) Observed spectrum light emitted from Al-Al_3O_3-Au junction biased at 3.28 V ($i=50$ mA). Units correspond to signal input to lock-in detector but with optical-system response corrections applied. (b) Electron-energy-loss function of Au calculated from optical data of Ref. 34.

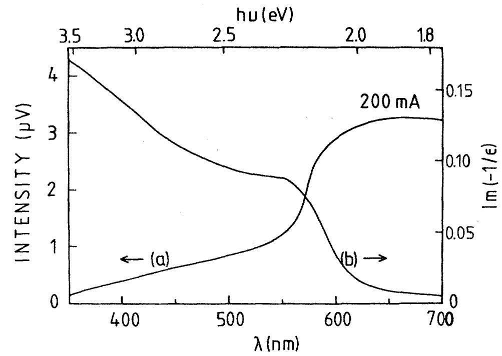

FIG. 7. (a) Observed spectrum of light emitted from Al-Al_2O_3-Cu junction biased at 3.80 V ($i=200$ mA). Units correspond to signal input to lock-in detector but with optical-system response corrections applied. (b) Electron-energy-loss function of Cu calculated from optical data of Ref. 34.

while the transition at 2.25 eV in Cu was identified some time ago by Ehrenreich and Phillipp.[36] Throughout the visible range, both ϵ and $\mathrm{Im}(-1/\epsilon)$ for the lower electrode, Al, are smoothly varying functions.

It is very surprising that the volume-loss function should account so well for the spectra of these thin-film devices. Traditionally, the function has been used to describe the energy loss of fast (~ 10 keV) electrons transmitted through thin metal films.[37] We might therefore suggest that the dip in the spectra of Figs. 6 and 7 towards higher energies occurs as a result of the absorption of hot tunneling electrons passing through the top metal film, from the junction interface to the outermost surface, prior to exciting fast SPP's. In other words, the output from statistically rough Au and Cu LETJ's essentially reflects the hot-electron-energy distribution incident on the outermost metal-air surface. This distribution is directly related to $\mathrm{Im}(-1/\epsilon)$; its sharp rise in each of the cases of Au and Cu is due to the excitation of interband transitions. Although such an interpretation of the results is in harmony with the traditional use of $\mathrm{Im}(-1/\epsilon)$, there is a strong counterargument. Consider, for example, the case of Au. The attenuation length[38] of 2-eV electrons in Au is ~ 10 nm and is comparable with the thicknesses, ~ 30 nm, of our films; thus we would anticipate two or three scatterings of hot electrons in transit through the film. These scatterings are from the free-electron gas. By comparison, scatterings which involve the excitations of an interband transition will be very much less common, probably by several orders of magnitude, as a consequence of the relatively low density of electrons in appropriate initial states. Hence the attenuation length associated with the interband excitation is expected to be very much greater than the film thickness and therefore the loss peak due to this process is unlikely to be significantly imaged in the electron distribution at the Au-air surface or in the emission spectra.

D. SPP damping

What then gives rise to the observed spectral shapes? Previously, we surmised that it was probably the damping of the SPP's themselves. Rather than pursue here a slightly tenuous line of argument relating the damping of a collective electron excitation to the bulk loss function of a single electron, we may consider the damping explicitly. First we recognize, with reference to Au LETJ's, that if the slow-mode SPP cuts off[28] at 2.2 eV, but the fast mode extends beond this energy,[39] then an examination of the damping of the latter mode should be instructive.

The fast-SPP mode, with its associated fields peaked at the outer surface of the top electrode, is, in fact, closely similar to the mode that exists at the boundary between a semi-infinite metal and a semi-infinite insulator.[8,40,41] The wave vector is given by the well-known relation

$$K=\frac{\omega}{c}\left[\frac{\epsilon_0\epsilon_1}{\epsilon_0+\epsilon_1}\right]^{1/2}, \tag{2}$$

where ϵ_0 is the dielectric response function of the insulator and ϵ_1 that of the metal. ϵ_1 is, of course, a complex quantity:

$$\epsilon_1=\epsilon_r+i\epsilon_i , \tag{3}$$

where ϵ_r nd ϵ_i denote the real and imaginary parts, respectively. The spatial damping of a SPP mode is given by the imaginary part of its wave vector, K_i. With some algebraic manipulation, it can be shown that, for the fast-SPP mode,

$$K_i=\pm\frac{\omega}{c}\left[\frac{-P\pm(P^2+R^2)^{1/2}}{2Q}\right]^{1/2}, \tag{4}$$

where

$$P=\epsilon_r\epsilon_0(\epsilon_r+\epsilon_0)+\epsilon_i^2\epsilon_0 ,$$
$$Q=(\epsilon_r+\epsilon_0)^2+\epsilon_i^2 ,$$
$$R=\epsilon_0^2\epsilon_i .$$

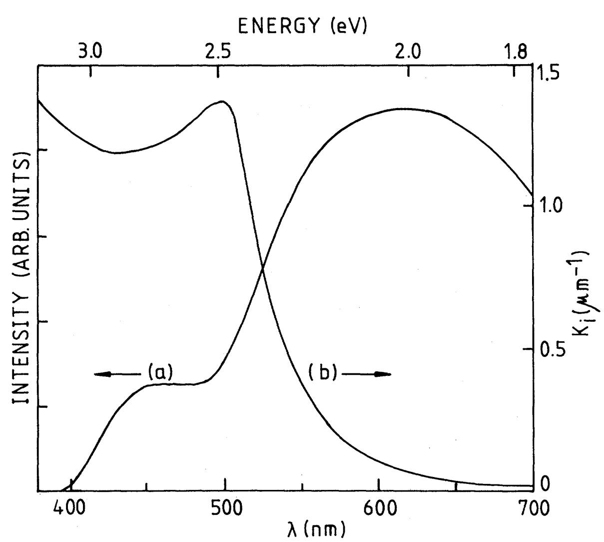

FIG. 8. (a) Observed spectrum of light emitted by Al-Al_2O_3-Au junction biased at 3.05 V (i=60 mA). Optical-system response corrections have been applied. (b) Imaginary part of wave vector K_i for SPP mode at Au—liquid-nitrogen interface; K_i was calculated from Eq. (4) using optical data of Ref. 34.

