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Near-field optical microscopy of plasmonic effects in anisotropic metamaterials

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ABSTRACT

Anisotropic plasmonic metamaterials are studied by means of scanning near-field optical dynamic polarimetry. Absolute measurements of near-field linear dichroism are performed for an array of plasmonic nanowires by measuring the linear dichroism value at the edge of the sample. The data are processed according to the Jones calculus. The results indicate nonuniform distribution of the linear dichroism value in the subwavelength vicinity of the sample with the mean value of -0.21 ± 0.03 which is in agreement with its far-field value of -0.20 ± 0.02.

1. Introduction

Polarization properties of anisotropic nanostructures have been an intense field of research for a long time. Wire-grid polarizers [1], form-birefringent waveplates [2] and tailored beamsplitters [3] have proven to be competitive against bulk-media optical elements. Anisotropic nanomaterials which optical response is defined by resonances of surface plasmon polaritons—so-called plasmonic metamaterials—possess high angular and frequency dispersion of birefringence and dichroism [4]. This implies high tunability of optical anisotropy allowing one to perform an arbitrary polarization transformation [5].

Scanning near-field optical microscopy (SNOM) [6] is a method which allows one to study optical properties of plasmon-active media by direct observation of the local plasmonic field intensity with spatial resolution below the diffraction limit. The possibility of studying polarization properties by means of SNOM gives the insight of how light polarization could be controlled by plasmonic nanostructures [7,8]. Sensitivity of maps of near-field optical intensity distribution to polarization state of the incident beam lies in the concept of spatial control of surface plasmons with polarization [9]. Although polarization-sensitive diagnostics of anisotropic nanostructures has been performed in various experimental configurations, there is a lack of metrological measurements in the field of near-field optical polarimetry of plasmonic nanostructures due to interpretation ambiguities occurring in the near-field microscopy results.

In this work we perform a novel scheme for absolute measurements of the near-field analog of linear dichroism (LD) effect in the subwavelength vicinity of a plasmonic nanowire array. It is based on simultaneous near-field dynamic polarimetry of the sample and its substrate combined with the results given by Jones calculus. The distribution of LD value measured with the subwavelength resolution is shown to be nonuniform with its mean value coinciding with the LD value measured in the far field.

2. Methods

Samples of plasmonic nanowire arrays were prepared using electron beam lithography with positive resist and lift-off process out of a thin golden film. The dimensions of the nanowires were extracted from atomic force microscope images and are as follows: the width of each wire is 80 ± 5 nm, the height of each wire is 40 ± 5 nm, the length of each wire is 30.0 ± 0.1 μm and the period of the grating is 300 ± 10 nm. The total nanostructured area of 30×30 μm^2 is surrounded by clear fused silica substrate. The transmission spectra of the samples were acquired using a micro-spectroscopy setup described in our previous work [4].

The polarization properties of the samples in the near-field regime were performed using a dynamic polarimetry setup combined with SNOM shown in Fig. 1. The setup is analogous to one described in our previous paper [10] and will be briefly described here. It relies on modulating the state of polarization of the light impinging on the sample with a photoelastic modulator (PEM) at a frequency *f*. The electrical signal from the photodetector is Fourier-decomposed to DC component and 2*f* harmonic amplitude; two raster images are built from the values of these terms in each

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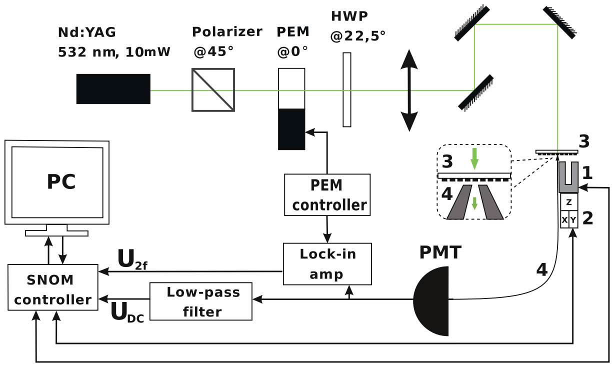

