Emission of light by the tunnel junction of a scanning tunneling microscope

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A new method has been developed for detecting the light emitted by the tunnel junction of a scanning tunneling microscope. Resonant features have been found on plots of the light intensity versus the applied voltage for Au–Au, Ag–Ag, Au–Pt, and Ag–Pt tunnel junctions. These features can be linked with the excitation of localized surface plasmons.

The information usually obtained by scanning tunneling microscopy consists of a map of the surface relief of the test sample and a spectrum of the electronic excitations, with a resolution reaching the atomic level. It seems promising to combine scanning tunneling microscopy with other methods for studying surfaces, in particular, observation of the spectra of inverse photoemission.[2] There is the hope that by combining methods in this way one could obtain information about the chemical composition of the surface with the spatial resolution characteristic of a scanning tunneling microscope.

The detection of light emitted from a tunnel junction between an iridium tip and a silver film deposited *in situ* was first reported in Ref. 2. These measurements were carried out in ultrahigh vacuum by means of a photomultiplier behind the window of the vacuum chamber. The light was detected at an angle $\sim 45°$ from the surface of the sample in a solid angle of 0.1 sr. Emission was observed over the interval $V_T \sim 2.5$–4.5 V of the voltage applied to the tunnel junction. Coombs *et al.*[2] suggested that this emission was caused by an excitation of localized surface plasmons:[3] collective excitations of the electron system analogous to plasmons in the interior of a metal. The nature of the observed emission, however, requires further study.

We have now developed a new method for detecting the light emitted by the tunnel junction of a microscope. As the tip of the tunnel microscope we use the end of a 200-μm quartz optical fiber (~ 0.3 m long), coated with a layer of metal along its generatrix. The second end of the fiber is cemented with Stycast-1266 to the input window of an FÉU-79 photomultiplier operating in a photon-counting mode. The geometry of these experiments has an obvious advantage over that of Ref. 2: The solid angle in which the photons emitted by the tunnel junction are detected approaches π sr. The experiments were carried out in air at room temperature. The design of the scanning tunneling microscope is described in Ref. 4.

This instrument is capable of operating as a scanning tunneling optical microscope:[2] At a fixed value of the voltage (V_T) applied between the tip and the sample, it

is possible to simultaneously record a relief map and a map of the intensity of the emission of light by the tip-sample junction. The resolution of this instrument reached ~ 10 Å. The light intensity varied by two orders of magnitude from one part of the surface of a sample to another, but there was no qualitative change in the V_T dependence of the intensity in these regions.

Figure 1 shows measurements of the light intensity as a function of the voltage V_T for gold and platinum samples and for optical-fiber tips coated with gold or silver. In each case, the results of ~ 50 measurements, carried out with various tips, on different parts of different samples, were averaged. These samples were metal films produced by vacuum deposition or by deposition from the plasma of a cathode discharge on the surface of glass or a metal foil. In the course of each measurement, the feedback circuit of the scanning tunneling microscope maintained a constant tunneling current, established at a point in the interval 30–100 nA.

The positions of the intensity maxima in Fig. 1 support the suggestion that the emission of light by the tunnel junction is associated with the excitation of localized surface plasmons in the volume between the tip and the sample. A comparison of the results for tunneling into Au, even at a qualitative level, shows that we are dealing with a collective phenomenon: The switch from Au to Ag as the junction material does

