Scanning tunneling microscopy and optical spectrum studies of light-emitting tunnel junctions

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We have measured the optical spectra of light-emitting tunnel junctions with two different roughnesses and measured the power spectrum of the roughness with scanning tunneling microscopy (STM). Our experiments show that most of the information on the roughness relevant to light emission can be inferred from STM measurements. We discuss the aspects of the STM data that are unique to the determination of long-wavelength roughness relevant to optical studies.

The earliest study[1] of light emission from metal–insulator–metal tunnel junctions shows that surface roughness of the films is responsible for the emission of light. Subsequent theoretical work[2] provides the following description of the emission process. Although nominally biased with a dc tunneling current, there are current fluctuations that extend to a frequency of $\omega = eV/\hbar$, where V is the dc bias voltage and e and $\hbar$ have their usual meaning. The current fluctuations have a broad distribution of Fourier components in $k_{\parallel}$, the wave vector parallel to the films. The current fluctuations excite electric fields in the junction. An alternative mechanism for the excitation of the electric fields has been suggested by Kirtley *et al.*[3] who attribute the excitation to hot electrons. There are resonances in the electric field strengths for particular values of $k_{\parallel}$ and ω. The resonant values of $k_{\parallel}$ and ω lie along trajectories in the $(k_{\parallel},\omega)$ plane that are called surface plasmon dispersion curves. The dispersion curves and the strengths of the resonances along them are determined by the dielectric functions and thicknesses of the films in the junction. Thus the current fluctuations or hot electrons create a broad spectrum in both ω and $k_{\parallel}$ of electric field strengths at the surface of the junction.

For the hypothetical case of perfectly flat junctions, not all of the Fourier components of the electric field at the surface can radiate light. Light emitted from the structure can be thought of as a plane wave traveling away from the junction with frequency ω and wave vector $k = \omega/c$ at an angle θ to the junction normal. The fields at the surface of the flat junction must obey the usual electromagnetic boundary conditions. These require, among other things, that the parallel components of the wave vectors of the fields on both sides of the surface be equal. Thus the electric field just inside the surface of the junction with frequency ω and wave vector $k_{\parallel}$ can only couple to a plane wave above the junction if $k_{\parallel} = (\omega/c)\sin\theta$ for some value of θ between 0° and 90°. Otherwise the field above the junction will die exponentially and will not represent the emission of light. Interestingly none of the resonantly enhanced fields along the surface plasmon dispersion curves can radiate in flat junctions because $k_{\parallel}(\omega)$ exceeds ω/c along all of the surface plasmon dispersion curves. This makes them true surface excitations.

The presence of surface roughness relaxes this boundary condition and allows the surface plasmons to radiate. The theory[2] shows that the power spectrum of the surface roughness determines the rate at which the surface plasmons radiate. Specifically if $\zeta(k_{\parallel})$ is the Fourier transform of the surface roughness profile, the power spectrum $P(k_{\parallel})$ is $|\zeta(k_{\parallel})|^2$. The coupling of the field inside the junction with wave vector $k_{\parallel}$ and frequency ω to light emitted at angle θ with frequency ω is proportional to $P[k_{\parallel} - (\omega/c)\sin\theta]$. Therefore, in order to study light emission from rough junctions the power spectrum of the roughness must be determined.

Figures 1 and 2 show that varying the surface roughness produces large changes in the emitted optical spectrum. Figure 1 shows the optical spectrum emitted from the gold side normal ($\theta = 0$) to a Au–insulator–Al junction evaporated on a glass substrate. The film thicknesses are shown on the inset in the figure. The junctions are biased at 2.8 V. The quantity plotted on the vertical axis is proportional to the number of photons per second per unit tunneling current per unit energy range emitted into a small angular range about $\theta = 0$. The proportionality constant has not been determined but it is the same for the data presented in Figs. 1 and 2, so they may be directly compared. For the sample shown in Fig. 1 no steps have been taken to roughen the junction deliberately. The nucleation and growth of the films guarantee some roughness anyway and this roughness is responsible for the emitted light. Nevertheless, we will refer to this as the smooth junction.

