# Unexpected polarization behavior at the aperture of hollow-pyramid near-field probes

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Polarization in the proximity zone beyond the illuminated aperture of a near-field optical microscope is determined by means of a thin dichroic layer of fluorescent molecules used as a near-field polarization analyzer. Near-field probes of the hollow-pyramid type, with a metal coating and about 100 nm apertures, are used. Surprisingly, it is found that the input polarization is always maintained in the near field, independently of the aperture geometry, in spite of the behavior of the transmitted far field, which may result either isotropic or strongly dichroic depending on the ellipticity of the aperture. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2137891]

Near-field scanning optical microscopy (NSOM) allows one to beat the diffraction limit in optical imaging by illuminating the sample through a subwavelength aperture brought at a distance much smaller than the wavelength [near field (NF)].[1,2] The most common practical implementation of the aperture is a tapered optical fiber, coated with a metal layer, which leaves a small hole at the apex; these probes enable a spatial resolution ranging from 50 to 100 nm, limited by the vanishing transmission for smaller aperture size. Recently, a novel class of NSOM probes has been introduced, based on a silicon or silicon nitride cantilever, similar to the ones used in atomic force microscopy (AFM), with a hollow pyramid tip.[3,4] The pyramid is metal coated, usually by aluminum or gold, leaving a hole at its apex, with a diameter ranging from 40 to 200 nm. These cantilevered tips offer several advantages[5] compared to tapered fibers, namely: (i) The larger taper angle produces higher power throughputs; (ii) the lower absorption allows one to couple higher average power before reaching the onset of thermal damage of the metal coating; and (iii) tip-sample distance stabilization methods used in AFM, such as the tapping mode, can be employed, thus ensuring longer probe lifetimes as compared to the standard shear-force method used with fiber probes.

The knowledge and control of the polarization state of the light in the NF is important for many applications of NSOM, e.g., polarization-sensitive mapping of the optical properties,[6] magneto-optical Kerr effect detection,[7,8] and optical nanowriting on photosensitive polymers.[9] Polarization properties of the NF have been theoretically investigated using the multiple-multipole method for a simplified case of a two-dimensional-NSOM, modeling the NSOM aperture as an infinite slit.[10] The optical behavior of the slit is found to be dramatically different for input polarizations parallel or orthogonal to the slit: Orthogonal polarization has a much larger transmitted power and a much lower degree of confinement than parallel polarization. Moreover, a nonzero longitudinal component of the electric NF is present.[10] This model can also be experimentally extended to the three-dimensional case[11] by defining a polarization plane that contains the tip axis and the polarization direction of the transmitted far-field (FF) wave, and by separately considering the radiation propagating along such a plane and orthogonal to it. However, the calculations assume a perfect taper structure, whereas imperfections of the real taper may play a significant role and lead to deviations from this theoretical framework. Moreover, such calculations are meaningful once the polarization of the radiation impinging on the taper is exactly known, which is hardly realized in practice with tapered fiber probes due to depolarization effects in the fiber. The use of hollow-pyramid cantilevered NSOM tips eliminates the need for an optical fiber, allowing a complete control of the polarization state incident on the taper. Additionally, the simplified structure of such hollow pyramids is promising toward a better theoretical understanding of the aperture behavior in the NF and indicates that the input light polarization should not be appreciably modified by the propagation inside the probe.

Since experimental access to the polarization state of NF light is not straightforward, it is common practice to characterize it by looking at the FF light.[12,13] However, a technique for probing the in-plane component of the electric NF was recently introduced.[14] It uses specially developed samples in which a fluorescent molecule layer with thickness comparable to that of the proximity zone is produced inside a host polymer film. The molecular dipoles are oriented along the sample surface by mechanical drawing, producing a strongly anisotropic absorption.[15] By recording the fluorescence excited by the local electric NF for different orientations of the film dichroic axis, the in-plane component of the linear po-

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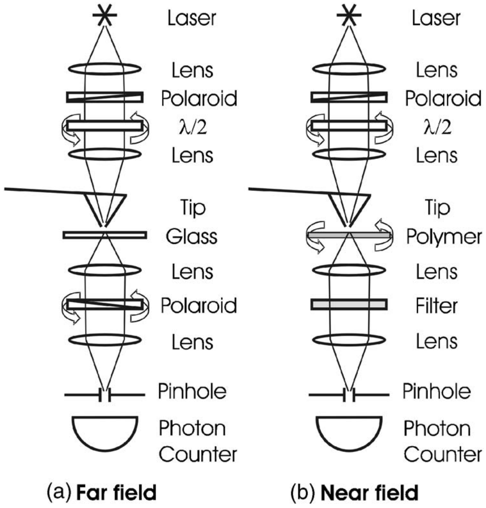


FIG. 1. Sketch of the experimental setups used for the characterization of FF (a) and NF (b) polarization properties.