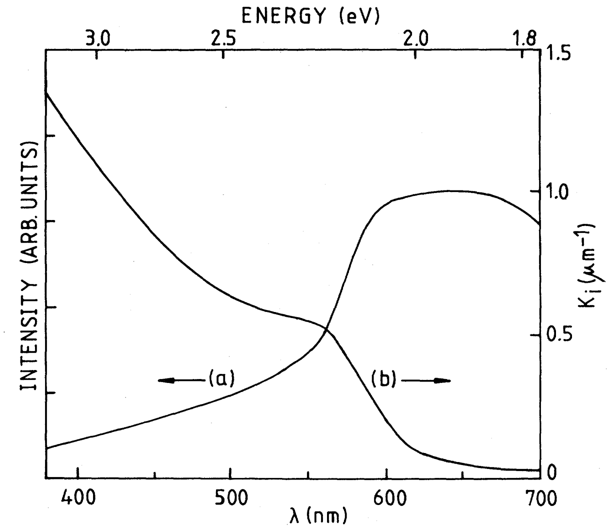

FIG. 9. (a) Observed spectrum of light emitted by Al-Al_2O_3-Cu junction biased at 3.80 V (i=200 mA). Optical-system response corrections have been applied. (b) Imaginary part of wave vector K_i for SPP mode at Cu—liquid-nitrogen interface; K_i was calculated from Eq. (4) using optical data of Ref. 34.

In order to have physically meaningful solutions, the positive sign is chosen in each case where Eq. (4) offers the choice. K_i was evaluated in the visible energy range for the SPP mode at Au—Cu—, and Ag—liquid-nitrogen (ϵ_0=1.45) interfaces using the optical data of Johnson and Christy;[34] the choice of liquid nitrogen for the insulator corresponds to our experimental arrangement. Commonly, the same quantity is calculated for infrared wavelengths in surface-electromagnetic-wave (SEW-) spectroscopy studies.[42] The spectrum of the light emitted from an Au LETJ and the corresponding damping curve are shown as Figs. 8(a) and 8(b), respectively; Fig. 9 presents the same information for a Cu LETJ. [In each case the damping curve closely resembles the corresponding Im($-1/\epsilon$) curve (cf. Figs. 6 and 7); the same is also true for Ag. This is an intriguing observation which, as far as we are aware, has not previously been made; it is perhaps worthy of further, more quantitative, investigation.]

Figures 8 and 9 being us directly to a much clearer understanding of the light-emission process. The information conveyed by these figures constitutes strong evidence that the bulk of the light emission from *statistically* rough tunnel junctions is mediated by the *fast*-SPP mode. The energy-dependent internal damping of the *fast*-SPP mode appears to dominate the spectral shapes. The sharp rise in K_i, above ~2.5 eV for Au and ~2.25 eV for Cu, is associated with the onset of interband transitions[35,36] which offer an alternative decay mode for the higher-energy fast SPP's. This decay mode competes most effectively with the radiative decay of the SPP's and, consequently, the spectrum of the emitted light dips sharply above the threshold for interband transitions.

To confirm the model, the spectrum for an Ag LETJ and the damping curve for Ag—liquid-nitrogen SPP's are presented in Fig. 10. In contrast to Au and Cu, Ag has no interband-transition thresholds in the visible energy range and, consequently, the damping curve is featureless. As

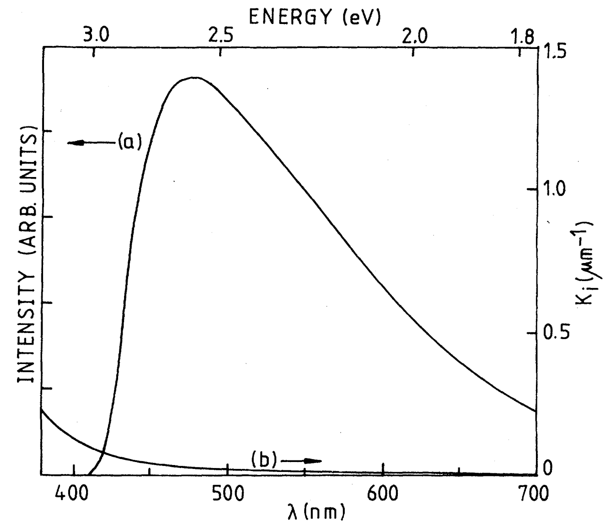

FIG. 10. (a) Observed spectrum of light emitted by Al-Al_2O_3-Ag junction biased at 3.00 V ($i=50$ mA). Optical-system response corrections have been applied. (b) Imaginary part of wave vector K_i for SPP mode at Ag—liquid-nitrogen interface; K_i was calculated from Eq. (4) using optical data of Ref. 34.

expected, the emitted-light spectrum also is featureless in that there is no strong "absorption" of higher-energy photons.

E. A slow-mode contribution?

Consider first the Cu LETJ. As yet, no calculation of the slow-mode damping curve nor the asymptotic energy of this mode has been reported in the literature. However, the asymptotic energy may be estimated from the approximate, but physically justifiable, condition[8]

$$\epsilon_r = -\epsilon_{Al_2O_3} \; . \tag{5}$$

Setting $\epsilon_{Al_2O_3}=3.0$ and using the optical data of Johnson and Christy[34] to evaluate ϵ_r, a slow-mode cutoff energy of 3.04 eV is obtained. Following the same procedure for the case of Au LETJ's yields a slow-SPP cutoff at 2.4 eV which is not far removed from the value of 2.2 eV rigorously calculated by LM.[28] For each type of junction it is noteworthy that the spectra fail to cut off at the slow-mode maximum energy. Moreover, they display no discernable structure around this energy. These observations lead us to conclude that there is an insignificant slow-mode contribution to the visible output from statistically rough tunnel junctions. The characteristic shapes of Figs. 3 and 4 may thus be unambiguously attributed to the form of the fast-SPP damping curves. We further note that in each type (Cu or Au) of junction the fast mode is available to mediate radiation well into the uv. Using the data of Ref. 34, we calculate the cutoff energy of the fast SPP to be 3.76 eV for the Cu—liquid-nitrogen interface and 4.86 eV for the Au—liquid-nitrogen interface.

