Determination of the spatial extension of the surface-plasmon evanescent field of a silver film with a photon scanning tunneling microscope

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A photon scanning tunneling microscope is employed to probe the surface-plasmon field in the evanescent region of a silver film for p (parallel to the plane of incidence) and s (perpendicular to the plane of incidence) polarizations of the light beam at several angles of incidence near the critical angle. The interaction between the field and the probe is measured and compared to theoretical calculations involving a single four-media model. A systematic analysis of images obtained for several positions of the optical fiber above the film is presented and it is shown that, for tip-to-sample distances smaller than half the wavelength of the incoming light, the collected intensity curves are identical in any area of the sample.

I. INTRODUCTION

In this paper, we study the evanescent behavior of the surface plasmons (SP), previously described by Ritchie,[1] excited in metallic films in the Kretschmann configuration.[2] The extension, above the surface, of the nonradiative surface plasmons is detected locally by the optical probe of the photon scanning tunneling microscope (PSTM). This microscope described by Reddick, Warmack, and Ferrell[3] at the Oak Ridge National Laboratory and since developed by several groups[4,5] is the optical analogue of the well-known scanning tunneling microscope (STM).[6] In our configuration a transparent sample is placed on the base of a hemicylindrical prism (index of refraction n_1) illuminated by a laser beam in total internal reflection (TIR) as represented in Fig. 1. Total internal reflection occurs at the prism-air interface and the evanescent field in the medium of lesser index of refraction ($n_3=1$ for air) is converted into progressive waves by a sharpened optical fiber brought sufficiently close to the sample (typically a few tens of nanometers). The probe is scanned over the sample surface and variations of the intensity detected by the fiber are measured and treated by a suitable detector-computer system.

A theoretical study of the field collected by the optical fiber is given in Sec. II. A comparison between these calculations and the experiment is developed in Sec. III. In Sec. IV we discuss the PSTM-field isointensity lines in the medium above the sample as a function of the sample probe distance. We give our conclusions in Sec. V.

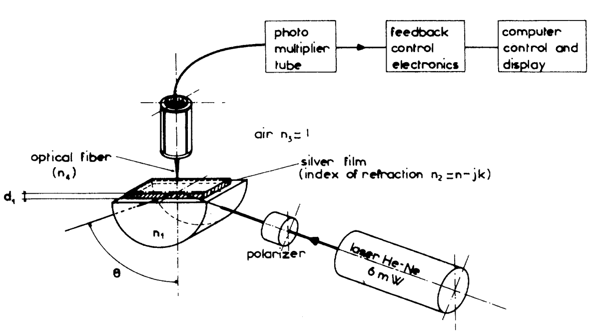

FIG. 1. Schematic of the photon scanning tunneling microscope.

II. TRANSMITTED FIELD: FOUR-MEDIA MODEL

The basic purpose of the PSTM is to locally detect the transmitted field in the evanescent region of diffraction of a sample. In the present study, the optical fiber of the microscope is first brought close to the surface and then moved back in order to follow continuously the intensity decrease as a function of fiber-sample distance. The problem to be studied is a four-media problem, since the presence of the fiber modifies the electromagnetic field transmitted by the silver film. The field is frustrated by the sensor and propagates up to the detector. To calculate the intensity of the light transmitted in the fiber we have considered the fiber as a semi-infinite medium of quartz with an index of refraction $n_4=n_1=1.458$. Indeed, we have shown in a previous paper[7] that even with this rough approximation the theoretical variations of the electric field above a plane surface were in good agreement with the experiments. This model described the shape of the decay but neither the value of the collected intensity nor the lateral variations of the electromagnetic field.

The transmittance in the fiber has been calculated by dividing the flux of the Poynting vector through a unit surface of the glass by the flux of the incident light. From the boundary conditions at the three interfaces $z=0$, $z=-d_1$, $z=-d_2$, the transmittance in p polarization in the fourth medium can be expressed as in Ref. 8.

The intensity of light transmitted in the fiber as a function of its distance to the sample is presented in Fig. 2 for several values of the angle of incidence in p polarization. The values of the parameters wavelength, indices of refraction, and film thickness are, respectively, $\lambda=632.8$ nm, $n_2=0.05-j4.3$,[9] and $d_1=50$ nm.

