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Near-field optical measurement of the surface plasmon field

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The intensity of the evanescent electromagnetic wave of optically excited surface plasmons was measured directly using a scanning tunneling optical microscope (STOM) setup. When resonant coupling of the driving field to the surface plasmons was achieved, the measured intensity was increased by a factor of 30 larger than the corresponding evanescent wave intensity on a bare glass surface, in agreement with the theoretical prediction. Experimental results are presented for three laser wavelengths (514 nm, 633 nm, 670 nm). Possible applications of the technique to study surface plasmon fields are discussed.

1. Introduction

Optically excited surface plasmons have become a well established probe for the sensitive analysis of metal surfaces [1,2]. The sensitivity of the method is based on the fact that the electromagnetic wave associated with the surface plasma oscillation is confined to a narrow region close to the metal surface [3]. Therefore small changes in surface roughness or adsorbate coverage considerably affect the phase velocity and damping of the surface plasmons.

Surface plasmons are usually excited by coupling them to an evanescent wave originating from the total internal reflection of a light beam at a glass surface. By varying the angle of incidence of the light beam, the wave vector of the evanescent wave can be varied. When the latter coincides with the surface plasmon wave vector, most of the photons of the illuminating beam are lost by conversion into surface plasmons. This resonance phenomenon is easily observed by measuring the attenuation of the light intensity reflected from the glass surface (ATR). Whereas the resonance enhancement of the electromagnetic surface plasmon field has been observed indirectly by measuring the light diffusely scattered from the metal surface [1], no direct measurement of the electromagnetic field of the surface plasmons has yet been performed.

2. Experiment

In the present work, a scanning tunneling optical microscope (STOM) is used to measure the evanescent optical field of surface plasmons propagating on the surface of a silver film. The Kretschmann geometry is used to excite the surface plasmons optically (cf. fig. 1). A fiber tip probes the amplitude of the exponentially decaying light field leaking into the half space above the sample. The intensity of the internally reflected light is measured separately in order to obtain an independent determination of the

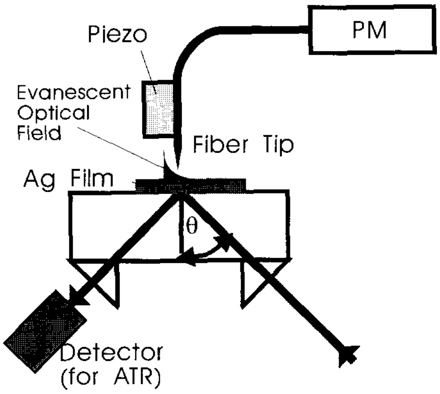

Fig. 1. Experimental arrangement used to measure the field enhancement. Surface plasmons are excited by total internal reflection in the Kretschmann geometry and detected from above by a STOM.

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surface plasmon resonance angle. The upper part of the apparatus in fig. 1 is essentially a STOM.

The silver film (with a typical thickness of 70 nm) covers only part of the substrate. Thus, by a lateral displacement of the sample, the evanescent field with and without the metal can be compared. The angle of incidence of the laser beam can be adjusted without changing the position of the spot on the sample by means of a telescope arrangement, and is calibrated with reference to the onset of total internal reflection. The fiber tip, attached to a piezo tube for vertical positioning, is centered above the laser spot which has a diameter of ≈0.3 mm. The fiber tip was fabricated from a monomode fiber designed for a wavelength of 633 nm by pulling it apart in a flame. The tip radius as determined by scanning electron microscopy is typically ≈1 μm. Of the incident light with an intensity of several mW, only a few nanowatts were extracted by the tip and detected by a photomultiplier tube. To within the precision of our apparatus, the intensity measured through the fiber was at a maximum when the total internally reflected intensity was at a minimum. This corresponds to resonant excitation of surface plasmons.