F. Antenna factor and power-density spectrum

The spectra for Au and Cu LETJ's can be regarded as simple Ag-type spectra with the high-energy end substantially attenuated. The matter of explaining the simple spectrum from Ag LETJ's remains. Clearly, these devices are much more efficient towards the blue. Indeed, the high-energy end of the spectra of Au and Cu LETJ's is also much more intense than would be anticipated on the basis of SPP-damping considerations alone. This observation is more evident from an examination of Fig. 11 rather than Figs. 8—10. The damping is portrayed in Fig. 11 as a plot of L versus λ for the Ag—, Au—, and Cu—liquid-nitrogen SPP's. Here L is the SPP propagation length and is related to K_i through the simple equation

$$L=(2K_i)^{-1} \; . \tag{6}$$

What then gives rise to the blue efficiency of the statistically rough tunnel junction?

In their work on grating LETJ's, Kirtley *et al.*[10] isolated an experimental "antenna-factor" curve which showed the LETJ to be a much more efficient antenna (by a factor $\sim 10^3$) at the blue end of the spectrum than at the red end. Such behavior is due[8,10] to the progressive localization (with increasing energy) of the fast-mode—SPP fields to the rough Ag-vacuum interface; the SPP thus becomes increasingly "sensitive" to the surface profile, resulting in a much increased scattering to photons. A more quantitative idea of the degree of localization may be given by es-

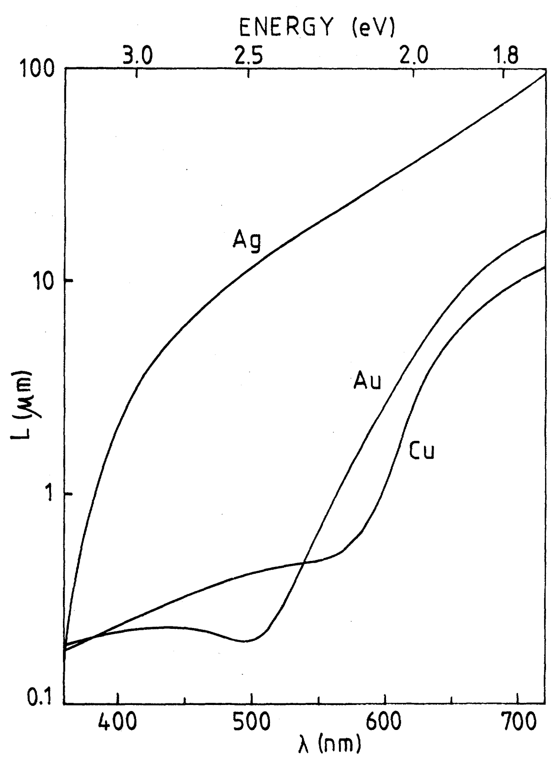

FIG. 11. Plots of L vs λ for Ag—, Au—, and Cu—liquid-nitrogen SPP modes. L is SPP propagation length.

timating $[\mathrm{Re}(K_{0z})]^{-1}$, which determines the distance above the top-electrode surface at which the SPP electric field has fallen to e^{-1} of its amplitude at the surface; $\mathrm{Re}(K_{0z})$ is given by[43]

$$\mathrm{Re}(K_{0z}) = -(\omega/c)(2Q)^{-1/2}\epsilon_0(\epsilon_r+\epsilon_0+Q^{1/2})^{1/2} , \quad (7)$$

where

$$Q=(\epsilon_r+\epsilon_0)^2+\epsilon_i^2 .$$

(The subscript z refers to the z direction, which coincides with the junction normal.) For an Ag-vacuum interface the decay distance of the SPP field, $[\mathrm{Re}(K_{0z})]^{-1}$, decreases from ~ 100 μm at 1.5 eV to only ~ 5 μm at 3.0 eV—the optical data of Ref. 34 were used for these calculations. The variation of the field-decay distance by a factor of ~ 20 across this energy range implies a variation of $\sim 4\times 10^2$ in the SPP-energy decay distance. It is this dramatic variation which underlies the remarkable range in the efficiency of the LETJ as a radiating antenna. Finally, we note that a theoretical antenna-factor curve[10] may be extracted from the rather complex expression for the radiative output given by LM (Ref. 15)—its variation with energy, though not its absolute values, agrees well with the experimentally derived curve of Kirtley *et al.*[10]

While the general increase in output from an Ag LETJ with decreasing wavelength is essentially due to the variation in SPP-photon coupling (i.e., the antenna factor), the sharp decrease in intensity from its maximum to zero at the energy given by Eq. (1) involves the tunneling-electron—SPP coupling. In a tunnel juncton the excitation of SPP's is caused by optical frequency fluctuations in the tunneling current.[5,8] Using a transfer Hamiltonian

