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Two-mode radiation from light-emitting tunnel junctions

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We have employed a prism coupler to cause two independent surface-plasmon polariton modes of a metal-insulator-metal tunnel junction to radiate. We have characterized the radiative coupling of the modes by attenuated-total-reflection spectrometry and measured the angular dependence of the emitted light. Each of the radiative modes is supported by one of the metal surfaces farthest from the tunneling barrier. A model calculation suggests that the modes are excited at the surfaces that support them.

In the paper announcing the discovery of light emission from metal-insulator-metal tunnel junctions,[1] Lambe and McCarthy identified surface plasmon polaritons (SPP's) as the coupling mechanism between tunneling electrons and the radiated light. Subsequent work has focused on identifying which of the three SPP modes a tunnel-junction structure supports in the visible[2] is responsible for the radiated light and how the SPP's are excited. Because the momentum of all three SPP modes exceeds the momentum of a photon of equal energy in vacuum or air surrounding the junction, none of the modes in perfectly flat junctions can radiate. To circumvent this restriction, some means of coupling the SPP's to the radiation field must be built into the junction structures. Unless the SPP-to-photon coupling can be quantified experimentally and compared to theoretical predictions without introducing arbitrarily adjustable parameters, the SPP-to-photon coupling mechanism can obscure important features of the emission process. An example is the difficulty of identifying the SPP mode responsible for the radiation from randomly rough junctions.[3]

Recently, we have developed a prism technique to provide the optical coupling between the SPP's and light.[4,5] In the prism technique, the junction is made very thin so that the fields of all the SPP's extend into the substrate. If the substrate has a dielectric function ϵ, the momentum of a photon in the substrate is a factor of $\sqrt{\epsilon}$ larger than the momentum of a photon of the same energy in vacuum. If ϵ is large enough, the momentum of a photon in the substrate can exceed the momentum of an SPP mode. Then photons traveling at an angle θ to the film normal can optically couple to an SPP of frequency ω and wave vector $k_{\parallel}$ if $\sin\theta = k_{\parallel}c/\sqrt{\epsilon}\omega$, where c is the speed of light.

We have modified the prism geometry to allow two SPP modes to radiate. We believe that this is the first observation of light emission from two clearly identified SPP modes.[6] We also show the results of attenuated-total-reflection (ATR) experiments on the junctions. These measurements demonstrate the accuracy with which the coupling between radiated light and fields in the junction can be calculated. As we shall discuss, the similarity between the calculation of the emitted light and the ATR calculation implies that if the spectrum of current fluctuations in the junction were perfectly known, we should expect to be able to calculate the emitted intensity as accurately as we can the ATR response of the junction. Finally, we will calculate the emitted intensity for some simple current fluctuation models and compare the results to the data.

The prism geometry used in this experiment is shown in Fig. 1. The flat face of a 1.27-cm-diam hemicylindrical prism of BK7 glass[7] is the substrate for a thin-film structure that includes the junction. The first layer is a 220-nm film of MgF_2 evaporated from a resistively heated alumina covered boat in a vacuum of about 4×10^{-4} Pa. The junction is formed on top of the MgF_2 layer. An 18-nm-thick film of aluminum is evaporated along the axis of the prism. The sample is then removed from the evaporator and baked in room air at 300 °C for 5 min to form the

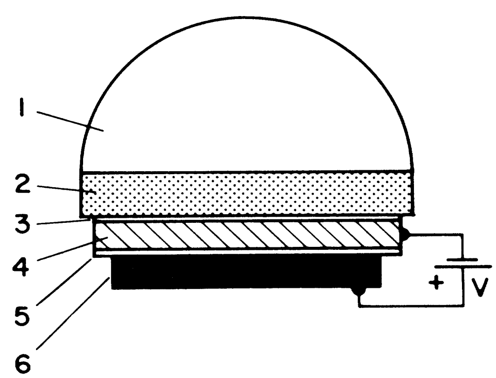

FIG. 1. Schematic drawings of the modified prism geometry. The film thicknesses are exaggerated for clarity, and the actual junction covers only a 2-mm-wide region in the center of the 6.4-mm-wide prism. The layers are 1, BK7 hemicylindrical glass prism; 2, 220-nm MgF_2 film; 3, 1-nm cermet layer; 4, 18-nm aluminum film; 5, tunnel barrier; and 6, 33-nm gold film. This geometry allows the Au-vacuum (fast mode) SPP and Al-MgF_2 (intermediate mode) SPP to radiate into the prism.

aluminum oxide tunneling barrier. The junction is completed by evaporating a 33-nm gold cross stripe over the oxidized aluminum. This process forms a $2\times2\text{-mm}^2$ tunnel junction on the axis of the prism and separated from the glass by the MgF_2 film.