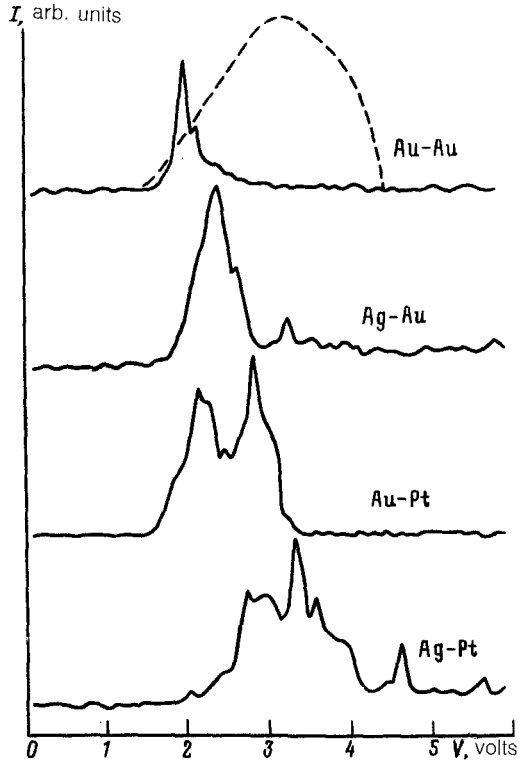

FIG. 1. Light intensity versus the voltage between the tip and the sample for Au–Au, Ag–Au, Au–Pt, and Ag–Pt tunnel junctions. The first metal of a pair is the tip material, and the second is the sample material. The dashed line shows the spectral sensitivity of the FÉU-79 photomultiplier.

not lead to an additional line, without a change in the existing line; instead, there is a shift in the position of the existing line. Rendell and Scalapino[3] have shown that a set of plasmon modes with frequencies no higher than the frequency of a surface plasmon with an infinite wave vector at a plane interface between two metals can be excited in a tunnel junction of arbitrary geometry:[5]

$$\omega_{sp} = \left(\frac{\omega_1^2 + \omega_2^2)^{1/2}}{2}\right), \tag{1}$$

where ω_1 and ω_2 are the bulk plasma frequencies of the metals. Under the assumption that the energy of the emitted photons is determined by the energy (eV_T) of the tunneling electrons, we would expect that if a plasmon excitation mechanism is operating, the positions of the peaks in Fig. 1 should depend linearly on the combination of the plasma frequencies of the tip and sample materials in (1). Figure 2 shows a plot of the voltage (V_T) across the tunnel junction at which the intensity of the emitted light reaches its maximum as a function of the combination ω_{sp} of the plasma frequencies of the tip and sample materials. Also shown in Fig. 2 is a point corresponding to the data of Ref. 2. As the point characterizing the Au–Pt junction in Fig. 2 we have shown the position of the peak on the right, which is at the higher value of the voltage between the tip and the sample. The peak on the left may be a consequence of the excitation of another geometric mode in the junction; the corresponding point is shown by a cross. The slope of the dependence in Fig. 2 is not 1; it is instead ~ 0.6. According to Ref. 3, one can work from this slope to estimate the typical dimensions of the microstructures at which the surface plasmons are excited. For a sphere of diameter a at a distance d from a flat metal surface, the frequency of a localized plasmon was estimated in Ref. 3

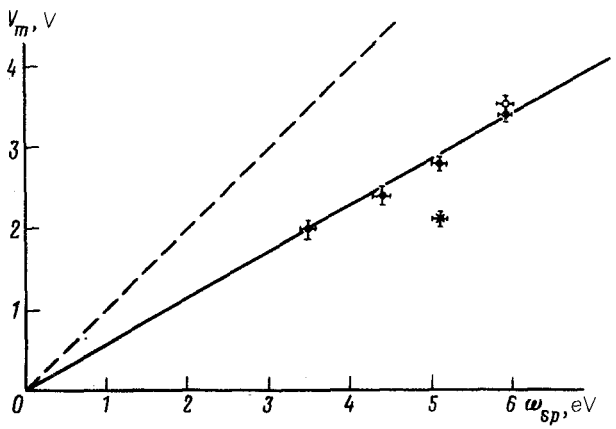

FIG. 2. Position of the peak of the light emission intensity versus the combination ω_{sp} of surface-plasmon frequencies of the metals forming the junction. Cross—Position of the left-hand peak for the Au–Pt junction; open circle–point corresponding to the data of Ref. 2 for an Ir–Ag junction; dashed line—straight line of unit slope. The frequencies ω_{sp} for Au, Ag, Pt, and Ir are 3.5, 5.4, 6.6, and 6.6 eV, respectively, according to Ref. 6.

to be $\omega \sim \omega_{sp}(d/4a)^{1/4}$. With $d \approx 5$ Å we thus find $a \sim 10$ Å. It thus appears that the peaks observed on the plot of the light intensity versus the voltage V_T stem from the excitation of plasma waves at microscopic irregularities at the surfaces of the tip and the sample with dimensions ~ 10 Å.

The strong coordinate dependence of the light emission intensity observed here agrees with the proposed plasmon mechanism. It is well known that light can be excited by surface plasmons only at microscopic irregularities of a surface,[7] which may be distributed in a nonuniform way along the surface.

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