Figure 2 shows the optical spectrum at $\theta = 0$ of a similar junction deposited on a glass substrate that had 120 nm of CaF_2 preevaporated on it. This spectrum is similar to the spectra on similar samples reported by Dawson and coworkers.[4] The CaF_2 film roughens the entire junction structure and produces striking changes in the emitted optical spectrum. First of all the junction in Fig. 2, which we shall call the rough junction, is a factor of 60 times brighter per unit tunneling current than is the junction of Fig. 1. Secondly, the peak in the rough junction optical spectrum is near 1.8 eV whereas the emission peaks in the smooth junction spectrum lie near 1.9 and 2.1 eV. Finally, there is a "knee" in the rough junction spectrum near 2.4 eV that is absent in the smooth spectrum. The energy dependence of the optical spectra of both kinds of junctions are satisfyingly reproducible from sample to sample. For both types of junctions the relative intensities agree to within a few percent for all frequencies. The maximum normalized intensities show more

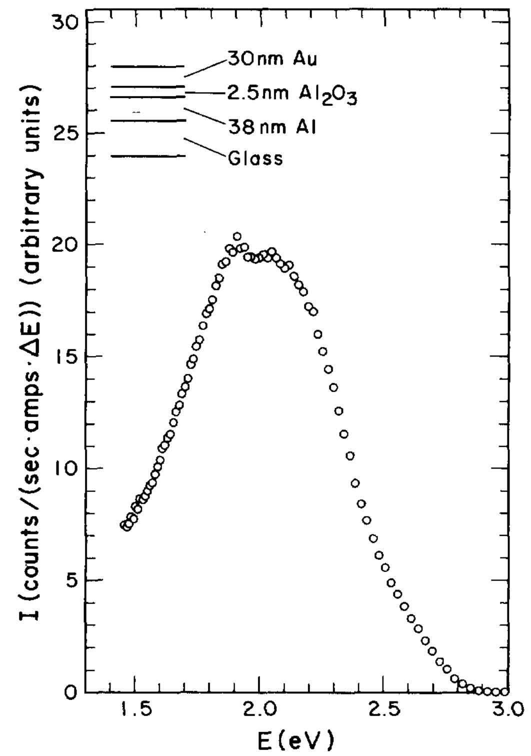

FIG. 1. Normalized photon flux vs photon energy for a "smooth" light-emitting tunnel junction. The junction geometry is shown in the inset.

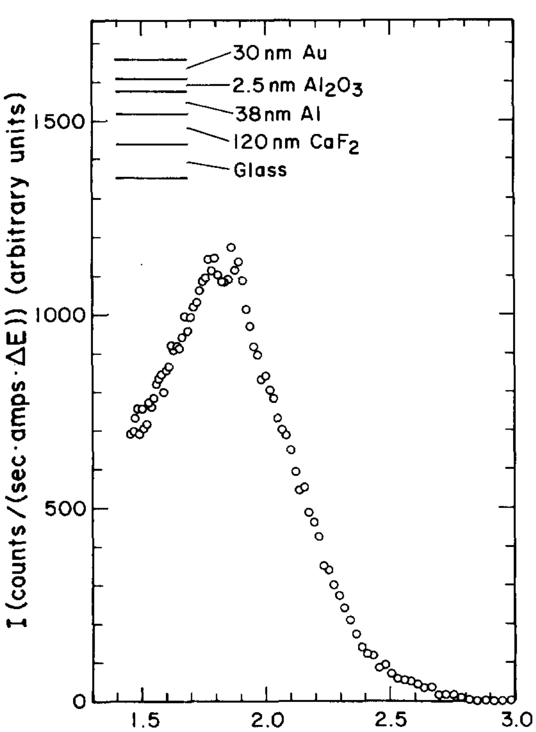

FIG. 2. Normalized photon flux vs photon energy for a CaF_2 roughened light emitting junction. The intensity units are the same as those for Fig. 1 so that the ratios of the intensities are the ratios of the ordinates in these two figures. Although the normalized intensity of this junction is a factor of 60 larger than the junction in Fig. 1 it was actually a factor of 20 less bright because it drew less current. The junction geometry is shown in the inset.

variation. For example, we have measured four samples like the one in the inset to Fig. 1. Three of them have the same normalized maximum intensity within 30% of the one shown, the other was twice as large. Within these limits, the differences between Figs. 1 and 2 represent reproducible changes in the optical spectra that are induced by changes in the roughness.