In Fig. 2 it can be seen that the intensity of light col-

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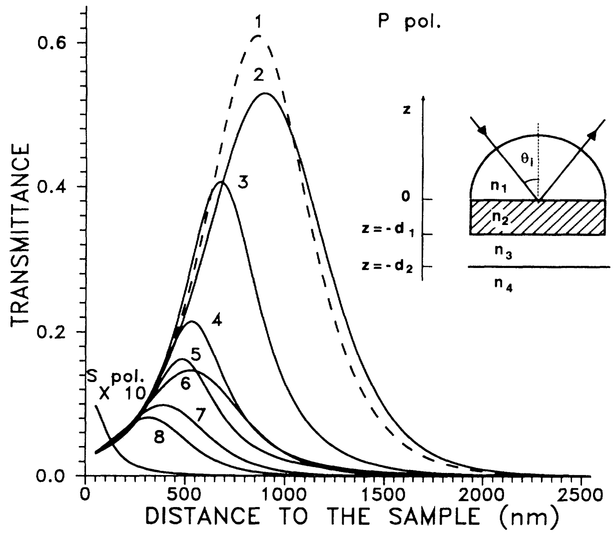

FIG. 2. Transmitted intensity in the fourth medium in p polarization as a function of the distance between the silver surface and the fiber tip. $\theta_p=44.9°$ in p polarization. (1) $\theta_p-0.1°$, (2) θ_p, (3) $\theta_p+0.5°$, (4) $\theta_p+1°$, (5) $\theta_p+1.5°$, (6) $\theta_p-0.5°$, (7) $\theta_p-1°$, (8) $\theta_p-1.5°$ and in s polarization θ_p.

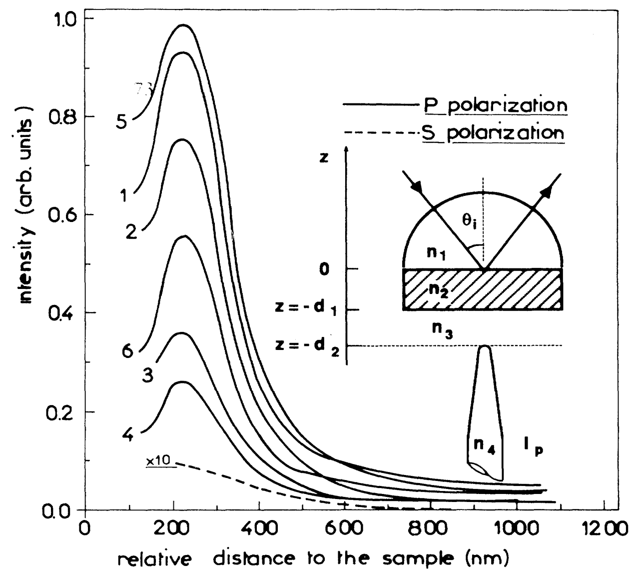

FIG. 3. Experimental transmittance in the fourth medium in p polarization as a function of the distance between the silver surface and the fiber tip. $\theta_p=44.9°$ in p polarization. (1) θ_p, (2) $\theta_p+0.5°$, (3) $\theta_p+1°$, (4) $\theta_p-1.5°$, (5) $\theta_p-0.5°$, (6) $\theta_p-1°$ and in s polarization θ_p.

lected by the fiber is no longer maximum at the surface. The position of this maximum varies as a function of the angle of incidence. This is a phenomenon already encountered for a four-media system without any losses.[8] It must also be noted that the maximum of transmittance is located at θ_p for distances greater than 1.2 μm and is progressively shifted to smaller values when the distance decreases. It shows that the electromagnetic field near the surface and, consequently, the plasma oscillations are greatly disturbed by the presence of a fourth medium.

III. EXPERIMENT

A. Intensity collected by the fiber

The experimental setup (Fig. 1) has been described previously.[7] The optical probe of the PSTM is a multimode optical fiber chemically etched in a fluorhydric acid solution. The radius of its extremity is about 50 nm. The optical fiber is attached to a piezoelectric tube and can be scanned over the sample. The light source used in this experiment is a 2.8-mW He-Ne laser ($\lambda=632.8$ nm), but any other laser or even white light can be used.

A thin silver film ($d_1=50\pm2$ nm) has been evaporated on the base of a hemicylindrical prism ($n_1=1.458$) using a standard technique ($p<10^{-5}$ mbar, room temperature, evaporation rate 1 nm/sec). Then the prism is placed on the PSTM sample holder and studied immediately after preparation to avoid oxidation or aging effects. All experiments reported here have been performed with the same optical fiber.

Figure 3 illustrates the variations of the intensity of light collected by the optical fiber for p polarization measured as a function of its distance to the sample for several angles of incidence near $\theta_p=44.9°$. In order to avoid any contact (and consequently probable modification of the tip) between fiber and sample, we have limited the measurements to a minimum distance probe of silver film of about 10 nm so that the study could be completed as a function of the different parameters with the same tip. Experimentally we have detected a shift between the curves (1–10 nm), but this determination is inaccurate due to the absence of a common origin for the different curves. Consequently, we chose to align the maxima in Fig. 3.