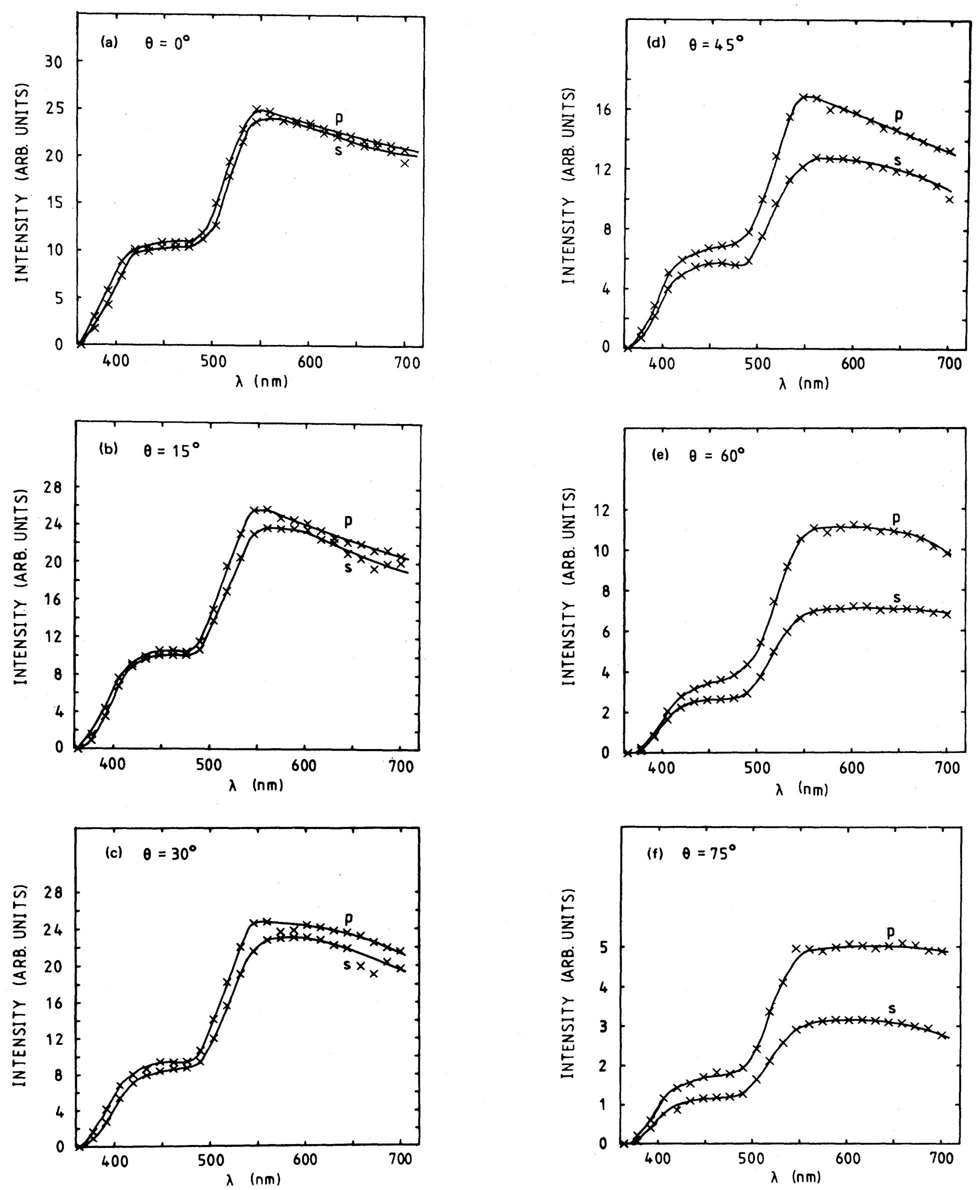

FIG. 12. Corrected spectra of s-polarized and p-polarized radiation emitted by Al-Al_2O_3-Au junction. (a) $\theta=0°$; (b) $\theta=15°$; (c) $\theta=30°$; (d) $\theta=45°$; (e) $\theta=60°$; (f) $\theta=75°$. Applied bias $V_0=3.40$ V ($i=30$ mA) throughout.

approach and assuming a free-electron model, Hone *et al.*[5] (also see Ref. 7) found a very simple expression for the power-density spectrum of the current fluctuations, namely

$$|i(\omega)|^2=\begin{cases} e\bar{i}(1-\hbar\omega/eV_0), & \hbar\omega<eV_0 \\ 0, & \hbar\omega>eV_0 \end{cases} \tag{8}$$

where $\bar{i}$ denotes the average tunneling current. [A modified form of this expression is also used by LM (Ref. 28) and AZ (Ref. 30) in their theoretical models.] Essentially, the sharp drop at the high-energy end of the spectra (Fig. 2) reflects the variation (with $\hbar\omega$) of the power-density spectrum of the current fluctuations, given by Eq. (8). Physically, the reason for such a variation is the much greater probability of a hot tunneling electron undergoing transitions (including those resulting in SPP excitation) involving a small energy change than transitions involving a large energy ($\sim eV_0$) change.

To summarize, the simple shape of the spectrum from statistically rough Ag LETJ's may be understood in terms of two competing factors. The sharp drop from the peak intensity to zero intensity (at $\hbar\omega_{\max}$) is determined by the power-density spectrum of the current fluctuations. Over the remainder of the spectral range, however, the dominant influence is that of the "antenna factor." It is this factor that causes the spectrum of Ag LETJ's to peak at the high-energy end and it is also the one responsible for boosting the blue end of the spectra in Au and Cu LETJ's to greater intensities than what would be expected on the basis of proportionality to the SPP propagation length. We might also remark that there may be other lesser influences on the form of the spectral output, notably the scattering of hot tunneling electrons in the conduction band of the insulator[20] and the form of the surface-roughness spectrum.[28,30]

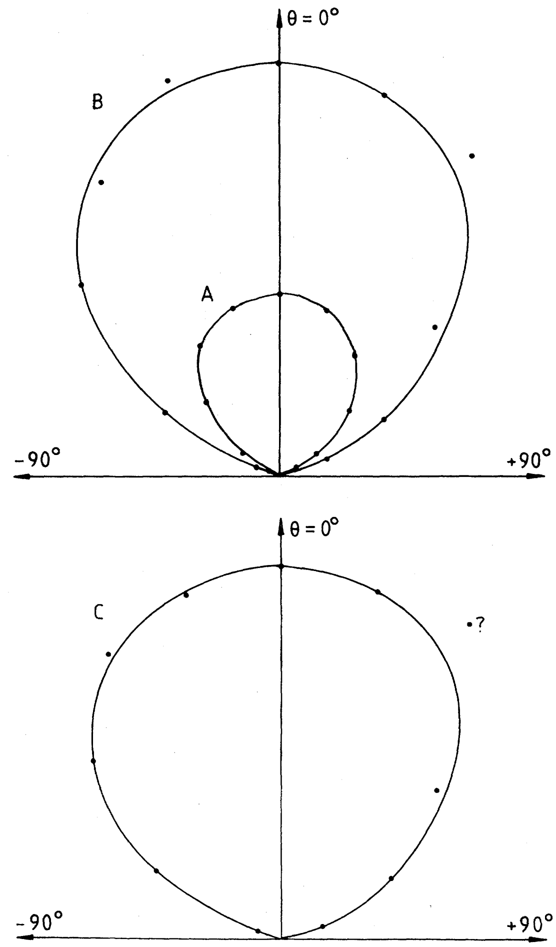

FIG. 13. Angular distribution of total (i.e., s- and p-polarized components) output from $Al\text{-}Al_2O_3\text{-}Au$ junction. A, $\lambda=450$ nm; B, $\lambda=550$ nm; C, $\lambda=650$ nm. Based on corrected spectra of Fig. 12.