We have begun to study the roughness in smooth and rough tunnel junctions by scanning tunneling microscopy. The aluminum film thickness on the rough junctions we have analyzed with the STM are not the same as those on which we measured the optical spectrum. The aluminum films in the optical spectra of Figs. 1 and 2 were 38 nm. The smooth power spectrum was measured on a junction with the same Al film thickness, but the rough power spectrum was measured on a junction with a 75-nm-thick Al film. All Au films were 30 nm thick and the CaF_2 was 120 nm thick on all rough samples. Although we have not yet produced reliable STM and optical data on identical samples, the STM results here give some indication of the power spectra of roughness on light emitting junctions and show how the roughness is distributed on the various metal interfaces.

The tunneling microscope consists of three orthogonally mounted tubular piezoelectric elements, each capable of sweeps up to 3 μm. Calibration of the x and y sweeps was done using a gold-plated diffraction grating. The sweeps were programmed with an IBM XT computer through 12-bit digital-to-analog converters (DAC's) to Kepco programmable high-voltage power supplies. The height information was acquired from a 12-bit analog-to-digital converter (ADC) monitoring the input of the z-piezo high-voltage op-amp supply.

The tunneling tips were cut from 0.5-mm platinum wire using scissors.[5] The microscope was run with the sample grounded, and the tip at + 100 mV. The tunneling current was held constant at 1 nA with an integrating feedback loop.

The STM was used to record height versus position along 2.25-μm-long straight-line trajectories on a rough and a smooth tunnel junction. Each height record was then fast Fourier analyzed and squared to provide a power spectrum of the roughness. Sixty such power spectra were averaged to obtain each of the spectra we show here. To analyze the noise in the microscope, the drive voltage to the piezoelectric element that moved the tip along the sample was disconnected and the apparent height variations recorded.[6] These apparent variations had a root-mean-square (rms) amplitude of 0.15 nm and a white power spectrum. This background has been subtracted from the power spectra discussed here.

We generated power spectra of the roughness of the Au film in four places: on top of the smooth junction (Fig. 3), on top of a continuation of the Au film off the smooth junction area on the glass substrate (Fig. 4), on top of the rough junction (Fig. 5), and on top of a continuation of the Au film off the rough junction area on the CaF_2. We do not show the power spectrum of the Au film on the CaF_2 off the junction area because within the scatter of the data it is indistinguishable from the result on top of the junction.

A general feature is revealed when the power spectra are compared. The power spectra for $k_{\parallel}$ values larger than

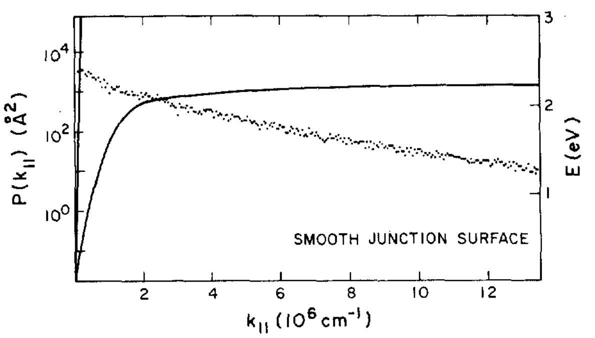

FIG. 3. Roughness power spectrum of the Au film on the smooth junction. The rms roughness is 16.5 Å. The solid lines are the dispersion curves for two of the surface plasmon modes. The energy is displayed on the right-hand ordinate.

$\sim 5\times 10^6$ cm^{-1}, corresponding to wavelengths shorter than about 12.5 nm, are identical for all four sets of data. There are two simple ways that this could come about. First, it is possible that the short-wavelength mean square roughness of the Au film evaporated on a flat surface is much larger than the short-wavelength mean square roughness of all four surfaces supporting the Au films, namely glass, CaF_2 and oxidized Al on glass or CaF_2. Thus "intrinsic" Au surface roughness would dominate the short-wavelength roughness.

