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Efficiency of light emission from surface plasmons

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The attentuated-total-reflection technique was used to excite surface plasmons in thin silver films on various substrates. The efficiency of conversion of the surface plasmons in the films into visible light at the silver-air interface was measured as a function of film thickness and roughness. Approximate efficiencies for 50-nm-thick films were 5% on glass substrates, 9% on CaF_2-roughened glass substrates, and up to 80% on holographic gratings.

I. INTRODUCTION

The excitation and decay mechanisms of surface plasmon resonances in thin metal films have been of recent interest to many investigators.[1-6] For example, those studying surface-enhanced Raman spectroscopy have considered the contribution of surface plasmons to surface-enhanced signals.[7,8] Our own interest has been motivated by work on light-emitting electron tunnel junctions.[9-21] Here, light emission is believed to be the result of surface plasmons and localized plasmons (excited by tunneling electrons) decaying radiatively.

The current maximum efficiency of these junctions is low, about 10^{-5} W of light emission for every watt of electrical input power.[12] Why? One hypothesis is that, though surface plasmons are efficiently excited by tunneling electrons, their radiative decay is inefficient. Thus it is of interest to measure the radiative decay efficiency directly. The attenuated-total-reflection (ATR) technique pioneered by Kretschmann and Raether[22] provides an excellent method for determining the efficiency for well-characterized surface plasmons; the power going into the plasmon fields and the light emission can be directly measured. Although estimates of surface plasmon light emission efficiencies ranging from 0.01% to 20% exist,[23,24] no previous systematic studies have been published to our knowledge.

II. EXPERIMENTAL

We prepared the films on glass microscope slides. The silver was evaporated at $\lesssim 10^{-3}$ Pa from 99.999% pure silver shot in a Ta boat. Film thickness and deposition rate (~ 1 nm per sec) were monitored continuously during deposition by a quartz crystal monitoring system. The films were aged in air for two days prior to optical measurements because we found this helped in obtaining reproducible results.

For films on CaF_2-roughened substrates, we evaporated optical grade CaF_2 from a Mo boat at 10^{-3} Pa and a deposition rate of ~ 1 nm per sec immediately before the silver evaporation.

Holographic gratings were made by exposing spun-on photoresist (AZ 1350) on a glass slide to two interfering beams from an Ar-ion laser ($\lambda = 457.9$ nm). The grating spacing was determined from the intersection angle of the beams. The grating amplitude h was adjusted by changing the photoresist exposure time and measured using a formula derived by Heitman,[25]

$$h = \frac{\lambda}{2\pi}\left[\frac{I_0}{I_{1-}}\frac{1}{\cos\theta_0\cos\theta_{1-}}\right]^{1/2}.$$

Here, h is defined to be the peak deviation from the mean plane, λ and I_0 are the wavelength and intensity of an s-polarized beam (He-Ne laser in our case) incident at an angle θ_0 from the normal in the plane perpendicular to the grating grooves, and I_{1-} and θ_{1-} are the intensity and angle of the reflected first principal maximum in the back scattered direction. Though the validity of this formula breaks down for larger amplitudes ($h > 20$ nm), it can still be used to demonstrate the efficiency trend as a function of h.

Our ATR apparatus consisted of a 90° glass prism mounted on a manual turntable (Fig. 1). The Ag-coated glass slides were optically coupled to the hypotenuse face of the prism using microscope immersion oil as an index-matching fluid. The light source was a p-polarized He-Ne laser

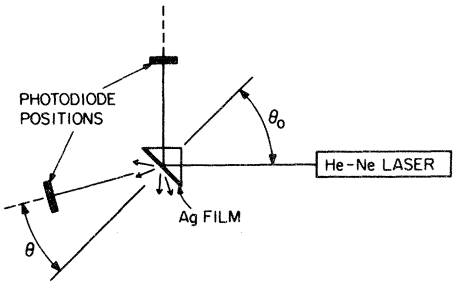

FIG. 1. ATR apparatus showing the He-Ne laser beam incident through the prism onto the Ag film and the positions of the photodiode detector.

($\lambda = 632.8$ nm) incident through the prism and slide onto the film. A large-area silicon photodiode 7.0 cm from the slide detected the intensity of the reflected specular beam and the plasmon emission from the Ag-air interface.