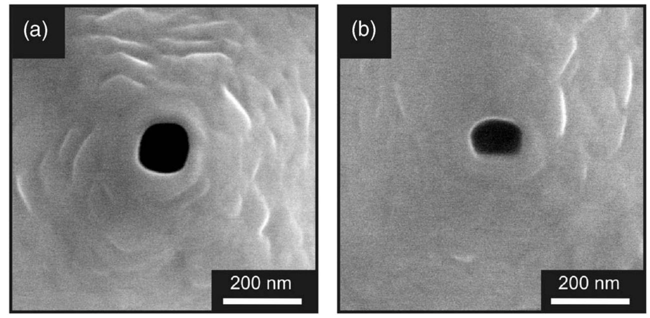


FIG. 2. Scanning electron microscopy images of the apex of two different tips, with circular (a) or elliptic (b) aperture.

larization at the aperture can be determined. The thin dichroic fluorescent film acts therefore as an effective NF polarization analyzer. Tapered fibers have already been characterized with this procedure[14] and polarization pinning is found in about one-half of the probes; however, it is not possible to say whether this effect is due to the birefringence of the fiber or to taper imperfections.

In this work, we perform an experimental study of the NF polarization of hollow-pyramid NSOM probes and compare it to the usual FF characterization. We find that the input polarization is always maintained in the NF, independently of the aperture geometry, despite the behavior of the transmitted FF, which may be either isotropic or strongly dichroic (pinning) depending on the ellipticity of the aperture. These results indicate the hollow pyramid as the most suitable probe for polarization-sensitive NF experiments nowadays and point out the need for a true NF characterization of the polarization state.

For our measurements, we used a commercially available NSOM (AlphaSNOM, WITec GmbH)[16] which we modified in order to accommodate all the optical elements needed to work with polarized light. The instrument employs hollow-pyramid probes supported on a flexible cantilever that acts both as an optical screen and as a force sensor, as customary in AFM; tip-sample distance control is performed in constant-force contact mode. The pyramid is coated with aluminum, thick enough to prevent any stray light from the pyramid sides, and presents at the apex an aperture with a diameter of about 100 nm. The excitation light (532 nm, 20 mW maximum power) is focused onto the NSOM probe. The propagating light, collected by a 0.8 NA objective, is sent through a pinhole, in a confocal collection geometry to improve stray light rejection, onto a photon counting photomultiplier.

The experimental setups used to characterize the polarization properties in the FF and NF are shown in Figs. 1(a) and 1(b), respectively. For the FF characterization, the tip is placed in contact with a glass substrate, to allow height stabilization without affecting the polarization of the transmitted light. Polarization analysis is carried out by a standard rotatable polaroid filter placed in front of the detector. In this way, the polarization direction of the propagating light can be determined for each polarization state of the light incident on the aperture, as selected by a half-wavelength plate. For the NF characterization, we use a dichroic thin film, prepared by casting and by subsequent mechanical stretching.[15] The sample is made from a xylene solution of ultrahigh molecular weight polyethylene and a terthiophene derivative, which is resonant with the 532 nm excitation and fluoresces with high efficiency. In this way, the chromophore molecules are mostly confined in about 250 nm thickness at the upper film surface[14] and the stretching aligns the molecules, giving rise to a strong absorption dichroism. We checked that the chromophore does not suffer from photodegradation. The tip is then contacted to the polymer, and the fluorescence signal, coming mostly from the NF region, is selected by means of a suitable long-pass filter (Schott OG570) rejecting the excitation light. The polymer film can be rotated and thus acts as a NF polarization analyzer. For each incident polarization direction, we measured the fluorescence signal from the sample for different orientations of its dichroic axis, thus obtaining a polar plot, from which we could extract both polarization direction and ratio in the NF.[14] In particular, since the strain axis of the film is perpendicular to the tip axis, only the in-plane polarization state of the NF is probed. It is worth noting that a standard polaroid filter, even when in contact with the tip, would not be suitable to the same purpose, due to its thickness (much larger then the extension of the proximity zone).

Ten tips have been characterized in total for this experimental work. For each tip, we first measured the total transmitted FF light as a function of the polarization state incident on the aperture. In doing this, we found peculiar differences among the tips, which were related to the aperture geometry by means of scanning electron microscopy imaging (Fig. 2). While the tips with an almost circular, highly symmetric aperture [Fig. 2(a)] show nearly the same transmission coefficient for every incoming polarization direction, the tips with a more elliptical hole [Fig. 2(b)] are highly dichroic, with a preferential direction of transmission for light polarized along the minor axis of the ellipse. The ratio between the transmission coefficients along the two main axes for the dichroic tips was generally between 10 and 20. This is a well-established FF behavior, as already pointed out in previously published theoretical[10] and experimental[11,13] studies. We will refer to such different kinds of tips as "isotropic" or "dichroic", respectively.

