Surface-plasmon microscopy with grating couplers

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Received 11 March 1993; revised manuscript received 27 May 1993

A novel surface plasmon microscopical technique with grating couplers is demonstrated. The rotation of the grating structure with respect to the plane of incidence provides a new contrast mechanism for the imaging of thin film samples of different optical thicknesses.

1. Introduction

Recently, surface plasmon microscopy (SPM) was introduced as a novel highly sensitive analytical tool for the optical characterization of interfaces and ultrathin films [1–3]. We could show that a thickness sensitivity of only a few tenths of a nanometer at a lateral resolution of a few μm can be achieved [4,5]. All examples given in the literature so far used a prism as surface plasmon (plasmon surface polaron or PSP for short) coupler [6]. The gratings used in these studies were prepared with a periodicity of A=0.5 μm [9].

In this communication we report on the use of the alternate coupling scheme for PSP excitation, namely grating structures, in a microscopic set-up. We demonstrate, in particular, that the rotation of the grating structure with respect to the plane of incidence provides a new contrast mechanism for the imaging of thin film samples of different optical thicknesses. For a quantitative analysis of the microscopic images this scheme requires only a rotation of the sample at a fixed angle of incidence of the exciting laser beam. This should facilitate the practical use of this microscopic technique.

2. Experimental

All experiments were performed with a linearly polarized HeNe laser at a wavelength of λ=632.8 nm (P=5 mW). A schematic of the set-up is shown in fig. 1. The gratings used in these studies were fabricated with a periodicity of A=0.5 μm [9]. The photoresist gratings were coated with ca. 200 nm Ag by evaporation in a vacuum chamber. Thin film test coatings were prepared by depositing monomolecular layers of poly[(γ-methyl-L-glutamate)-co-(γ-octadecyl-L-glutamate)] (PG, each monolayer is Δ=1.75 mm thick) [10] by the Langmuir-Blodgett-Kuhn (LBK) technique [11]. The mounted gratings could be rotated by an angle θ with respect to the plane of incidence defined by the incident (angle of incidence φ with respect to the sample normal) and the reflected laser beam. At θ=0 the grating vector G=2π/λ was parallel to the photon wave-vector projection kγ=cos φ in the case of 

![Fig. 1. Schematic of the experimental set-up for surface plasmon microscopy with grating couplers. The plane of incidence is defined by the incoming (angle of incidence φ) and the reflected laser beam. The grating can be rotated around an axis parallel to its normal by θ. The polarization of the (linearly polarized) laser beam can follow this rotation.](image-url)
an angular scan the reflected intensity was monitored by a photodiode as a function of $\phi$. For the microscopic mode, the reflected and scattered plasmonic light was Fourier-backconverted by a lens and monitored by a TV (CCD) camera [12].

3. Results

Figure 2 summarizes a series of angular scans (reflectivity $R$ versus angle of incidence $\phi$) taken at various rotation angles $\psi$ as indicated. Two sets of data are shown: the first was taken for the bare Ag grating and shows the well-documented shift of the PSP resonance to higher angles $\phi$ with increasing rotation $\psi$ of the grating [13–15]. In order to compensate for the increasing angular mismatch between the grating vector $G$ and the direction of the initially $p$- (i.e., linearly in the plane of incidence) polarized light the polarizer in the exciting laser beam was also rotated for each $\phi$-scan by $\psi$. This way, all resonance curves are nearly equally deep. At a certain cut-off angle $\psi_0$ no PSP excitation at any angle $\phi$ can be found. If the grating was coated by four layers of PG the expected angular shift of the PSP resonance curve was observed. For $\psi=0$ this finding corresponds to the usual PSP spectroscopical mode for the valuation of the optical thickness within the Fresnel theory for ultra-thin organic coatings [16]. Since we are concerned with a grating constant of $A=0.5 \mu m$, we are operating in the second Brillouin zone where any thin film coating shifts the PSP resonance to smaller angles $\phi$ [17]. With increasing rotational angle $\psi$ this angular $\phi$-shift relative to the bare Ag-curves increases. As a result, the LBK coating allows for PSP excitation even at angles $\psi$ which are already beyond the cut-off value $\psi_0$ for the bare Ag. But also in the coated areas, eventually the characteristic cut-off angle $\psi_0$ is reached.

