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LETTER TO THE EDITOR

Possible observation of the direct emission of radiation from tunnel junctions

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Abstract. Light emission from tunnel junctions occurs when the tunnelling electrons excite surface plasmon polaritons (SPPs) and these SPPs decay by photon emission. Laks and Mills predicted an additional direct process in which optical frequency fluctuations in the tunnel current couple directly to the radiation field. We have observed an anomalous component in the light output from a nominally smooth aluminium–aluminium oxide–gold tunnel junction which has many of the features expected in direct emission.

It is now well established that the bulk of light emission from metal–oxide–metal tunnel junctions (Lambe and McCarthy 1976) is mediated by surface plasmon polariton (SPP) modes of the structure. The stimulation of such intermediate excitations and their subsequent decay to photons is the central theme of much recent experimental (Donohue and Wang 1986, Pappas *et al* 1988, Soole and Hughes 1988, Watanabe *et al* 1988) and theoretical (Kurdi and Hall 1986, Takeuchi *et al* 1988) work. However, in an earlier theoretical analysis of the light emitting tunnel junction, Laks and Mills (1979) (LM hereafter) described a direct emission process involving no intermediate excitation; in it, optical frequency fluctuations in the tunnel current couple directly to the radiation field. The direct emission is broad band, though peaked at the interband transition energy of the top metal electrode, namely at 2.5 eV in the case of gold and at 3.8 eV for silver. (These metals are the most frequently used top metal electrodes.) The direct emission is entirely p polarised irrespective of the degree of junction roughness (within the limitations of the theoretical description); this feature arises from the physically reasonable restriction that only tunnel current fluctuations normal to the plane of the thin film structure are considered. The calculated intensity is similar to that arising from scattering of SPPs off slightly roughened junction surfaces or interfaces. It is likely that the perturbative treatment of the surface roughness and its interaction with SPPs, as carried through by LM, is applicable only to the type of residual roughness found in nominally smooth devices such as those studied here, and does not extend to the more severe roughness of deliberately roughened devices.

In a subsequent theoretical study of similarly roughened tunnel junctions Arya and Zeyher (1983) again found a direct emission contribution. Their non-perturbative treatment of the surface roughness includes the effects of multiple scattering of SPPs. The emission due to SPPs is found to be a factor of about six greater than in the LM analysis. Thus, on the theoretical front the importance of the direct emission process is

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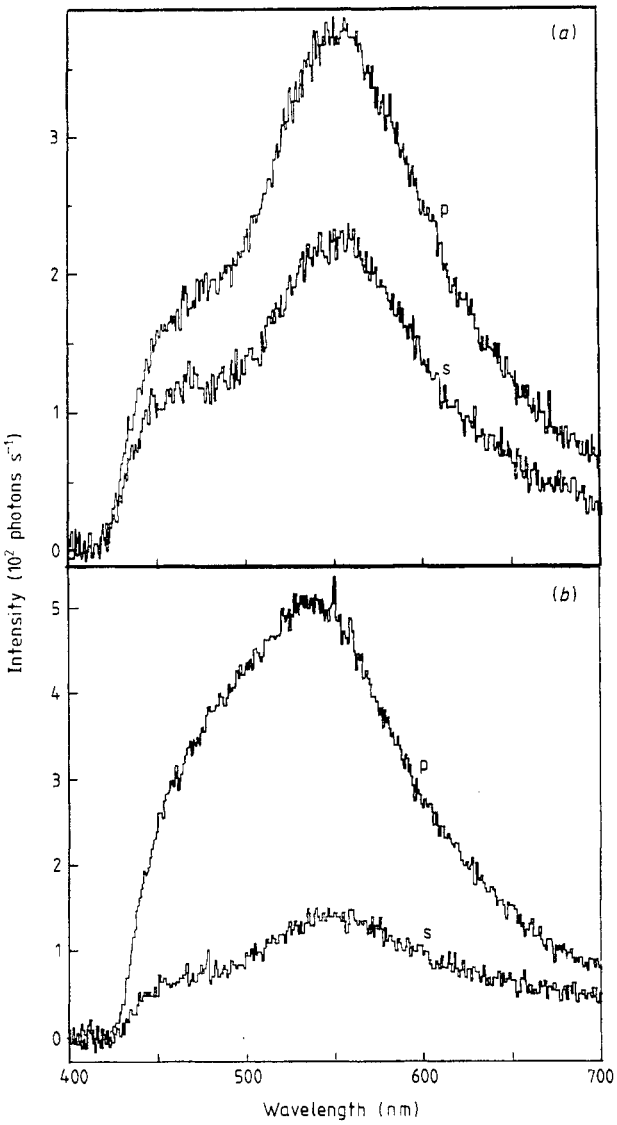

Figure 1. (*a*) Measured intensity of light (photon count) emitted by nominally smooth aluminium–aluminium oxide–gold junction at 0° to normal. p and s polarisation are shown as a function of wavelength. Applied DC bias = 2.95 V; current = 170 mA. (*b*) As (*a*) but at 60° to normal. Applied DC bias = 2.90 V; current = 150 mA.

somewhat eclipsed. Furthermore, no appeal has been made to the direct emission process to explain experimental results.