Figure 3(a) shows the FF polarization behavior for an isotropic tip, plotting the polarization angle $\theta_{\text{out}}$ of the propagating light as a function of the polarization angle $\theta_{\text{in}}$ of the incident light. As expected, the polarization is very well pre-

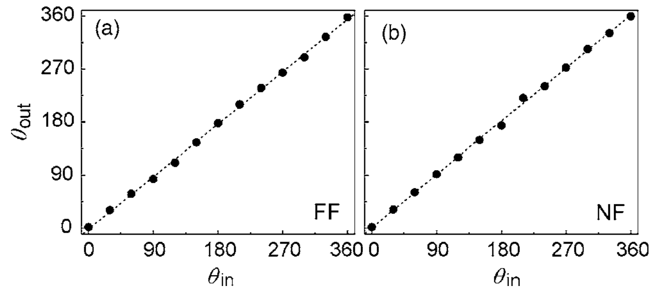


FIG. 3. Plot of the FF (a) and NF (b) polarization direction $\theta_{out}$ as a function of incident polarization direction $\theta_{in}$ for an isotropic tip. $\theta=0°$ refers to one of the pyramid base sides.

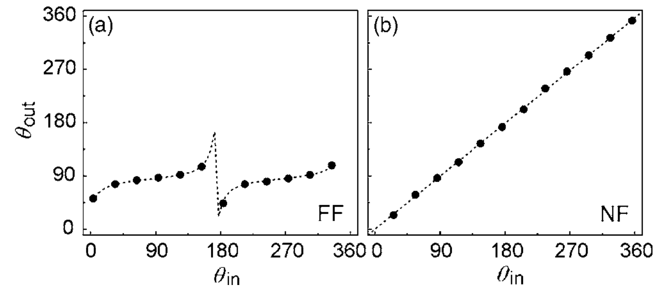


FIG. 4. Plot of the FF (a) and NF (b) polarization direction $\theta_{out}$ as a function of incident polarization direction $\theta_{in}$ for an isotropic tip. $\theta=0°$ refers to the long axis of the ellipse.

served. The result of the NF characterization for the same tip is shown in Fig. 3(b): It confirms the FF behavior, showing that the input polarization is translated also in the NF with high fidelity.

Figure 4(a) shows the FF polarization behavior for a dichroic tip, indicating that it strongly pins the polarization along its preferential direction of transmission. The data can be fitted with the expression

$$\tan \theta_{out} = \sqrt{\frac{T_x}{T_y}} \tan \theta_{in},$$

where $T_x$ and $T_y$ are the transmission coefficients for light polarized along the short and long axis of the aperture, respectively. The NF polarization behavior of the dichroic tip is shown in Fig. 4(b): We find that, at variance with the FF result, the tip closely maintains the incident polarization in the NF. This happens with all the dichroic tips we characterized. To verify this surprising result, we repeated the characterization with the NF setup by increasing the tip/sample distance by 500 nm, therefore going out of contact in order to mimic the FF behavior. Under these conditions, we recovered the polarization pinning behavior also from the fluorescence signal, thus excluding any artifact from the polymeric sample in the NF measurements. From the fitting of our NF polar plots we could also extract the value of the polarization ratio in the NF, which is found to vary between 20 and 200 for different tips.

Our unexpected experimental results point out the importance of an effective probe for the NF polarization out of a subwavelength aperture, and seem to require a reconsideration of previous experimental characterizations performed in the literature, where only the FF propagating light was analyzed. They also may provide hints on the NF generation mechanisms at NSOM apertures, suggesting that the source of the local NF is not the whole aperture, as in the usual diffraction-theory approach where the geometric shape of the aperture plays a considerable role. Rather, we tentatively suggest that the illuminated area of the sample mostly sees the evanescent NF coming from the nearest zone of the aperture, which locally behaves like an isotropic center and scatters the incident light without affecting its polarization direction. However, further studies are necessary to clarify this issue.

It is worth noting that the polarization state of inhomogeneous waves, characterized by a complex wave vector and therefore by an evanescent component, needs to be correctly defined. Such a definition is still being discussed,[17] but for practical purposes we choose as the polarization direction the one of the local electric field, averaged within the interaction volume. In particular, our experimental characterization deals only with the in-plane polarization of the NF, since we are not sensitive to the longitudinal NF component. The discrimination of longitudinal fields would be a further task to be undertaken by means of differently structured samples, e.g., composed by vertically oriented fluorescent dipoles.

In conclusion, we have characterized the NF polarization properties of hollow-pyramid NSOM probes, demonstrating that, irrespective of the geometrical asymmetry of the aperture, they are able to translate the incident polarization into the NF. This result points out that such probes are the most indicated for polarization-sensitive experiments in the NF. In addition, it emphasizes that the polarization state in the NF cannot be characterized and optimized simply by a FF measurement, and that a method able to extract the NF contribution from the total propagating light is always required.

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