It is also possible that as the Au film is deposited it first buries or snow drifts over the short-wavelength roughness of its substrate. Again if the Au is thick enough only its intrinsic short-wavelength roughness is visible on the surface. These possibilities could be sorted out by a systematic study of the Au film surface roughness as a function of the Au film thickness.

The identity of the Au film surface power spectra on and off the rough junction area has important implications for the optical emission. As we shall discuss, the important part of the roughness power spectrum for optical emission is the part with $k_{\|} \simeq 2\times 10^6$ cm^{-1} and smaller. Here, the smooth Au film power spectrum is an order of magnitude or so less than either the rough Au film or rough junction. Clearly the Au film intrinsic roughness cannot be responsible for the roughness on the CaF_2 samples. Rather it seems likely that the long-wavelength roughness of the CaF_2 surface is much larger than the intrinsic long-wavelength roughness of either the Al or Au films and that all the films in the structure approximately replicate the CaF_2 surface. Thus in this region, all four metal interfaces can be thought of as rough but locally parallel. This is sometimes referred to as correlated roughness.

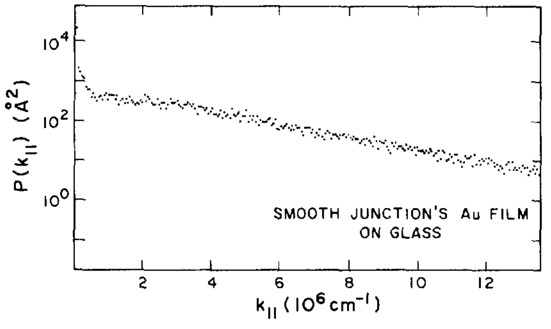

FIG. 4. Roughness power spectrum of the Au film on the glass substrate. The rms roughness is 12.6 Å.

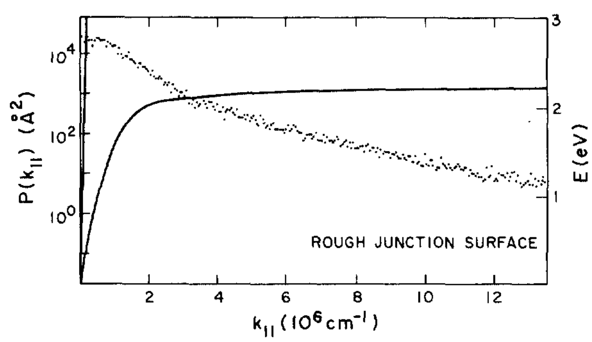

FIG. 5. Roughness power spectrum of the Au film on the rough junction. The rms roughness is 39.8 Å. The roughness power spectrum of the Au film on the CaF_2 surface is indistinguishable from this. The solid lines are the dispersion curves for two of the surface plasmon modes. The energy is displayed on the right-hand ordinate.

A comparison of the power spectra for the smooth junction and the smooth Au film indicates that the top surface of the oxide barrier is intrinsically rough. Figures 3–5 show that the long-wavelength surface roughness of the Au film over the smooth junction is an order of magnitude or so rougher than the Au film on glass and about an order of magnitude less rough than the Au film on the rough sample. This suggests that the top surface oxide barrier is about an order of magnitude rougher at long wavelength than the intrinsic roughness on a Au film and an order of magnitude less rough than the CaF_2 surface. It is not clear whether the roughness of the surface of the oxide barrier is due entirely to a rough Al surface or whether the oxidation layer is rough as well. It is unlikely that all of the excess roughness is due to uneven barrier thickness because comparison of the power spectra in Figs. 3 and 4 shows that the barrier would need to have thickness variations as large as its thickness to explain the $\sim$600–1000 Å^2 differences below $k_{\|} = 2\times 10^6$ cm^{-1}. In any case one cannot expect any of the existing theories[2,7] to account quantitatively for the light emission from junctions because the theories assume only the top surface is rough. Nevertheless we shall use the theory to draw qualitative conclusions about the optical data.