The magnitude of the evanescent field was obtained as follows: as the fiber tip is approached to the sample, the intensity increases exponentially with decreasing separation (fig. 2). On a bare glass plate the exponential decay length is given by the index of refraction, the wavelength and the angle of incidence θ [4]. Typical values for this length are 100 to 1000 nm. When the tip touches the sample surface the signal saturates and then exhibits a complicated and not very reproducible behavior indicating optical contact between tip and sample. The intensity at the onset of saturation (I_0) is a direct measure of the amplitude of the evanescent light field. Since the coupling between the evanescent field and the fiber tip strongly depends on the tip shape (which is not well characterized), it is difficult to estimate absolute intensities. Therefore we will give the measured field intensity in arbitrary units.

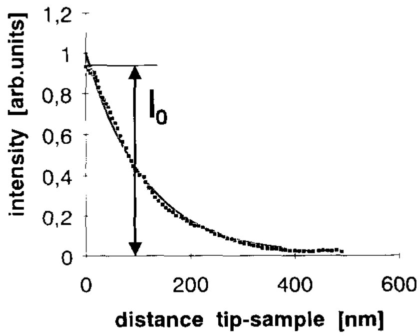

Fig. 2. Typical behavior of the evanescent field above a glass plate, measured by varying the tip–sample distance z and monitoring the fiber output. The decay length of the exponential fit (solid line) is 116 nm, while the theoretically expected value is 138 nm for the given geometry.

3. Results and discussion

Resonance curves are measured at three wavelengths (514, 633, and 670 nm) from different laser sources. The laser power is kept below 17 mW to avoid damage to the silver film. For each wavelength we compare the detected signal on the silver film to the signal on the uncoated glass surface. The experimental results are summarized in table 1; the resonance curves for two wavelengths (670 nm and 514 nm) are shown in fig.3. The intensity of the evanescent wave was calculated numerically using the transfer matrix representation of Fresnel's formulae [5,6] (see full lines in fig. 3). The data for the optical properties of silver were taken from ref. [7].

We define the field enhancement ratio F as the ration between the maximum intensity of the evanes-

Table 1

Comparison of measured and calculated values of the plasmon field enhancement factor, resonance width and angle for different laser wavelengths. For details see text.

	Wavelength (nm)		
	514	633	670
F_m (measured	17.0±2.3	12.8±0.9	34.2±9.3
F_c (calculated)	22.1	17.0	38.6
F_m/F_c	0.77	0.75	0.88
θ_m (measured)	43.90°	42.89°	42.67°
θ_c (calculated)	43.89°	42.83°	42.57°
$\Delta\theta$ (measured)	0.45°	0.21°	0.25°
$\Delta\theta$ (calculated)	0.23°	0.15°	0.075°
Film thickness used in calculation (nm)	70	70	70