IV. FURTHER EVIDENCE FAVORING FAST-SPP MEDIATION

A. Polarization and angular distribution of light emitted by Au LETJ's

The polarization and angular distribution of the light emitted by Au LETJ's should form a critical testing ground for both the theoretical models of LM (Ref. 28) and AZ (Ref. 30). Previously, what little attention has been given to polarization and angular distribution has been concentrated on the Ag LETJ, where it is reported that the light is essentially unpolarized[16,19] and has an approximate $\cos\theta$ angular distribution.[22] Parvin and Parker[22] report that the emission from Au LETJ's also is most intense along the direction normal to the device. Here, Figs. 12–15 offer a much more comprehensive description of the emission from Au LETJ's.

The corrected spectra of the s- and p-polarized components of the output from an Au LETJ are shown in Figs. 12(a)–12(f) for various angles of emission θ. The slight excess of p-polarized light in the $\theta=0°$ direction is due to the fact that light emitted within a small angular range ($\pm 7°$ from the junction normal) is actually being examined. It is convenient to divide the output from Au LETJ's into two parts: an unpolarized component which accounts for the bulk of the output, and a small, excess p-polarized component (i.e., p-s intensity). The angular distribution, at various wavelengths, of the former component is shown in Fig. 13, while that of the latter component is shown, again for the same selected wavelengths, in Fig. 14. The form of these angular distributions was confirmed by the use of optical interference filters at $\lambda=401$, 455, and 554 nm. Finally, Fig. 15 presents spectra of the excess p-polarized light for angles of emission $\theta=30°$, 45°, 60° and 75°. The margin of error in determining similar spectra for smaller emission angles is large enough to make the shape of such spectra uncertain.

The results of Figs. 12–15 seriously disagree in at least one respect with each of the theoretical descriptions of LM (Ref. 28) and AZ (Ref. 30). For example, Fig. 12 shows no evidence of the very substantial excess p-polarized output (due to multiple scattering of SPP's) described by AZ. Nor do we find the entire higher-energy (>2.2 eV) light output to be p polarized and to display a dipole-radiation-like angular distribution, features which LM predicted for the directly emitted radiation. On the

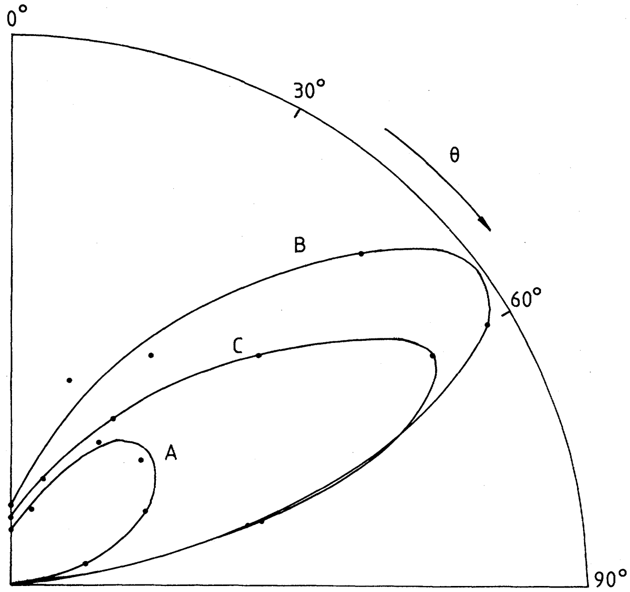

FIG. 14. Angular distribution of excess p-polarized (i.e., p- minus s-polarized component) light emitted by Al-Al_2O_3-Au junction. A, $\lambda=450$ nm; B, $\lambda=550$ nm; C, $\lambda=650$ nm. Based on Fig. 12.

contrary, the uniform structure of the emitted radiation (in terms of the ratio of the p- to s-polarized intensities) throughout the entire spectral range is a strong indication that one single emission mechanism operates. From the discussion of Sec. III it is clear that, in Au LETJ's, the play plausible mechanism is the excitation and subsequent radiative decay of the *fast*-SPP mode; it alone can give rise to radiation over a broad spectral range, including the crucial region above 2.2 eV. (Fortunately, the results on angular distribution are consistent with LM's description[28] of SPP-mediated radiation.) There would thus seem to be no need to appeal to a direct-emission process. Further weight is added to this point of view when it is considered that Parvin and Parker[22] found no evidence of

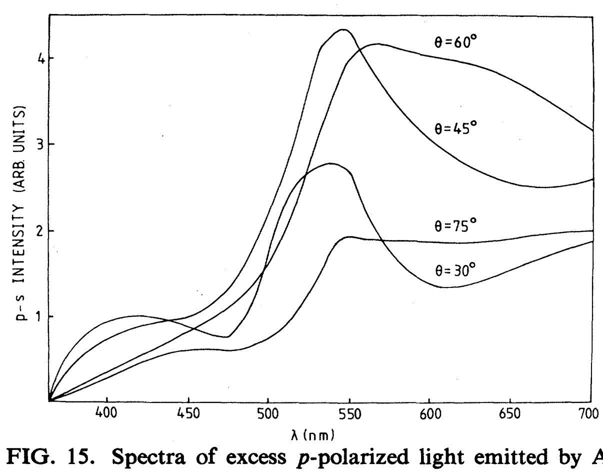

FIG. 15. Spectra of excess p-polarized light emitted by Al-Al_2O_3-Au junction at various angles of emission θ. Based on Fig. 12.

directly emitted radiation from Ag LETJ's.

What then of the small, excess p-polarized component of the light output described in Figs. 14 and 15? Could it be due to the direct-emission process? An examination of the spectrum of this component (Fig. 15) would seem to rule out any such possibility. The calculated spectrum of the directly emitted radiation peaks sharply in the very region ($\sim$2.5 eV, or 500 nm) in which the intensity of the excess p-polarized component is falling sharply. In fact, the general similarity of the spectra of Fig. 15 to those of the total emission (Fig. 3) strongly suggests the involvement of SPP's. On one hand it may be argued that the excess p-polarized emission is simply due to the roughness-induced scattering of the fast-SPP mode. This component of the radiation is thus indistinguishable in origin from the unpolarized bulk of the output and its isolation is simply an artifact of the manner in which we have chosen to treat our results. The calculation by AZ (Ref. 30) concerning radiation due to the scattering of the slow-SPP mode shows an "excess" of the p-polarized component; presumably, the scattering of the fast-SPP mode would likewise give rise to p-polarized radiation that is more intense than the s-polarized component. However, assuming that the AZ predictions do carry over to fast-mode scattering, it appears that the perturbation (i.e., LM-type) calculations, and *not* the more rigorous calculations which take multiple scatterings into account, yield p- and s-polarized light intensities that are more in accord with those recorded experimentally. Hence it is not entirely clear that the observed small excess p-polarized component should be attributed to the scattering of the fast-SPP mode.

