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LETTER TO THE EDITOR

Dependence of light output from nominally smooth Al–Ox–Au tunnel junctions on electrode morphology

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Abstract. The light output from nominally smooth Al–Ox–Au tunnel junctions is observed to be substantially independent of the deposition rate of the Au film electrode. Films deposited quickly (2 nm s^{-1}) and those deposited slowly (0.16 nm s^{-1}) have similar spectral dependences and intensities. (This is in contrast to roughened films where those deposited quickly give out less light, especially towards the blue end of the spectrum.) The behaviour can be interpreted in terms of the ratio l_{ph}/l_{em} where l_{ph} and l_{em} are the mean free paths of surface plasmons between external photon emissions and internal electromagnetic absorptions respectively. Once l_{ph}/l_{em} exceeds 100, as it does on smooth films, grain size has little further effect on the spectral shape of the light output. In fast-deposited films there are two compensating effects on the output intensity: grain boundary scattering decreases it and greater surface roughness increases it.

Excitation of the free electrons at the surface of a metal can give rise to collective oscillations in the form of surface plasmons (Raether 1988). The decay of surface plasmons to photons has been extensively studied because of its intrinsic interest and also its significance in the context of light-emitting flat screen displays. Two channels are important in determining the decay: electromagnetic absorption within the material supporting the plasmon and external photon emission. The principal experimental methods of investigation are through the excitation of surface plasmons either by photons during attenuated total reflection of light (ATR), or by electrons in tunnel junctions. In the ATR type of study, surface plasmons are excited in a thin film under appropriate conditions of photon energy and incidence angle, and the plasmon decay is monitored through observation of light from the opposite face of the film (Kretschmann 1972). Surface plasmons are excited in tunnel junction sandwiches by tunnelling electrons and the subsequent plasmon decay is seen through light emission from an external surface (Lambe and McCarthy 1976).

Of these, the ATR studies relate to a simpler system, a single film structure, though the data in most cases are limited to a single wavelength. By contrast, the tunnel junctions involve two metal electrodes separated by a thin insulating tunnel barrier. The arrangement allows surface plasmons of a wide range of energies to be excited but the process is less confidently understood. Therefore it is desirable to explore both methods and compare the outcomes for compatibility of results. Ideally, the same metal would be studied in both situations. However, it is usually more convenient to study Au films in tunnel junctions because of fabrication considerations, and Ag films in ATR because of their low losses.

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Already it is known that Al–Ox–Au tunnel junctions emit more light when deposited over a layer of CaF_2 which serves to roughen all surfaces and interfaces in the structure (McCarthy and Lambe 1977). Furthermore, if the film supporting the plasmon is deposited quickly ($\simeq$ 2 nm s^{-1}) the tunnel junction light output is reduced by a factor of 10 at the red end of the spectrum and up to 50 at the blue end (Ferguson *et al* 1989), as compared with that from slow deposited ($\simeq$ 0.03 nm s^{-1}) films: increased elastic scattering of surface plasmons at grain boundaries is responsible.

ATR studies (Kretschmann 1972, Hall and Braundmeier 1978, and Naoi and Fukui 1990) of the angular distribution of light from Ag films reveal that the emission during decay of plasmons is determined by surface roughness in a direct way: the power spectrum of the roughness dictates the details of the light distribution. Analysis of the emitted light under 632.8 nm excitation shows (Ferguson *et al* 1993) that on *nominally smooth* substrates, such as glass, thinner films ($\simeq$ 45 nm), within the useful range, emit with greater intensity than thicker ($\simeq$ 60 nm) films. Furthermore, if the films are deposited quickly the emission is increased by a factor of about 2.5. Thinner, faster-deposited films can be deduced to be rougher. The light output from CaF_2 underlaid, i.e. *deliberately roughened*, films is much stronger than from nominally smooth films. It is greater by a factor of up to 10 if the films are deposited slowly ($\simeq$ 0.1 nm s^{-1}). However, if the films are deposited quickly ($\simeq$ 10 nm s^{-1}) elastic scattering of surface plasmons reduces that factor to 5. The question of interest is whether these relationships apply to light emitting tunnel junctions.