Due to the interaction between the surface and the fiber tip, the intensity of the field is not maximum at θ_p. It goes through a maximum for $\theta_p+0.5°$ and decreases on both sides of this angle, which is in good agreement with the theoretical results shown in Fig. 2.

The widths of the experimental peaks (Fig. 3), as well as the shifts of the maxima, are smaller than those obtained from the theoretical calculations, but it must be pointed out that the curves in Fig. 2 have been calculated from a rough model (four-media model) in which the tip was modeled as a perfectly semi-infinite medium.

In Fig. 4 we have illustrated the intensity of light measured in s polarization with the same experimental parameters. We note the rapid decay as a function of the distance between sample and fiber, as was expected from the theoretical data (Fig. 2). The intensity collected by the fiber is roughly a hundred times weaker than in p polarization, as can be seen in Fig. 3 in which, for comparison, we have reported the experimental results (dashed line). It confirms the concentration of the electromagnetic field density at the silver/air interface in p polarization near the critical angle.

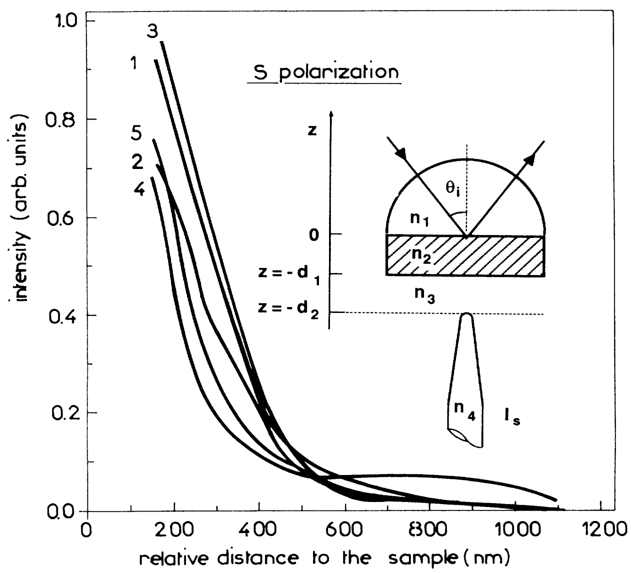

FIG. 4. Experimental transmittance in the fourth medium in s polarization as a function of the distance between the silver surface and the fiber tip, in s polarization. (1) $\theta_p=44.9°$, (2) $\theta_p+0.5°$, (3) $\theta_p-0.5°$, (4) $\theta_p+1°$, (5) $\theta_p-1°$.

B. Penetration depth

For the general case of rough surfaces, one defines a local penetration depth $d_{pl}(z_0)$ at a distance z_0:[8]

$$d_{pl}(z_0)=\frac{2I(z_0)}{\left[\frac{dI}{dz}\right]_{z_0}},$$

where $I(z_0)$ is the intensity of light collected by the fiber at a distance z_0. The local penetration depth is related to the local slope at each point of the decay curve. We have

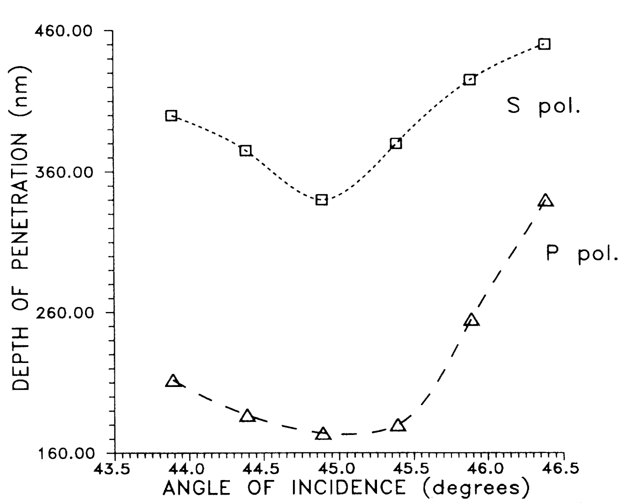

FIG. 5. Depth-penetration minimum d_{pl} as a function of the angle of incidence in the two modes of polarization.