Superimposed on the power spectra in Figs. 3 and 5 are calculated dispersion curves[8] for two of the surface plasmon modes on the junction. The energy of the surface plasmon mode is displayed on the right-hand ordinate. The nearly vertical dispersion curve drawn to 1.6×10^5 cm^{-1} and 3 eV is the so-called fast surface plasmon that has its maximum electric fields on the Au–air surface. The other mode is the slow mode with maximum field intensity in the oxide barrier. There is a third mode with field maximum at the Al surface farthest from the oxide barrier which has a dispersion curve about 30% less steep than the fast mode. We have not drawn it on the figure because with the wide $k_{\|}$ scale we have used it is difficult to distinguish.

One expects the proportionality of the emitted intensity to $P[k_{\|} - (\omega/c)\sin\theta]$ to survive the generalization of the theory to more than one rough surface, although it may be necessary to sum contributions proportional to the roughness power spectrum of each rough interface when calculating the electric field.[9] In this case, we see that the power spectrum near $k_{\parallel} = 1\times10^5$ cm^{-1} will determine the contribution to the emission of 2-eV photons from the fast mode and the Al mode, whereas the contribution to 2-eV emission of the slow mode will be determined by the power spectrum near 1.8×10^6 cm^{-1}. These are the relevant wave vectors near the 2-eV emission peaks in Figs. 1 and 2.

The STM is a useful device for limiting the range of roughness parameters that must be considered when calculating the emission of light from tunnel junctions. There are limitations that prevent the STM from determining those parameters exactly. Most obviously the STM can only measure the roughness of the Au surface. The roughness of the other interfaces is inaccessible to the STM. Even if one were to succeed in measuring the roughness of the Al or oxide barrier away from the junction area, one could not be sure that the result represented the roughness in the junction. The Au deposition affects the condition of the surfaces at the barrier because even the tunneling resistance is affected by the deposition rate. Second, the power spectra of the individual traces of the Au film on glass, our smoothest film, show a great deal of variability for $k_{\parallel}$ smaller than $\sim 3\times10^6$ cm^{-1}. We are investigating the possibility that the films contain large intrinsically smooth areas with a low density of defects. If the intrinsic areas are smooth enough, the power spectrum will be strongly affected by the number and details of the defects the tip encounters on a particular sweep. If the film is intrinsically rougher than the defects they will have less severe effects on the data. Finally, the $k_{\parallel}$'s that matter to the fast and Al surface modes correspond to long wavelengths requiring long sweeps of the tip across the surface.

In summary, we have measured optical emission spectra from light-emitting tunnel junctions with different roughnesses. We have shown for the first time that the energy dependence of the spectrum is affected by the roughness. We have characterized the roughness by measuring the power spectra of the height profiles of gold films on and off the junction area using STM. The surface roughness of gold on CaF_2 has the same power spectrum as that on the rough junction suggesting that the roughness of the CaF_2 dominates the roughness of each interface in the rough tunnel junction. A quantitative comparison with theory is not attempted because we have not yet measured the optical spectra and roughness on the same junctions. Because the theory includes only one rough interface and our STM results indicate roughness on all interfaces, it is probable that the theory will need to be extended before it can explain the data.

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[1]J. Lambe and S. L. McCarthy, Phys. Rev. Lett. 37, 923 (1976).
[2]B. Laks and D. L. Mills, Phys. Rev. B 20, 4962 (1979).
[3]J. R. Kirtley, T. N. Theis, J. C. Tsang, and D. J. Di Maria, Phys. Rev. B 27, 4601 (1983).
[4]P. Dawson, D. G. Walmsley, H. A. Quinn, and A. J. L. Ferguson, Phys. Rev. B 30, 3164 (1984).
[5]We thank Professor P. K. Hansma for demonstrating this technique.
[6]We thank Dr. J. R. Matey for suggesting this measurement.
[7]K. Arya and R. Zeyher, Phys. Rev. B 28, 4080 (1983).
[8]S. Ushioda, J. E. Rutledge, and R. M. Pierce, Phys. Rev. B 34, 6804 (1986).
[9]Recent theoretical work under way in Japan confirms these statements about the generalization of the theory to multiple rough interfaces. S. Ushioda (private communication).