Alternatively, we may follow Adams *et al.*[3] (as we have in the treatment of our results on the polarization and angular distribution) and postulate that the excess p-polarized emission from statistically rough Au LETJ's arises from the radiative decay of localized SPP modes. We may then offer the following overview of our results and those[3,4] which relate to LETJ's incorporating small metal particles in their structure. These junctions are fabricated in a manner which boosts the intensity of the excess p-polarized emission originating with the localized plasmon modes of the metal particles; its intensity becomes comparable with and even outweighs that of the unpolarized emission arising from the radiative decay of nonlocalized SPP modes in the outermost metal film which electrically connects the metal-particle resonators.[4] We may suggest that the reverse is true of our structures. The bulk of the emission originates with the nonlocalized fast-SPP mode, with only a small contribution to the total light output coming from localized SPP modes. One difficulty with this interpretation is that the localized mediation of light above 2.5 eV from Au LETJ's would imply[5–7] modes localized on the scale of 10 nm or less. If the physical structure of the thin Au film was such that plasmon modes localized on this scale could be supported, then it might be argued that the radiative decay of the slow-SPP mode should also proceed. However, we find no support elsewhere in the results for the participation of the slow-SPP mode and thus, by implication, no evidence for surface structure on the scale of 10 nm or less.

In concluding this section of the discussion it may be said that the results on the angular distribution and polarization of the light emitted by Au LETJ's lend further support to the idea of fast-SPP mediation throughout the spectral range. However, further experiments involving a variation in the detailed morphology of the Au electrode would be needed in order to determine the origin of the small, excess p-polarized component.

B. Emission under reverse bias

The paucity of experimental data concerning the emission of light from reverse-biased tunnel junctions is rather surprising in view of the general tenor of the article by Davis.[27] Apart from several comments in the early articles of Lambe and McCarthy,[1,16–18] only Jain *et al.*[19] offer a description of such emission—they report that for Mg-I-Ag (where I is an insulator) junctions the spectral shape for both bias polarities is similar, but with a reduced emission intensity per unit tunnel current for the reverse-bias polarity. Here we confirm this observation for the case of the Al-I-Ag junction (the structure actually studied by Davis) in Fig. 16, which presents both the forward- and rear-emission spectra for the reverse-bias polarity (Al electrode positive). With reference to Fig. 16 it was thought that a comparison of the light outputs was more meaningful for the case where the forward and reverse currents, rather than applied biases, were of equal magnitude; moreover, the LETJ's generally proved to be rather unstable under a reverse bias of 2 V or more.

According to Davis,[27] the excitation of the slow-SPP mode (up to an energy of ~ 3 eV) should proceed symmetrically with respect to the sense of the applied bias. It follows that the intensity of the emitted light should likewise be insensitive to the bias polarity. These predictions are in disagreement with the experimental results of Fig. 16, which clearly demonstrates a considerable degree of asymmetry in the light emission. The asymmetry in the tunneling current, which is, in turn, due to an asymmetric barrier potential, is also evident from Fig. 16. This source of asymmetry should, for any given energy below that corresponding to the applied bias, favor the reverse-bias emission. However, even with the tunneling-current asymmetry working to its advantage, the total light output under the reverse-bias condition is about 4 times weaker than that under the forward-bias condition.

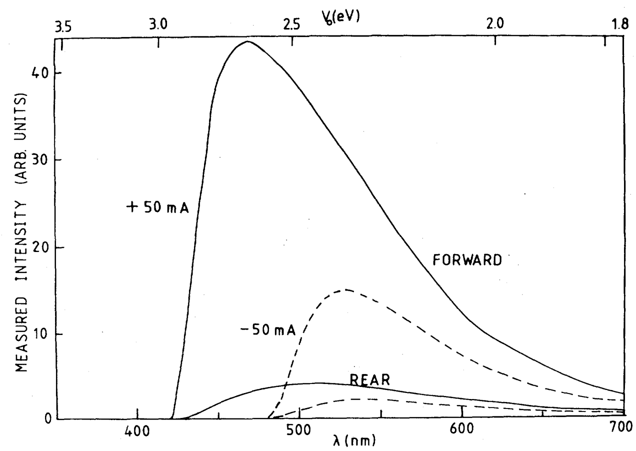

FIG. 16. Solid lines: Recorded (uncorrected) spectra of light emitted in forward ($\theta=0°$) and rear ($\theta=180°$) directions from Al-Al_2O_3-Ag junction *forward* biased with a current of 50 mA. Dashed lines: Recorded (uncorrected) spectra of light emitted in forward ($\theta=0°$) and rear ($\theta=180°$) directions from same junction *reverse* biased with current of 50 mA.

What conclusions are to be drawn? It could be construed, for example, that the identification of the SPP excitation as a truly inelastic process is mistaken. Davis himself argues in the introduction to his article that an injection mechanism would be expected to lead to a large degree of asymmetry. Laying aside such considerations, however, we could have here further evidence which mitigates against the role of the junction SPP in the light-emission process. If, as has been proposed, the light emission is dominantly due to the decay of the fast-SPP mode at the Ag-air interface, then the asymmetry of light emission with respect to bias direction may be readily understood in a qualitative manner. If electrons tunnel in the direction from Al to Ag, then the fast-SPP mode is excited in a relatively efficient manner. By contrast, the excitation of this mode would proceed somewhat less efficiently if the electrons tunnel in the opposite direction. The possibility of light emission mediated by the junction SPP should perhaps not be excluded altogether. The excitation efficiency of this mode, relative to that of the fast mode, may be even greater in a reverse-biased junction than theoretical calculations indicate for a forward-biased junction. Nonetheless, there still remains the highly debatable point of whether there is roughness on a sufficiently small scale to couple the junction SPP to radiation (see Sec. V A).