Here we report, after an extensive search, what is possibly the first positive observation of the directly emitted radiation described by LM and Arya and Zeyher. Though a large number of samples was studied the effect was prominent in only one. Alternative explanations of this result are considered.

The tunnel junctions were fabricated as nominally smooth aluminium–aluminium oxide–gold thin film structures in order to minimise SPP mediated emission. First a 50–60 nm thick, 7 mm wide, film of aluminium was deposited at 1.0–1.5 nm s^{-1} on a soda

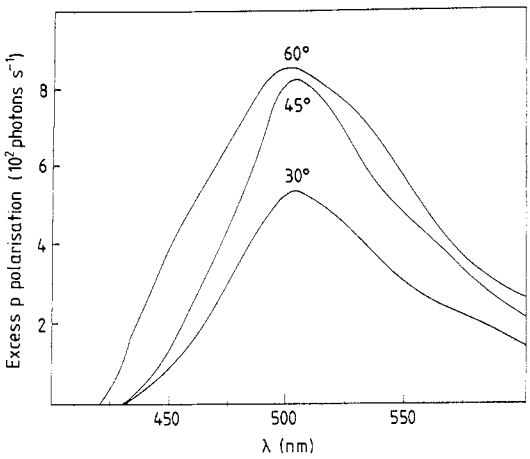

Figure 2. Excess p-polarised component (calibrated) emitted at 30°, 45° and 60° as function of wavelength. The p-polarised component associated with SPP mediated emission has been subtracted.

glass substrate. This was oxidised at 100–150 °C in air for 30 min; smooth films require longer oxidation times than rough films to produce the same barrier resistance. On top, a gold film 15–18 nm thick and 7 mm wide was cross deposited at 0.1–0.4 nm s^{-1}. The samples were then aged at room temperature until the barrier resistance had risen to the required value. Typical ageing times were 5–15 d. Thereafter the samples were stored and studied at 77 K as previously described (Ferguson *et al* 1989). Although the films were not deliberately roughened some intrinsic rugosity arises from the glass substrate surface finish and the grain structure of the evaporated metal films.

In all samples, except one, the light output was the same as, or showed only slight deviation in spectral form, polarisation and angular intensity variation from, that of deliberately roughened samples (Dawson *et al* 1984) though it was lower in intensity by a factor of 20 to 30.

In the exceptional case, anomalous behaviour was consistently observed over the several weeks' life of the sample. Spectra of the light emitted by this sample in both s and p polarisations for emission angles of 0° and 60° with respect to the junction normal are shown in figure 1. With regard to the spectra taken at 0° the distinction in polarisation is clearly nominal: the salient feature is a prominent absorption knee in the region of 2.5 eV which also is characteristic of the emission from deliberately roughened junctions. Away from the junction normal the emitted light takes on an interesting property: figure 1(*b*) shows that at 60° the strong 2.5 eV absorption knee is present in the s-polarised but completely absent from the p-polarised radiation. The excess p-polarised component reaches a broad maximum at 2.5 eV (figure 2). This excess p-polarised component is the only radiation component from either smooth or statistically rough gold junctions which we have found to peak rather than dip in the region of 2.5 eV. We tentatively identify it with the direct emission process first discussed by LM. This identification is strengthened when the angular variation of the excess p component is plotted (figure 3): a maximum at 60° is in accord with the LM prediction. Also, as predicted by LM, the intensity of the component identified as direct emission is comparable with that of the SPP mediated output.

In disagreement with the experimental results, the LM theory shows *roughness coupled* radiation has disappeared in the region of 2.5 eV; its absence arises from the assumptions by LM that the SPP junction mode is the intermediate excitation and that

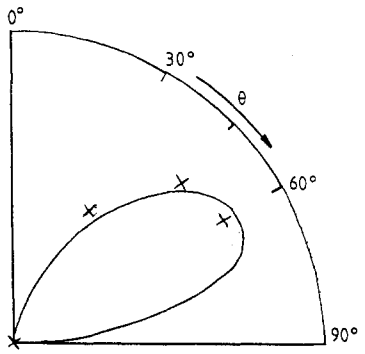

Figure 3. ×: Excess p-polarised component at 500 nm as a function of angle. The full curve is the LM prediction for the direct emission process.

this mode cuts off at 2.2 eV. Neither assumption is now accepted (Laks and Mills 1980, Soole and Hughes 1988, Kurdi and Hall 1986).