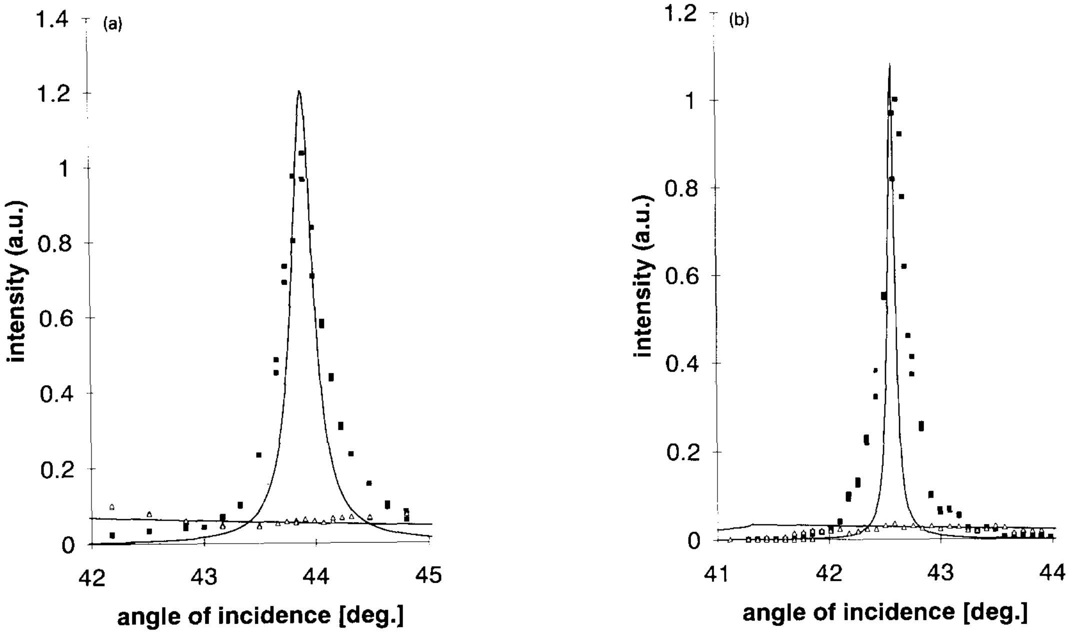

Fig. 3. Detected intensity of the evanescent field at the wavelengths 514 nm (a) and 670 nm (b). The "squares" correspond to the field on the silver film (plasmon field), while the "triangles" are the measurements on the bare glass substrate. The solid lines represent the numerically calculated intensities. Theory and experimental data are scaled such that the evanescent waves on bare glass have the same intensity.

cent field with plasmon excitation and the field intensity of the evanescent wave on the bare glass surface:

$$F=\frac{|\text{field with metal film at resonance}|^2}{|\text{field without metal field}|^2}\,.$$

Table 1 shows the measured and calculated field enhancements F as well as the resonance angles θ and the widths $\Delta\theta$ of the resonances.

The angle measurement is accurate within 0.15°. The good agreement with theory in the position of the resonance is manifest. The experimental values of the resonance width are 2 to 3 times larger than the calculated values. Possible reasons for this are that the laser beam divergence varies for the different laser types and that the plasmon excitation is damped by the fiber. For an incident intensity of $\approx 100\ \text{nW}/\mu\text{m}^2$, an assumed probe area of the order of $1\ \mu\text{m}^2$ and an observed intensity at the end of the fiber of ≈ 1 nW, about 1% of the intensity in the volume between tip and sample is estimated to be coupled out by the fiber. This results in a non-negligible local damping of the surface plasmons. This local damping, however, does not appear in the ATR measurement, which integrates over the global light beam.

The experimental field enhancement on a freshly evaporated film is ≈ 70 to 80% of the theoretically calculated value of about 30. The field enhancement strongly depends on the thickness and quality of the film. Apart from the uncertainty of the film thickness which could be responsible in part for the differences between measurement and calculation, the aging of the silver (structural changes and chemical reactions with the air) leads to a pronounced decrease of the enhancement factor within a few days. Thus we can conclude that our measurements are in accordance with the theoretical predictions.

4. Conclusions

Figure 3 shows that we are able to determine the surface plasmon resonance angle with the STOM fiber tip, i.e. with a spatial resolution roughly given by the fiber tip diameter. Hence a combination of surface plasmon excitation and STOM is an appealing novel instrumental technique: The first method has a submonolayer thickness resolution for adsorbates [8] and a high sensitivity to changes in the optical properties of the surface region. Its spatial resolution is limited to the surface plasmon decay length, which is on the order of 10 μm. The STOM, on the other hand, has a lateral resolution appreciably better than the laser wavelength [4,9].

Moreover, information on the propagation of the surface plasmons on structured surfaces could be extracted as well. Such local measurements might be useful as a test for theories of plasmon propagation on rough surfaces [10]. Unlike measurements of the interaction of an STM tip with surface plasmons [11,12] where the measured quantity is the disturbant of the plasmons caused by the tunneling tip or the rectification of the tunneling current, the STOM directly measures the optical plasmon field on the surface with a detector placed in the near field region. Finally, the enhanced light intensity of surface plasmons could improve the signal to noise ratio of normal STOM operation.

The combined technique has a number of possible applications including, for example, the characterization of mirrors for neutral atoms based on evanescent fields [13,14]. Atoms interacting with the strong field gradient of the plasmon field are deflected by the induced dipole force. Since the atoms will approach the plasmon carrying surface to a distance less than an optical wavelength, the near field structure of the plasmon field will become important for the performance of the mirror. For instance a metal surface with nanometer-sized corrugation has a considerable variation in the plasmon near field and might produce some unwanted scattering of neutral atoms.

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