Figure 2(a) shows the relative intensity of the *p*-polarized light emitted in a 1-nm bandwidth at 590 nm as a function of angle from a junction like that in Fig. 1 biased at 2.7 V. A smooth[8] background due to scattering off the small intrinsic roughness in the junction has been subtracted from the data to produce the open circles in the figure. The peaks near 43° and 68° are due to surface plasmons. When the optical-coupling condition is satisfied the fields in the junction and in the prism are enhanced and the emitted intensity goes through a peak. Calculations of the SPP dispersion relations and electric-field patterns like those in Ref. 5 allow us to identify the SPP's responsible for the two peaks in the data. The peak at 43° is due to the SPP mode with maximum field strength at the Au-air interface. This SPP is called the fast mode and has been identified in every emission experiment that can distinguish among the modes.[2,4,6,9] The peak near 68° has its maximum field strength at the Al-MgF_2 interface. Emission from this mode has not been reported before. It radiates in this geometry because its velocity is determined primarily by the dielectric function of the MgF_2. Because the dielectric function of the MgF_2 is smaller than that of the prism, the wave vector of this mode is smaller than the wave vector of light in the substrate, and thus it will radiate at a particular angle. In this case, that angle is 68°. We will call this mode the intermediate mode because its velocity lies between the velocities of the fast mode and the slow mode which has maximum field strength in the oxide barrier. The slow mode is about an order of magnitude too slow to radiate in this geometry.[2,5]

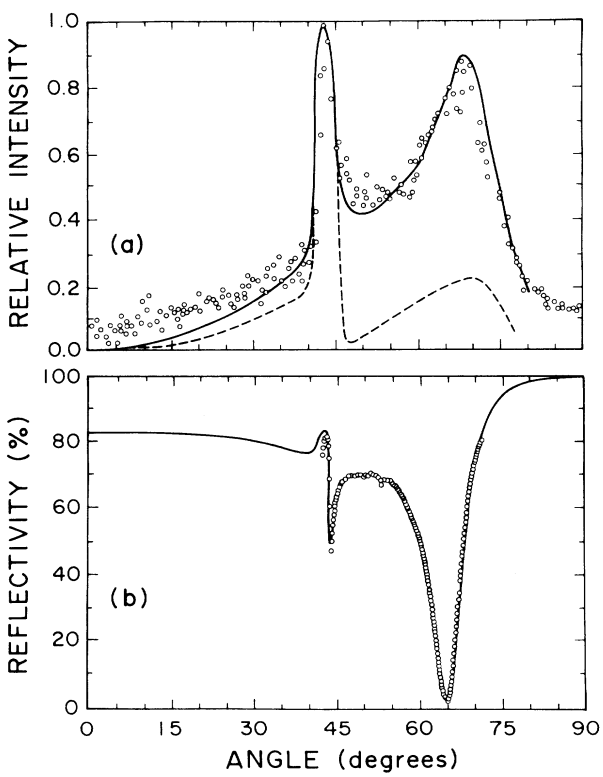

FIG. 2. The open circles in (a) show the angular dependence of 590-nm light emitted from a prism-coupled junction. The dashed curve is a calculation which assumes current fluctuations in the barrier are solely responsible for the emission. The solid curve is a similar calculation with current fluctuations at both the Au-air and Al-MgF_2 interfaces. The open circles in (b) show the ATR response. The solid line is a calculation of the ATR response described in the text.

To quantify the coupling between radiation in the prism and the fields in the junction the bias voltage was removed and ATR measurements were made on the junction. An ATR experiment[10,11] measures the angular dependence of the intensity of specularly reflected light from the junction when it is illuminated through the prism with monochromatic, *p*-polarized light. ATR is sensitive to the dielectric functions and thicknesses of the layers of the structure studied. The open circles in Fig. 2(b) show the results of an ATR measurement performed at 585 nm on the light-emitting junction. The ATR data shows the presence of the SPP modes because the enhancement of the fields when optical coupling occurs increases the resistive losses in the films and causes the reflected intensity to drop.