III. RESULTS AND DISCUSSION

Figure 2 shows experimental results for the intensity of the specularly reflected laser beam and the light emitted at the Ag-air interface as a function of incident angle θ_0. The peak absorption [Fig. 2(a)] and emission [Fig. 2(b)] were estimated as shown. The angle at which peak emission occurred was found by putting a large area photodiode (1×2 cm^2 active area) approximately 1 mm from the Ag film so that it collected as much of the emitted light as possible ($\simeq 60\%$ collection). The value of the peak emission was not, however, determined from this measurement because of the difficulty in accurately estimating the amount of light collected. Instead, peak emissions were determined by integrating over the angular power distributions shown in Fig. 3. The results agreed to within 10–20% of those obtained using an estimated collection efficiency for the photodiode held close to the film but were probably more reliable, since the amount of collection depended on the angular distribution.

The results of the absorption, emission, and efficiency measurements as a function of Ag thickness are displayed in Figs. 4 and 5; efficiency is defined as the power emitted divided by the power absorbed at a given angle. The efficiencies were measured at the peak emisssion angle which is slightly smaller than the peak absorption angle for Ag films thinner than 30 nm. The large errors reflect the difficulty in making reproducible samples. We do not understand all the contributing factors but imagine that they include irreproducible film nucleation and gas inclusions that depend on substrate temperature and residual gas composition during evaporation.

To investigate the roughness dependence, we looked at CaF_2-roughened 50-nm Ag films. There was a large uncertainty in the data, again indicating the difficulty in making identical films. Further, for CaF_2 thicknesses ≥ 200 nm the surface plasmon resonance was so broad that the light emission, though large, was only slightly peaked about the resonance angle. There is a discernible trend, however, toward saturation at 9% efficiency for CaF_2 thicknesses > 100 nm.

We see that randomly rough thin films have light emission efficiencies between 5% and 9%. This agrees with the theoretical calculations of Mills *et al.* where they estimate the probability

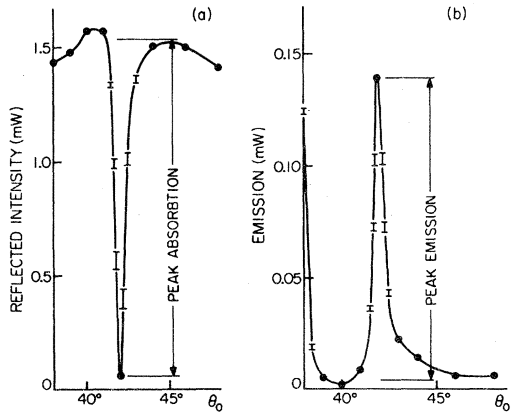

FIG. 2. (a) Specularly reflected light intensity and (b) surface plasmon light emission for a 40-nm Ag film as a function of incident angle θ_0.

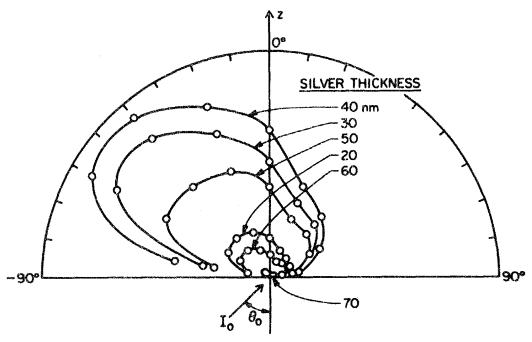

FIG. 3. Angular power distribution of emitted light in the plane of incidence; $\theta_0 \simeq 42°$, the surface plasmon resonance angle.

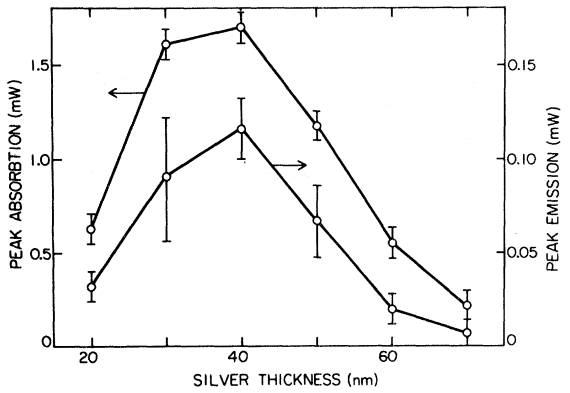

FIG. 4. Peak absorption and peak emission vs Ag film thickness.