The present study is further motivated by the thought that they might be simpler to understand than deliberately roughened tunnel junctions, since eliminating the roughening layer reduces the number of possible sources of variability in the morphology of the top Au layer. There are few published data from light emitting Al–Ox–Au tunnel junctions on smooth substrates. Nominally smooth tunnel junctions of high resistance are relatively difficult to prepare. The reason for the difficulty in their manufacture rests in part in the long ageing time required for their preparation. This increases the possibility of irreversible degradation of the junctions. Why the smooth junctions require longer is not clear. The oxide growth, according to the usually accepted Cabrera–Mott theory (1949), involves the diffusion of Al^{3+} ions from the Al electrode through the already grown oxide. The oxide film on smooth films may be more compact and offer fewer ionic diffusion paths along grain boundaries (see Schaefer and Adkins 1991).

The Al–Ox–Au tunnel sandwiches were prepared on soda glass substrates by first depositing a 50 nm thick Al film at 1 nm s^{-1}. This film was then oxidized in air at 100–150 °C for 5 minutes. Subsequently 20 nm thick Au films were cross-deposited, some at a slow (0.16 nm s^{-1}) and some at a fast (2 nm s^{-1}) rate. The resulting sandwiches then had to be aged at room temperature for periods of 10–30 days for the barrier resistance to increase to the required value to sustain DC biases up to 4 V.

To perform the optical measurements the substrate was suspended from an edge connector in a bath of liquid nitrogen. When operated at 77 K the resistance of the samples is more stable and their lifetimes are longer compared with room temperature. The optical detection system has previously been described (Dawson *et al* 1984) with the exception that a photon counting system (PAR model 1109/1121A) replaces phase-sensitive detection. Using this set-up the spectrum of the light output normal to the film surface was examined for each tunnel junction.

The spectral outputs from two tunnel junctions with slow-deposited Au films are shown in figures 1(*a*) and 1(*b*) and from two tunnel junctions with fast-deposited films in figures 1(*c*) and 1(*d*). All the outputs from nominally smooth samples examined in this work were lower in intensity by factors of 10–50 compared to CaF_2 roughened samples

(Dawson *et al* 1984). An important cause is the smaller RMS heights, δ, of roughness features on nominally smooth films: the intensity of the emitted light varies as δ^2 (Hunderi and Beaglehole 1970).