To summarize, the results on reverse-bias emission possibly constitute further evidence for the dominance of the fast-SPP mode in the light-emission process, but certainly do nothing to enhance the status of the slow or junction mode to that of a serious contributor.

C. Consistency of the fast-SPP-mediation model with other experimental results

We consider the fast-SPP-mediation model to be consistent with a number of experimental results beyond the scope of those presented in this paper; we highlight two in particular.

The first concerns the early work of Lambe and McCarthy who observed light emission from statistically rough Au LETJ's at energies up to 4 eV.[16] It was the emission of photons above 2.2 eV (the slow-mode cutoff energy) from such tunnel junctions which expressly motivated LM (Ref. 28) to develop a comprehensive description of the direct-emission process. This direct process should operate in tunnel junctions having perfectly smooth films. Yet McCarthy and Lambe found[1] negligible emission from a nominally smooth Au LETJ above ~ 2.3 eV. The significant emission of higher-energy (>2.3 eV) photons occurred only upon the introduction of surface roughness (or small-metal-particle resonators) to such a structure.[1] Since the enhancement of the light output upon the introduction of small-scale surface roughness was one of the key factors used to adduce the involvement of SPP's in the emission process, it must again be concluded that the radiation at all energies, and especially that at the blue end of the visible spectrum in Au LETJ's, is SPP mediated.

A second, salient experimental observation is that by Parvin and Parker[22] concerning the emission from Ag junctions; they report that it extends only to 3.5 eV even when the applied bias exceeds 4.0 eV. (As yet, our measurements do not extend into the uv region). Continuing in the vein of Sec. III E, we note that the rigorously calculated asymptotic energy for the slow mode in the Ag LETJ is ~ 3.25 eV (Refs. 28 and 30) [Eq. (5) also yields a value of ~ 3.25 eV], whereas that of the fast mode lies at ~ 3.5 eV.[30] Hence we consider the observation of Parvin and Parker, viz., compliance with the quantum cutoff condition [Eq. (1)] up to ~ 3.5 eV, to be harmonious with the idea of fast-mode mediation, but quite inconsistent with slow-mode mediation. In addition, the absence of spectral structure at ~ 3.25 eV strongly mitigates against the idea of radiation originating with the slow mode.

Incidentally, Parvin and Parker also observed an enhanced emission from Ag LETJ's in the near-ir range; they attributed this feature, in accordance with the LM view, to slow SPP-photon decay assisted by surface roughness on the scale of ~ 50 nm. However, on this point we prefer to follow the localized-plasmon interpretation of Kroó *et al.*,[20] who observed the feature only in the case of junctions having a very thin ($\lesssim 20$ nm), granulated Ag film. It is noteworthy that Parvin and Parker completed their junctions with a slowly evaporated, very thin (~ 20 nm) Ag film.

V. FURTHER DISCUSSION

A. Scale of the surface roughness

The condition for efficient SPP-photon coupling may be crudely expressed[8] as $K_r\sigma \approx 1$ (K_r is the real part of the SPP wave vector and σ is the transverse correlation length for the rough surface). If the slow mode is held to be responsible for the visible output from tunnel junctions, then there is necessarily an appeal to very-small-scale surface roughness:[8,28,30] K_r for the slow mode runs rapidly to very high values. For example, AZ (Ref. 30) shows that the slow-mode emission from Ag LETJ's drops very sharply at 2.5 eV for $\sigma = 20$ nm and at lower energies for larger values of σ. In their original article[28] LM proposed a new model of CaF_2-roughened surfaces. They suggested that, superimposed on a larger scale of ($\sigma = 40$ nm) roughness,[44] there were steps and terraces on a much smaller scale ($\sigma \approx 3$–5 nm).

The negligible observed intensity of slow-mode-mediated radiation (Sec. III E) leads us to the counterpostulate that surface roughness with $\sigma \approx 3$–5 nm is substantially absent in CaF_2-roughened LETJ's. However, it is well known that the fast-SPP mode can couple to visible radiation via CaF_2-induced surface roughness,[45,46] which is definitely known to have $\sigma \approx 50$–100 nm;[47–49] indeed, some of the methods employed in the determination of transverse correlation lengths depend on this very interaction. Furthermore, recent measurements[50,51] using an attenuated-total-reflection (ATR) technique yield very high efficiency values ($>10\%$) for (fast-) SPP-photon coupling. Although such results provide independent evidence which tends to favor the participation of the fast-SPP mode in the light-emission process of statistically rough tunnel junctions, they also raise a couple of paradoxical points. First, $K_r\sigma$ is of the order of 10^{-1} for CaF_2-roughened surfaces. Clearly, the surface roughness is not ideally matched to fast-SPP scattering. However, the idea of a "mismatched" surface roughness and the efficient radiative decay of SPP's are perhaps compatible if the decay is visualized to proceed by a multpile-scattering process involving two or more surface-roughness vectors, each of which is not individually matched to the task in hand. A second point of difficulty is how to reconcile the very high SPP-photon decay efficiencies[50] ($\sim 80\%$) at an energy of 1.96 eV on Ag-grating surfaces with the antenna-factor curve[10] of Kirtley *et al.*; this curve indicates that in going from 2.0 to, say, 2.5 eV, the efficiency of the grating as a radiating antenna should increase by about an order of magnitude. We do not propose to pursue this matter any further here.

In concluding subsection A, we suggest that there may be two possible ways to increase the efficiency of the statistically rough LETJ. The first is to fabricate the device on a rough surface with $\sigma \sim 500$ nm; this should facilitate a more efficient radiative decoupling of the fast-SPP mode. (However, ATR investigations would indicate that the scope for improvement may be rather limited.) The second method is the introduction of surface roughness with $\sigma \sim 5$ nm, thus facilitating the decay to visible photons of the efficiently excited slow-SPP mode. LM have already suggested[29] that the absorption of large molecules (ranging from 5 to 10 nm in size) on the outer surface of the LETJ could increase the emission intensity. In the view presented here the absence of such small-scale surface roughness effectively closes the radiative decay channel for slow-mode SPP's. If this view is correct, then the motivation for introducing surface roughness with $\sigma \sim 5$ nm (or localized scattering centers on the scale of 5 nm) is considerably increased.