Fig. 1. Schematic of the experimental setup for scanning near-field measurements of the linear dichroism effect. PEM is a photoelastic modulator, HWP is a half-wave plate, PMT is a photomultiplier tube; 1 is a tuning fork which is mounted on a *XYZ* piezostage 2; the sample 3 is scanned by a SNOM probe 4 in a constant-height regime at a distance of about 10 nm from the surface.

lateral point of the sample. From these images the map of the near-field value of LD is extracted as [11]:

$$D_{nf} = C\frac{U_{2f}}{U_{DC}}, \tag{1}$$

where U_{2f} and U_{DC} are the values of DC and $2f$ components of the electrical photoinduced bias at the detector and C is a normalization constant defined by measuring D_{nf} for a “perfect” polarizer like Glan-Taylor prism.

3. Compensation for the linear dichroism of the probe

The data on linear dichroism of the plasmonic nanostructures obtained before [10] does not provide one with the proper absolute values of LD of the sample. The LD values are obscured by the intrinsic dichroism of the near-field probe which shape could be anisotropic as well. Here we present a method of compensating for the spurious linear dichroism value originating from the probe in an approximation of the probe as a polarizer with its transmission axis oriented at an angle θ with respect to the vertical axis. In this case the probe is described by the following Jones matrix:

$$P_\theta = R^{-1}(\theta)P_0R(\theta) = R^{-1}(\theta)\begin{pmatrix} a_0 & 0 \\ 0 & b_0 \end{pmatrix}R(\theta), \tag{2}$$

where P_θ is the matrix of the probe in the laboratory frame, P_0 is the matrix of the probe in its eigenvector basis and $R(\theta)$ is the rotation matrix. With no sample between the incident beam and the probe the intensity of light at the PMT is expressed as follows (see Appendix A):

$$I(t) = 2\left(a_0^2 + b_0^2\right) + 4J_2(A)\cos(2\theta)\left(b_0^2 - a_0^2\right)\sin(4\pi ft), \tag{3}$$

where $J_n(A)$ are Bessel functions of the first kind and A is the maximum phase delay brought by PEM; its value is set to $A \simeq 2.405$ so that $J_0(A) = 0$. In Eq. (3) only DC and $2f$ components of the Fourier expansion are included. Define ξ variable as:

$$\xi = \frac{U_{2f}}{2J_2(A)U_{DC}} = \frac{\cos(2\theta)\left(b_0^2 - a_0^2\right)}{a_0^2 + b_0^2}. \tag{4}$$

The sample is described by the following Jones matrix:

$$J_{sam} = \begin{pmatrix} ae^{i\phi} & 0 \\ 0 & b \end{pmatrix}. \tag{5}$$

The intrinsic dichroism of the sample is equal to:

$$D_{nf} = \frac{b^2 - a^2}{b^2 + a^2}, \tag{6}$$

which is equivalent to the expression Eq. (1) if no other dichroic elements are placed before or after the sample. Note that this definition of LD differs from the canonical one; it is defined for transmission coefficients rather than for absorption coefficients and can take values from -1 to 1 rather than from 0 to 1. With the sample between the incident beam and the probe the ξ' value is expressed as a function $\xi'(a, b, a_0, b_0, \theta)$ given by Eq. (A.5). Using the expressions for D_{nf}, ξ and ξ' one obtains the expression for the LD value in the following form:

$$D_{nf} = \frac{\xi - \xi'}{\xi\xi' - 1}. \tag{7}$$

We recall that values ξ and ξ' are the ratios of U_{2f} and U_{DC} measured with and without the sample installed into the setup, correspondingly. Thus the resulting D_{nf} value is an absolute LD value free from the contribution of the near-field optical probe.

4. Results and discussion

The measurements of the near-field LD are performed on the samples of nanowire arrays excited with a monochromatic laser source with wavelength $\lambda = 532$ nm. The wavelength of the source is situated in the spectral proximity of the local plasmon resonance indicated as a dip in the transmission spectra in Fig. 2 taken for the polarization of the incident light oriented perpendicularly to the wires. The spectra are shown for polarization of incident light along (gray line) and perpendicular to (black line) the wires.