The solid line in Fig. 2(b) is a calculation of the ATR response of the junction. In the calculation, we used the thicknesses of the MgF_2 layer, the Al film, and the Au film measured by a quartz thickness monitor while the films were deposited. We determined the dielectric functions of the Al and Au films in a separate ATR measurement on single films of each. The results of these measurements were within the scatter of the values compiled by Ordal *et al.*[12] for these quantities. We use 1.88 for the dielectric function of MgF_2.[13] We assumed that the oxide barrier consisted of γ alumina with a dielectric function of 2.79 (Ref. 14) and a thickness of 3 nm. The calculation was performed by assuming that the electric field in each layer is the superposition of one plane wave propagating toward and another plane wave propagating away from the surface of the prism. Each plane wave has the same frequency and wave-vector component parallel to the film surfaces as the incident light. The wave-vector component perpendicular to the films depends on the dielectric function of each layer and is in general complex. To complete the calculation one sets the electric-field incident on the structure from the air side to zero and uses the usual boundary conditions to calculate the amplitudes of the fields in the succeeding layers until the amplitudes of the incident and reflected fields in the prism are calculated. To produce the result shown in the figure, we found it necessary to add a cermet layer between the Al and MgF_2 layers.[15] Without this layer the calculated position of the large angle dip is 3° larger than the measured value. The depth of the dip and the rest of the curve are not strongly affected by the cermet layer.

Calculations[16-18] of the emission intensity of a biased junction share some important features with the calculation of the ATR response. First, an inelastic scattering mechanism for the tunneling electrons is envisioned. The interference of the incoming and scattered wave functions gives rise to ac currents in the film which, because of the symmetry of the junction, are located in a plane over the junction area. After the current fluctuations are Fourier analyzed in frequency and in the two spatial dimensions parallel to the plane, plane waves identical to those used in each layer in the ATR calculation are joined together at each interface until the emitted field strength in the prism is calculated. The success of this procedure in the ATR case implies that if the current fluctuations were known exactly, the emission intensity from prism-coupled junctions could be calculated as accurately as the ATR response.

The smooth curves in Fig. 2(a) show calculations of the angle dependence of the emitted p-polarized intensity of 590-nm light for two different assumptions about the current fluctuations. In both cases only current fluctuations along the axis perpendicular to the films are considered.[16] The dashed curve shows the calculated angle dependence assuming the current fluctuations are confined to the tunnel barrier. When the calculated curve is scaled to match the intensity of the fast mode peak, it fails to reproduce the rest of the measured curve. Furthermore, the agreement between the calculated and measured emission cannot be substantially improved by adjusting film thickness or dielectric functions without using values ruled out by the ATR results. We conclude that current fluctuations in the tunnel barrier cannot be solely responsible for the emitted spectrum. We have also been unable to reproduce the emission data by assuming a single current-fluctuation layer of any width anywhere in the junction.

The solid curve in Fig. 2(a) results from adding the emitted intensities from two incoherent current-fluctuation layers. By adding the intensities from two current-fluctuation layers in arbitrary amounts it is, of course, possible to reproduce the measured peak intensities of both the fast and intermediate modes for many choices of the location of the layers. However, the agreement between the data and calculation away from the peaks varies considerably for different choices of the current layer locations. The depth of the local intensity minimum near 50° is particularly sensitive to this choice. The solid curve is generated by placing 0.5-nm-thick current dipole fluctuation layers at both the Au-air and Al-MgF_2 interfaces. The current fluctuations at the Au-air interface are three times larger than those at the Al-MgF_2 interface. This calculation suggests that each SPP mode is created at the surface that supports it.

The microscopic mechanism responsible for the current fluctuations is not clear. A hot-electron mechanism[19] in which electrons tunnel through the barrier and collide with the Au-air interface and hot holes in the Al film collide with the Al-MgF_2 interface before they thermalize could be responsible for the current fluctuations. The residual roughness in our nominally smooth junction presents another possible source of current fluctuations. A roughness layer in an oscillating **E** field can act as a current fluctuation source.[20] Perhaps the tunneling current strongly excites the slow mode and the slow-mode fields overlap the roughness layers at both surfaces. In other words, the slow mode scatters into the fast and intermediate modes by residual roughness.[21,22]

There are undoubtedly other possible scattering processes that can produce SPP's, and it is possible that several of them are simultaneously responsible for the light emission from tunnel junctions. In any case, the use of ATR to characterize the radiative properties of the SPP's of light-emitting junctions can clearly establish some of the parameters needed to calculate the light emission intensity. The ability to examine two independent SPP modes on opposite sides of the tunneling barrier places additional constraints on the emission problem that can be used to evaluate theoretical models of the tunneling electron-SPP coupling.

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