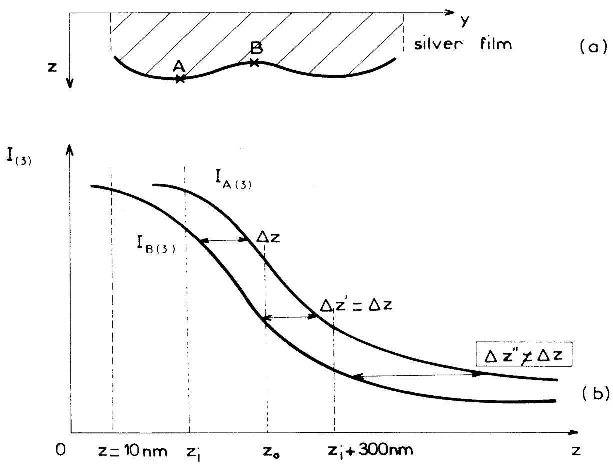

FIG. 6. Intensity collected by the fiber tip vs the probe-to-sample distance before two different areas of the silver film.

calculated the minimum value of the local penetration depth (d_{plmin}) for each curve of Figs. 3 and 4. In Fig. 5, d_{plmin} is shown as a function of the angle of incidence for s and p polarizations. For s polarization, d_{pl} is twice as large as for p polarization, and it appears that the minima occur for angles of incidence close to θ_p. We conclude that a better vertical resolution is obtained when the PSTM is operated in p polarized light for these angles of incidence. The figures presented in Sec. IV have been obtained in these experimental conditions.

The curves shown in Figs. 3 and 4 have been obtained for a fixed position of the tip, say A, above the sample [Fig. 6(a)]. For another position of the fiber, say B, the decrease in intensity as a function of the distance presented the same characteristics in terms of local penetration depth for probe-to-sample distances ranging from z_i to z_i+300 nm, as has been schematically represented in Fig. 6. It can also be seen in Fig. 6(b) that when the fiber-sample gap increases, the local slopes of the decaying field display stronger relative variations. Propagative waves become indeed dominant (diffused light due to the roughness of the surface) with respect to the evanescent ones. They give rise to interferences modifying significantly the local slopes. Thus, it can be expected that the amplitudes Δz of the isointensity lines of the electromagnetic field depend on the position z of the tip.

IV. PSTM IMAGES OF A THIN SILVER FILM

In the constant-intensity mode,[4] the PSTM gives an image which is a map of the isointensity lines of the electromagnetic field due to the interaction of light with the sample-fiber system. The feedback loop of the electronic controller maintains the current constant by acting on the elongation of the piezoelectric tube in the direction perpendicular to the sample surface. A PSTM image is the recording of these corrugations when the optical fiber attached to the piezoelectric tube is scanned over the sample.

We have examined in a previous paper[10] the PSTM

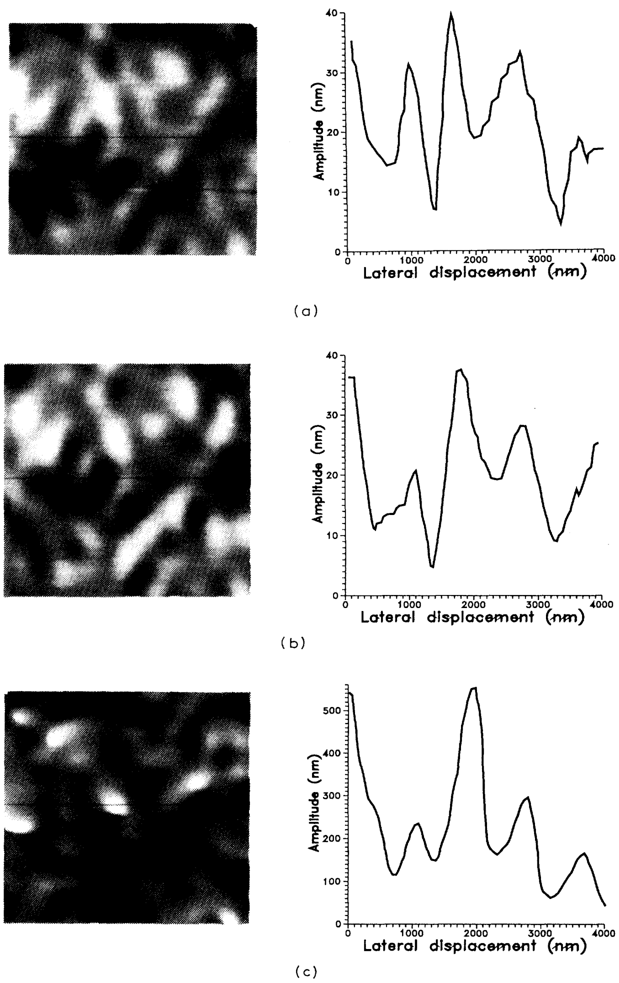

FIG. 7. PSTM images of the 50-nm silver film for different distances between the metal surface and the fiber tip (from z_0 to z_0 +630 nm, the scan range is 4×4 μm^2). (a) z_0, (b) z_0+120 nm, (c) z_0+630 nm.