A LD map was measured according to the aforementioned procedure for the edge of the sample as shown in Fig. 3; the mean value of the $U_{2f}/(2J_2(A)U_{DC})$ above the substrate area with no sample on it was used for ξ' value. The map demonstrates a well resolved array of stripes which is free from speckle structure of the illuminating field. The period of the stripes coincides with the period of the nanowire grating. Bright stripes (low absolute values) are situated in the site of the wires and dark ones (high absolute values)

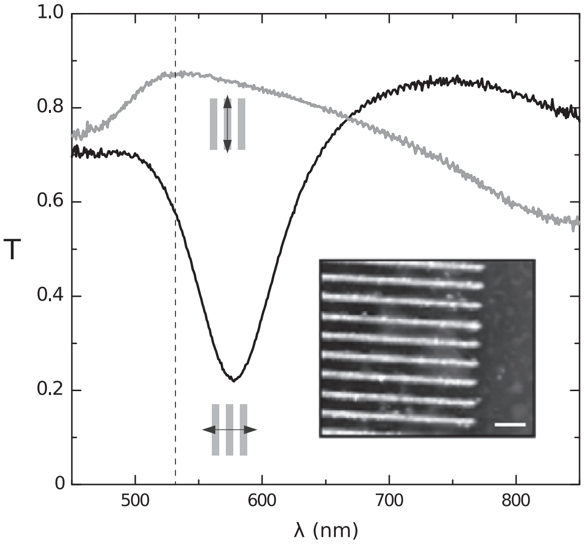

Fig. 2. Transmission spectra of the nanowire array sample for polarization along (gray line) and perpendicular to (black line) the wires. The dashed line denotes the wavelength used in SNOM experiments. The inset shows the topography of the sample obtained in an atomic force microscope. The scale bar equals 500 nm.

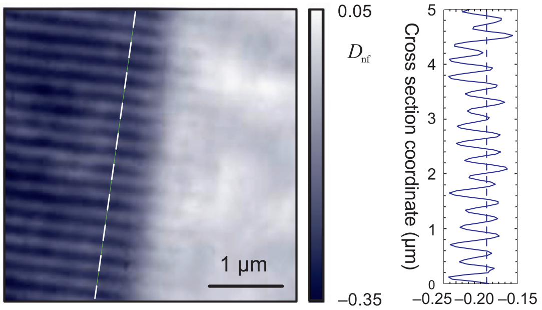

Fig. 3. (Left) The map of linear dichroism of the sample near one of its boundaries. (Right) The section of the LD map with the dashed line on the left panel. The dashed line on the right denotes the far-field value of LD.

correspond to the spaces between the wires which is proved by comparing the shear-force topography images and SNOM images [10].

The absolute value of LD is oscillating as a function of coordinate between the values of from approximately −0.25 to approximately −0.18 with the mean value of −0.21 ± 0.03 which is in a good agreement with the measured far-field value of LD −0.20 ± 0.02. The provided data give possible grounds to the validity of the method used to determine the absolute values of the near-field LD.

5. Conclusions

In conclusion, absolute measurements of the linear dichroism of a plasmonic nanowire array were carried out using the scanning near-field dynamic polarimetry method. The data processed according to Jones calculus indicate nonuniform periodic distribution of the linear dichroism in the subwavelength proximity of the sample surface. The near-field linear dichroism value is varying from −0.25 to −0.18 and its mean value is −0.21 ± 0.03 which corresponds to the value of linear dichroism obtained in the far field −0.20 ± 0.02. The established technique and obtained data are a step towards understanding the problem of polarization in optical near fields.