The most disturbing feature of the experimental results is that the direct emission intensity is not reproducible from sample to sample though some evidence of it is seen in other junctions. The explanation of this variation must lie in subtle differences between different electrode films; little else is involved in the devices. It remains to be determined what is the important underlying factor, for example, whether the thin gold top electrode has pinholes or voids which allow energy to be radiated from the junction interior. Characterisation of the electrode films by electron microscopy and scanning tunneling microscopy could be most informative. Further reduction of the top film thickness would be likely to increase the void area in the film and would be a useful additional diagnostic measure.

McCarthy and Lambe (1977) showed that silver particles with diameters of the order of a few tens of nanometers at a distance of 5 nm above a junction with gold top electrode can efficiently scatter the plasmon evanescent field outside the gold electrode. This structure emitted p-polarised light with a maximum intensity at 60°. Hansma and Broida (1978) showed related behaviour with 30 nm gold particles deposited directly on the tunnel barrier and then overcoated with a thin gold film. These two studies were restricted to lower DC bias and did not present polarisation or angular variation data so comparisons with the present work are necessarily limited. While a similar origin cannot be entirely ruled out for our results it seems most improbable since we took none of the extreme measures needed to produce metal particle electrodes.

An important related point concerns the possibility of direct emission centred around 3.8 eV in tunnel junctions having a *silver* top electrode. A sharp emission peak has been observed at 3.8 eV for both nominally smooth and statistically rough junctions of this type. In common with others we have attributed (Dawson and Walmsley 1986) the emission to a radiative plasmon mode supported by the thin silver electrode. We would maintain that this view is still correct and that the direct emission contribution is comparatively small. This duality of interpretation concerning the gold 2.5 eV and silver 3.8 eV emission is justified as follows.

For an ideal metal the plasma resonance energy, $h\omega_p$, occurs at the point where $\varepsilon_{real} = 0$; a sufficiently thin film of such a metal will support coupled surface oscillations, the high-frequency branch of which constitutes a *radiative* plasmon at the energy $h\omega_p$. For both silver and gold the bound d electrons make a contribution to ε_{real} in the energy range of interest but to somewhat different effect. In silver this positive contribution is strong and causes the condition $\varepsilon_{real} = 0$ to occur at the surprisingly low energy of 3.8 eV,

which is immediately below the onset of strong d-band absorption and an associated rise in $\varepsilon_{\text{imaginary}}$. The bulk plasma resonance at 3.8 eV may be viewed as a collective oscillation of the conduction electrons screened by the out-of-phase bound d electrons; were it not for the d electrons the plasma oscillation energy would be 9.2 eV. The occurrence of volume plasmons and thin film radiative surface plasmons at 3.8 eV in silver is well attested in the literature (Raether 1980). By contrast the d electrons of gold influence its dielectric function in such a way that the condition $\varepsilon_{\text{real}} = 0$ is brought about at a somewhat higher energy around 5.0 eV. (The energy is sensitive to the exact value of the optical data which differ slightly from one report to another. See Weaver *et al* 1981.) A d band to Fermi level transition occurs at 2.5 eV in gold where $\varepsilon_{\text{imaginary}}$ increases sharply. Consequently neither volume plasmons nor thin film radiative surface plasmons occur near the region of the interband transition (namely 2.5 eV). (Indeed because of the strong absorption above 2.5 eV in gold these plasma phenomena are not observed even when $\varepsilon_{\text{real}} = 0$.) In the gold tunnel junctions the p-polarised emission that peaked at 2.5 eV cannot be associated with a radiative plasma mode of the thin gold film.

The physical arguments outlined above are reiterated by two simple observations, namely bandwidth and intensity.

(i) For gold junctions the excess p-polarised emission is broad band in nature in agreement with the direct emission described in the LM theory. For silver junctions the emission (Dawson and Walmsley 1986) at 3.8 eV is considerably more narrow band (FWHM = 0.12 eV) than the theoretically predicted direct emission.

(ii) The second observation concerns the intensity of the radiation. For the excess p-polarised component at 2.5 eV from gold junctions we find the peak emission to be 750 photons s^{-1}, for a tunnel current density of 2 mA mm^{-2} and comparable with that of the SPP mediated emission from nominally smooth junctions. In the LM analysis the intensity of the direct emission from silver junctions is predicted to be a factor of seven or eight greater than that from gold junctions. For the same tunnel current density we find the emission from silver at 3.8 eV to be approximately 40 times as great as that from gold at 2.5 eV; the silver 3.8 eV emission is considerably more intense than the SPP mediated emission from nominally smooth junctions and is comparable in intensity with that from deliberately roughened silver junctions.

This preliminary report is aimed at stimulating further study of the effect observed.

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