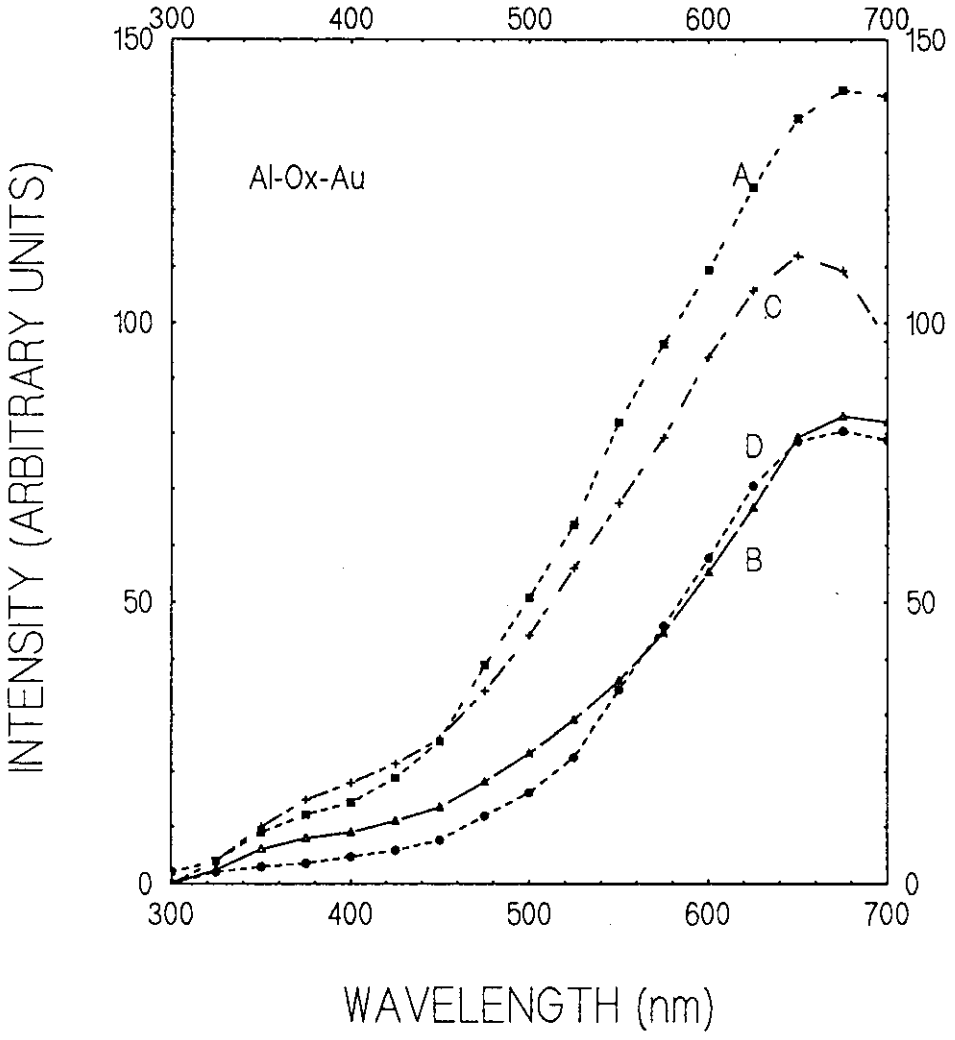

Figure 1. Intensity (photon count) of light emitted normal to the surface by Al–Ox–Au tunnel junctions: (*a*) Au deposited at 0.16 nm s^{-1}, $V = 4.3$ V, $i = 140$ mA, (*b*) Au deposited at 0.16 nm s^{-1}, $V = 4.3$ V, $i = 130$ mA, (*c*) Au deposited at 2 nm s^{-1}, $V = 4.2$ V, $i = 200$ mA and (*d*) Au deposited at 2 nm s^{-1}, $V = 4.0$ V, $i = 230$ mA. The intensity units are arbitrary but consistent for all junctions; corrections have been applied for the wavelength dependence of the optical detection system response.

Referring to figures 1(*a–d*) it is apparent that there is no strong influence on intensity or spectral shape with deposition rate. The same range of spectral forms is observed from

tunnel junctions both with slow- and fast-deposited Au films. These observations are in striking contrast to the behaviour of roughened films where fast evaporation reduced the overall intensity by a factor of 10 at the red end of the spectrum (Ferguson *et al* 1989).