isointensity lines of nonabsorbing calibrated samples—quartz gratings or quartz-crossed gratings—and shown theoretically and experimentally that in p polarization the amplitudes of the field lines (corrugation) were amplified when the tip was moved back off the sample surface. For the simple grating, the interferences appearing in the far field of diffraction are related to the existence of propagative orders.

We have prepared a 50-nm-thick silver film on quartz and repeated the experiments in the constant-intensity mode, in p polarization, and with the following parameters: $\lambda=632.8$ nm, $\theta_p=44.9°$. We have studied systematically the variation of the corrugation as a function of the fiber-sample distance on a 4×4 μm^2 scanning range with a random noise in the feedback loop corresponding to a corrugation of a few nanometers. Figure 7(a) illustrates the PSTM image obtained when scanning at a given distance z_0 very close to the sample. In the zone where the evanescent field is dominant, Figs. 7(a) and 7(b) (z_0+120 nm) show that the relative amplitude of the lines of electromagnetic fields are slightly affected by the probe-to-sample distance. On the other hand, in Fig. 7(c) (z_0+630 nm) important variations of the amplitude are recorded. This behavior is close to that observed for a calibrated surface.[10] Note that the corrugations obtained for θ_p [Figs. 7(a)–7(c)] are the same for any other angle of incidence $\theta>\theta_p$.

V. CONCLUSION

In this paper, we have shown that surface excitations can be studied with the PSTM. The spatial extension of the surface-plasmon field of a thin silver film has been probed by the sharp optical fiber of the microscope brought into the evanescent region. A qualitatively good agreement has been demonstrated between experimental results and a theoretical analysis based on a very simple model tip. The interaction between the tip and the plasmon field results in a shift of the resonance incidence angle and a minimum of the local penetration depth for p polarization which can be used to improve the vertical resolution of the instrument. Finally, a systematic study of the experimental decay of the field for sample-to-tip distances up to 300 nm showed that collected intensity curves are identical in any area of the sample.

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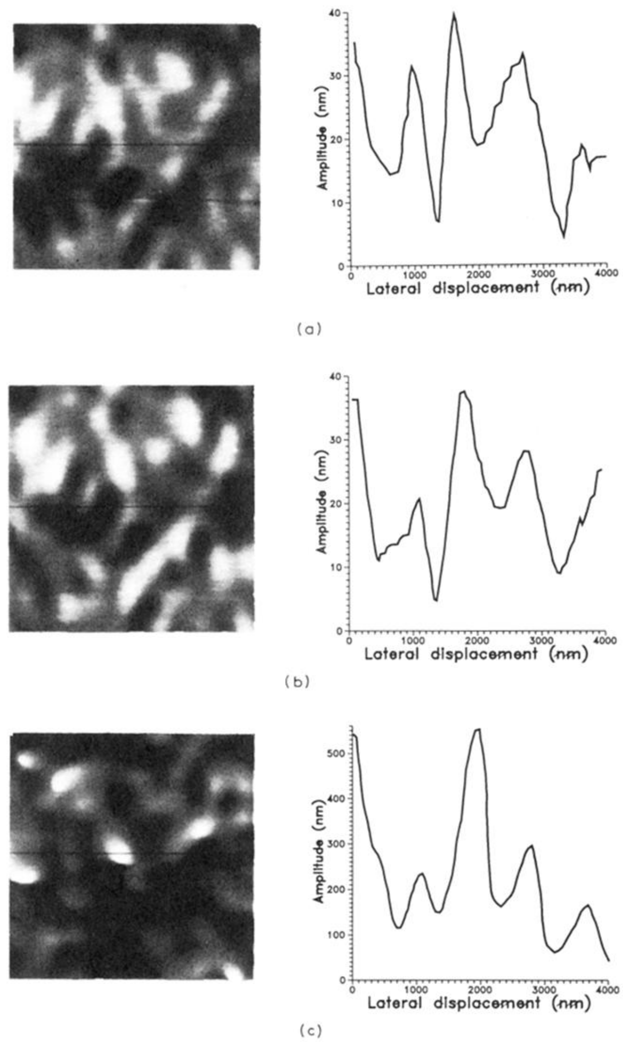

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