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Appendix A. Evaluation of Eq. (7)

The vector of the polarization state of the incident light is composed by three optical elements, namely a polarizer, a PEM and a half-wave plate, transforming the initial horizontally polarized state of light coming out from the laser. It is written down as a set of Jones matrices acting on the horizontally polarized wave:

$$\mathbf{P}_{in} = \begin{pmatrix} -1 & -1 \\ -1 & 1 \end{pmatrix} \begin{pmatrix} 1 & 0 \\ 0 & e^{i\delta} \end{pmatrix} \begin{pmatrix} -1 & -1 \\ -1 & 1 \end{pmatrix} \begin{pmatrix} 1 \\ 0 \end{pmatrix} = \begin{pmatrix} 1 + e^{i\delta} \\ 1 - e^{i\delta} \end{pmatrix}, \tag{A.1}$$

where $\delta = A\sin(2\pi f\,t)$. The SNOM probe is described by a matrix in the form of Eq. (2). Total intensity of the light coming to the PMT after passing the setup without the sample installed is expressed by the following Jones vector:

$$\mathbf{P}_{out} = \begin{pmatrix} \cos\theta\sin\theta(a_0 - b_0)(e^{i\delta} + 1) + (b_0\sin^2\theta + a_0\cos^2\theta)(1 - e^{i\delta}) \\ (a_0\sin^2\theta + b_0\cos^2\theta)(e^{i\delta} + 1) + \cos\theta\sin\theta(a_0 - b_0)(1 - e^{i\delta}) \end{pmatrix}. \tag{A.2}$$

Its intensity is calculated as $I = |\mathbf{P}_{out,1}|^2 + |\mathbf{P}_{out,2}|^2$. Then we simplify the expression for I' using the Fourier expansion of the functions:

$$\begin{aligned} \sin(A\sin(x)) &= 2\sum_{i=1} J_{2i-1}(A)\sin[(2i-1)x], \\ \cos(A\sin(x)) &= J_0(A) + 2\sum_{i=1} J_{2i}(A)\sin(2ix). \end{aligned} \tag{A.3}$$

After disregarding all the terms except the time-independent one, which is measured after a low-pass filter, and one oscillating at the $2f$ frequency, which is measured with a lock-in amplifier, the expression for the electromagnetic field intensity takes the form of Eq. (3). The value of ξ given by Eq. (4) is measured.

Then, the same values are measured with the sample installed into the setup. The sample is described by the matrix in the form of Eq. (5). In this case the polarization state of light at the PMT is described with the following vector:

$$\mathbf{P}'_{out} = \begin{pmatrix} b\cos\theta\sin\theta(a_0 - b_0)(e^{i\delta} + 1) + ae^{i\phi}(b_0\sin^2\theta + a_0\cos^2\theta)(1 - e^{i\delta}) \\ b(a_0\sin^2\theta + b_0\cos^2\theta)(e^{i\delta} + 1) + ae^{i\phi}\cos\theta\sin\theta(a_0 - b_0)(1 - e^{i\delta}) \end{pmatrix} \tag{A.4}$$

The intensity is evaluated as:

$$\begin{aligned} I' = {} & 2J_2(A) \\ & \times \sin(4\pi ft)\Big(\cos(2\theta)(b^2 + a^2)\big(b_0^2 - a_0^2\big) + \big(b_0^2 + a_0^2\big)(b^2 - a^2)\Big) \\ & + \cos(2\theta)\big(b_0^2b^2 - a_0^2b^2 - b_0^2a^2 + a_0^2a^2\big) + b_0^2b^2 + a_0^2b^2 \\ & + b_0^2a^2 + a_0^2a^2, \end{aligned} \tag{A.5}$$

and ξ' value is expressed as follows:

$$\xi' = \frac{\cos 2\theta(b^2 + a^2)\big(b_0^2 - a_0^2\big) + \big(b_0^2 + a_0^2\big)(b^2 - a^2)}{\cos 2\theta\big(b_0^2b^2 - a_0^2b^2 - b_0^2a^2 + a_0^2a^2\big) + b_0^2b^2 + a_0^2b^2 + b_0^2a^2 + a_0^2a^2}. \tag{A.6}$$

Using explicit expressions of ξ, ξ' and D_{nf} as functions of unknowns a/b, a_0/b_0 and θ Eq. (7) is obtained. Note that θ, although not being intentionally excluded from the system, is not present in the final expression for D_{nf}.

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