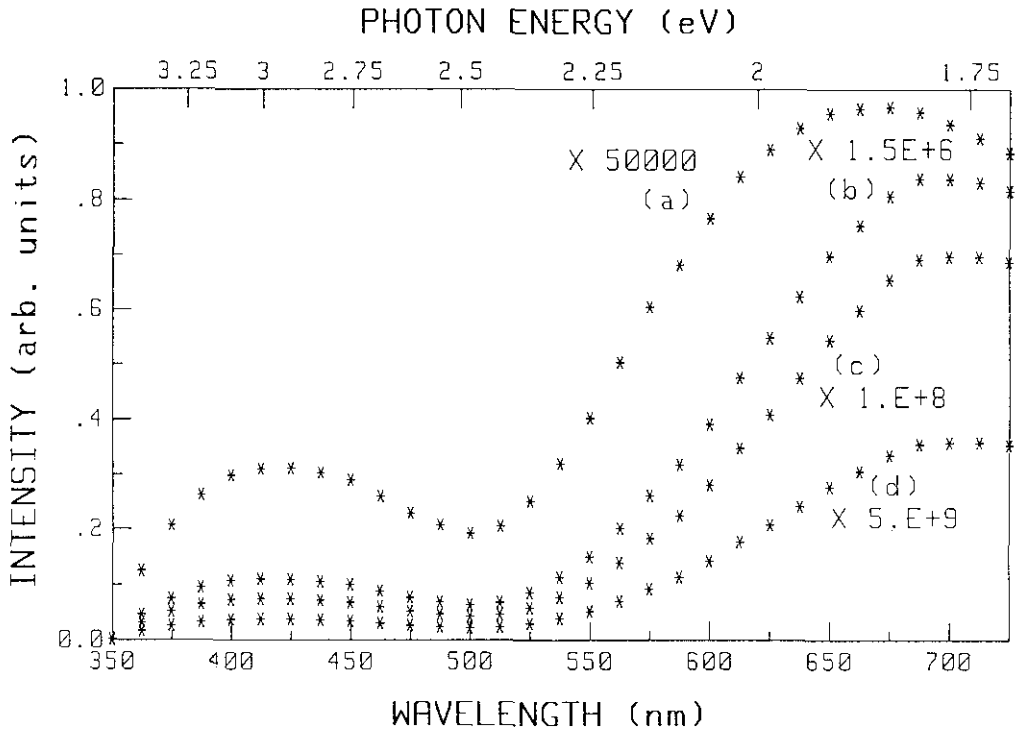

Figure 2. Calcualted light emission (photon count) as a function of wavelength from the nominally smooth Au surface of Al–Ox–Au tunnel junction for the following ratios of l_{ph}/l_{em}: (*a*) 10, (*b*) 100, (*c*) 1000, (*d*) 10000. For clarity the intensities have been scaled up by the values shown. In accordance with the l_{ph}^{-2} expectation at large ratio, curve (*d*) superimposes on (*c*) if a scaling factor of 10^{10} is used. In all cases the upper frequency cut-off is at 350 nm.

The model developed there (Ferguson *et al* 1989) to explain the results from roughened tunnel junctions considered the effect of elastic scattering of surface plasmon-polaritons (SPPs) at grain boundaries on the competition between the two plasmon absorption processes—emission of a photon and electromagnetic absorption within the film. These processes can be attributed to mean free paths of l_{ph} and l_{em} respectively. Fast-deposited films have smaller grains giving rise to greater elastic scattering of SPPs than in slow-deposited films.

With decreasing grain size, elastic scattering increases the SPP path between one incidence with a surface roughness feature and the next. Since electromagnetic damping per unit path length is unchanged, the ratio l_{ph}/l_{em} increases and the relative probability of internal dissipation of the SPP increases compared with photon emission. The grain boundary scattering was assumed to be Rayleigh-like and therefore more severe at short

wavelengths. A quantitative version of this model reproduced well the spectral forms observed for roughened Al–Ox–Au junctions (Ferguson *et al* 1989). Typical roughened junctions behave as if $l_{ph}/l_{em} \sim 10$–100 with $l_{ph} \sim 1$ mm in the red.

One would anticipate this model to be as applicable to nominally smooth tunnel junctions as to deliberately roughened ones. On smooth films the distance between roughness features capable of outcoupling a SPP to radiation, l_{ph}, is greater than on deliberately roughened films. An estimate from the emitted intensity would suggest a factor of 10 increase in l_{ph} corresponding to 10 mm in the red and $l_{ph}/l_{em} = 100$–1000. Of course, l_{em} has the same value irrespective of roughness. Consequently the ratio l_{ph}/l_{em} is greater for smooth films than for roughened ones. The effect of elastic scattering which should further increase l_{ph}/l_{em} is slight in this regime since l_{ph}/l_{em} is already large. The model calculation for the cases $l_{ph}/l_{em} = 10, 100, 1000, 10000$ is shown in figure 2. It illustrates that the curve shape is little modified as the ratio increases beyond 100 although the output intensity suffers further decrease (as l_{ph}^{-2}) as the ratio rises.

The intensity curves shown in figure 1, though showing some variation, are consistent in shape with the asymptotic form calculated for $l_{ph}/l_{em} > 100$. Remaining variations in the spectra are probably due to random factors in sample preparation. A greater contrast in output between fast- and slow-deposited films might have been predicted. In fast-deposited films with smaller grains, increased elastic scattering should reduce output. However, the development of surface roughness (Ferguson *et al* 1993) increases it and there appears to be substantial compensation between the two effects. The net outcome of a junction emission substantially independent of deposition rate is in contrast with the ATR result but is understandable. The film is only 20 nm thick in the tunnel junctions as compared with 50 nm in the ATR experiments and the roughening effects are therefore stronger in the junction. Differences arising specifically from the distinct properties of Ag and Au will not be addressed until suitable Al–Ox–Ag junctions can be fabricated and ATR measurements are made on Au films.

In summary, the spectral shape of the output from the tunnel junctions prepared on nominally smooth substrates is independent of deposition rate because grain boundary scattering increases further the ratio of l_{ph}/l_{em} which is already > 100, a region in which the increasing ratio has little consequence on shape. Essentially the spectral consequences of internal electromagnetic absorption of the surface plasmons are saturated. The intensity of the emitted light is not sensitive to deposition rate because additional roughening in films deposited at high rates leads to an increased emission which compensates the decrease expected when grain boundary scattering is greater. There is consistency between data from ATR and tunnel junctions.

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