B. Statistically rough LETJ in relatoin to other work

By way of a general remark, we note that the considerable attention paid to SPP dispersion contrasts sharply with the relative neglect of SPP damping. The interest in dispersion seems to go hand in hand with the use of grating surfaces[45] which yield detailed information (the position of the sharp photon-emission peaks or absorption dips, depending on the particular experimental arrangement) directly bearing on SPP dispersion. Furthermore, it is often argued that the use of statistically rough metal surfaces blurs the available spectroscopic information. Kirtley *et al.* took this view and were thus motivated to investigate LETJ's grown on grating substrates,[9–11] as were Kroó *et al.* in their independent study.[12–14] However, such an approach does not have a monopoly on advantage because, as we have shown here, statistically rough

Au and Cu LETJ's allow for a rather valuable scan of SPP-damping properties and thus of the dielectric properties of the metal forming the top electrode. Certainly, with a grating LETJ, information on SPP damping is not lost since it contributes to the half-width of the emission peaks; obviously, though, the nature of the variation in SPP damping across the spectral range is less accessible. Thus, while it is true that information on SPP dispersion is blurred by the use of statistically rough LETJ's, it is equally true that information on SPP damping may be obscured by the use of grating LETJ's. Indeed, the statistically rough LETJ perhaps represents the first experimental arrangement which is capable of scanning visible-regime SPP damping properties in such a direct manner.

This insight on LETJ operation allows us to view the device as a potentially useful spectroscopic tool. It may, in fact, be regarded as a broadband, visible-regime extension of SEW spectroscopy.[42] In SEW spectroscopy the propagation length L of ir-region SPP's (i.e., SEW's) may be determined as a function of energy;[52,53] thin films of dielectric media or adsorbed molecules overlying the base metal cause a reduction in L at the eigenfrequencies of the film vibrations.[54] Similarly, we might anticipate that thin-film overlayers on LETJ's would cause variations in the fast-SPP propagation length, which would, in turn, be monitored by the light output. Currently, we know that the output from Au and Cu LETJ's monitors the variation in L for the fast-SPP mode due to the influence of an interband transition in the top metal electrode (Au or Cu) itself.

The radiation SPP-coupling techniques[42,52,55,56] used at the input and output stages in SEW experiments require the SPP to have a macroscopic (>1 mm) propagation length. These techniques preclude the extension of SEW spectroscopy into the near-ir and visible regions where the SPP propagation length is on a microscopic scale (see Fig. 11). Thus, the great advantage of the statistically rough LETJ is its potential to detect variation in the microscopic propagation length of SPP's at visible energies. The principal disadvantages of the LETJ are its low-intensity output and the fact that there is no simple, direct relation between the output intensity and the absolute value of L; neither of these difficulties is encountered in SEW spectroscopy. In particular, the "antenna factor" discussed in Sec. III F tends to mask the magnitude of the variation in L. On the other hand, the antenna factor endows the output with a strong blue bias, thus enabling the LETJ to probe SPP behavior sensitively in the rather esoteric, but physically interesting, region above the interband transition in Au and Cu. We know of no other experimental arrangement which makes SPP excitations in this energy regime accessible to investigation.

The work on statistically rough LETJ's is also of possible relevance to surface-enhanced Raman spectroscopy (SERS) where it is thought that SPP's are excited.[57] Clearly, if the long-range part of the enhancement mechanism involves SPP excitation, then SPP damping will influence the magnitude of the enhancement. For example, we expect the effect to be much reduced (by a factor $\sim 10^2$) when Au or Cu substrates are used in conjunction with laser excitation in the blue or green. Experiment confirms this expectation.[58–62] A more quantitative expression of this physical interpretation is found in the close similarity in form between the SPP propagation-length plots of Fig. 11 and Schatz's graphs[63] of the SERS-enhancement factor versus energy for pyridine adsorbed on Ag, Au, and Cu. The form of each of Schatz's calculated graphs, and thus that of the corresponding graph of Fig. 11, is in harmony with the available experimental data.

VI. CONCLUSIONS

The striking correlation between the forward-emission spectra of statistically rough Al-Al_2O_3-M (M=Au, Cu) tunnel junctions and the wavelength dependence of the fast-SPP damping constitutes strong evidence for the dominance of the fast-SPP mode in mediating the radiative output. The idea of fast-SPP mediation has been shown to be in harmony with the remainder of the experimental data presented here, in Sec. IV, and with those of other workers. In particular, the results relating to the polarization and angular distribution of the output from Au LETJ's point to a common source of emission throughout the visible-energy regime and that source can only be the fast-SPP mode. In parallel, a number of areas of disagreement between the experimental results and the predictions of extant theoretical models have been highlighted. To summarize, we find no evidence for the direct-emission process originally proposed by LM,[28] nor for a significant contribution by the slow-SPP mode which, in both the models of LM and AZ,[30] accounts for the bulk of the visible output. Also, we observed only a *small* excess p-polarized component (described in Figs. 14 and 15), rather than the very considerable excess p-polarized output which AZ find to result from multiple scattering of the SPP's mediating the emission.

Further interesting features of the output from the statistically rough LETJ are the spectra of the emission to the rear of the device (Figs. 5 and 16) and the spectra of the emission for the reverse-bias condition (Fig. 16). The low intensity of the former spectra was important to the formulation of some initial arguments (Sec. III B and Ref. 25) which eventually led towards the idea of fast-SPP mediation, an idea which finds further possible support in the low intensity of the latter spectra.

The view presented here of the operation of the statistically rough LETJ implies that a change in the scale of the surface roughness from that employed at present should increase the emission efficiency (Sec. V A). A much more efficient LETJ would be commercially attractive on account of its ease of fabrication and the rather convenient, voltage-tunable nature of the emission color.

Viewing the LETJ as a device capable of scanning SPP-damping properties (Figs. 8 and 9) opens up further avenues of investigation. It would seem, for example, to be particularly useful for probing SPP behavior above the interband transitions in Au and Cu. We have also argued that the emission from the statistically rough LETJ may be regarded as an extension of SEW spectroscopy into the visible spectral range and we suggest that the understanding of the device presented here may be of relevance to SERS investigations.

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