This PSP resonance behavior can be understood on the basis of the momentum matching scheme given in fig. 3. Shown are quarter circles with radii corresponding to the grating vector $|G|$, and the PSP wavevectors $|k_{sp}^{Ag}|$ and $|k_{sp}^{Ag+4PG}|$, respectively. Now, for a given sample rotation $\psi$, the grating vector $G$ corresponds to the arrow from the origin to the respective circle. The photon wavevector projection as obtained from the corresponding reflectivity scan (cf. fig. 2) adds vectorially parallel to the $x$-direction which is defined by the geometry of the reflectivity measurement. All thus determined values for the resonance momentum matching conditions, indeed, fit to the respective PSP wavevector circles. This picture also explains the different cut-off angles $\psi_0$. Beyond these, the addition of the photon wavevector projection does not reach a point on the respective PSP-circles.

If we plot the reflected intensity at a fixed angle $\phi$ (taken from the angular scans in fig. 2) as a function of $\psi$, we find the following behavior:

![Fig. 2. Series of angular $\phi$-scans of the reflected intensity for a bare Ag-grating (a) and after deposition of four layers of polyglutamate (b) at various sample rotations $\psi$, as indicated. Note the shift of the PSP-resonance to higher $\phi$-values with increasing $\psi.$](image-url)
Fig. 3. Momentum matching scheme for the PSP excitation experiments presented in fig. 2. Given are quarter circles corresponding to the grating vector $|G|$, and the PSP wavevectors $|k_{SP}^0|$ and $|k_{SP}^0 + 4m|$, respectively. By rotation of the grating by $\psi$ and the vector-addition of the photon wavevector projection $k_{\text{photon}}$ (parallel to the x-axis) a point on the surface plasmon wavevector circle can be reached which leads to the resonant excitation of PSP waves. Above a certain cut-off angle $\psi_c$ this is not possible anymore.

Fig. 4. Plot of the reflected intensities at a fixed angle of incidence, $\phi = 35^\circ$, as derived from $\phi$-scans at fixed angles of rotation $\psi$. The different angular positions for the PSP resonance in bare Ag-gratings (---) and after deposition of four PG LBK-layers (-----) is the basis for the $\psi$-contrast in SPM. Of the various $\psi$-values, we see the striking difference between bare Ag and the polymer coated sample: the $\phi$-contrast of the “classical” PSP-resonance spectroscopy is converted into a $\psi$-contrast at a fixed angle $\phi = 35^\circ$.

Fig. 5. Series of SPM pictures taken from a sample consisting of bare Ag, a narrow stripe coated with two layers of PG and an area coated with four layers. The fixed angle of incidence was $\phi = 35^\circ$, the varied angle of sample rotation $\psi$ is indicated. The bar corresponds to ca. 1 mm.
ngle of incidence. This is documented in fig. 4 for the reflectivity data taken at $\phi = 35^\circ$. The $\psi$-resonance curves are about as narrow as the $\phi$-scans with a comparable shift induced by the LBK-layer. We expect, therefore, a similar contrast obtainable in this novel surface plasmon microscopical mode as compared to the $\phi$-SPM.

This is demonstrated in fig. 5 for a sample which was only partly coated by four layers of polyglutamate. Shown is a series of pictures taken in the usual microscopic configuration, however, at the fixed angle of incidence $\phi = 35^\circ$ but at various grating rotations $\psi$. At $\psi = 53^\circ$ the region coated by four PG layers is at resonance and hence appears dark in reflection as it was inferred from the data in fig. 4. Rotation of the grating to $\psi = 54^\circ$ detunes this sample area out of resonance whereas the transition region to the bare Ag which is coated with two PG layers now appears dark. At $\psi = 54.3^\circ$ this sample area is fully at resonance with excellent intensity contrast to the neighboring sample areas. Note that a double layer PG is only $d = 3.5$ nm thick [10]. Increasing $\psi$ further eventually tunes the uncoated Ag-area in resonance at $\psi = 56^\circ$ (cf. also fig. 4) until at still higher angles the whole sample is bright.

4. Conclusions

We have demonstrated a novel surface plasmon microscopical technique with grating couplers that allow for a transformation of the $\phi$-contrast based on the sensitivity of the resonance coupling angle for different coating thicknesses into a $\psi$-contrast, i.e. reflecting differences for different sample rotations. For the practical use of SPM this should give a substantial advantage because it replaces the difficult angular scans by much simpler sample rotations. E.g., any commercial light microscopy can easily be equipped with a rotary table. Other advantages of using grating couplers instead of prisms are currently elaborated in this laboratory.

References