of converting a surface plasmon into a photon to be 6%.[26] Their result is based on modeling the roughness as a perturbation to a flat surface using a Gaussian correlation function

$$\langle \zeta(\vec{x})\zeta(\vec{x}+\vec{r})\rangle = \delta^2 \exp(-r^2/a^2) ,$$

where ζ is the deviation from the flat surface, δ is the rms roughness height, and a is the transverse correlation length. They obtain the formula,

$$P = \tfrac{4}{3}(\delta a)^2 k^4 \omega_p \tau ,$$

where k is the surface plasmon wave vector, ω_p the electron plasma frequency, and τ the electron relaxation time. The close agreement depends on their probably fortuitous but unbiased (they had not seen our data) choices of $\delta = 5$ nm, $a = 50$ nm, $k = 10^5$ cm^{-1}, $\omega_p = 10^{16}$ sec^{-1}, and $\tau = 10^{-14}$ sec. Indeed, since δ and a were not measured and are probably different for different Ag thicknesses, we were satisifed to be within an order of magnitude of their estimate. Other investigations have found δ and a to be[27] $\delta = 0.7$ nm, $a = 300$ nm and[28]

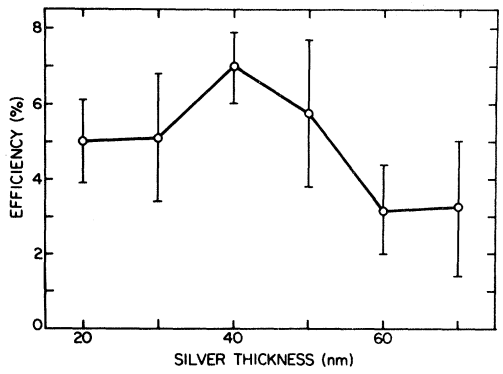

FIG. 5. Efficiency of surface plasmon light emission vs Ag film thickness.

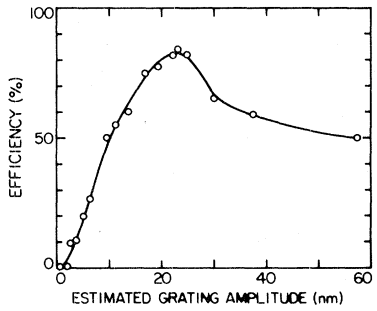

FIG. 6. Efficiency of surface plasmon light emission from 50-nm Ag films on 450-nm sinusoidal gratings vs estimated grating amplitude.

$\delta = 1.8$ nm, $a = 210$ nm. These values in the formula of Mills *et al.* (assuming k, ω_p, and τ are unchanged) give probabilities of 6% to 17%.

The total plasmon light emission from a surface is believed to be the sum of the emitted light from each Fourier component of roughness. We also looked at the light emission from 50 nm of Ag evaporated on sinusoidal holographic gratings. The efficiency results for grating spacing ~ 450 nm are plotted in Fig. 6 as a function of estimated grating amplitude. Note that the efficiency rises sharply with increasing h to $\gtrsim 80\%$ near $h = 20$ nm and decreases slowly thereafter. The results are consistent with those of Heitmann[29] who made similar measurements, and with the perturbation theory of Mills *et al.* when $h < 10$ nm. (Perturbation theory breaks down for larger h.[30,31]) Note also that a grating of appreciable amplitude is a much better light emitter than a randomly roughened film, as has been demonstrated by Kirtley, Theis, and Tsang.[13,14]

IV. SUMMARY

We find that surface plasmons in silver films radiatively decay with relatively high efficiency.

(i) The efficiency for films on glass substrates is not strongly dependent on film thickness for thicknesses between 20 and 70 nm. It is of order 5%.

(ii) The efficiency increases with CaF_2 underlayer thickness to a limitng value of 9% for CaF_2 thickness > 100 nm.

(iii) The efficiency increases rapidly with grating amplitude to a maximum of $\gtrsim 80\%$ near $h = 20$ nm and decreases slowly thereafter for gratings with a wavelength of about 450 nm.

(iv) The efficiencies for the films on glass slides and low amplitude gratings can be understood within the framework of a theory developed by

Mills *et al.* The efficiencies for films on higher amplitude gratings ($\gtrsim 10$ nm) cannot be modeled with existing theories because perturbation theory breaks down.[29–31]

Finally, because the decay process is relatively efficient, we conclude that if these types of surface plasmons could be efficiently excited by tunneling electrons, then tunnel junctions could be efficient light emitters.

ACKNOWLEDGMENTS

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