Polarization tomography of metallic nanohole arrays

E. Altwiescher, C. Genet, M.P. van Exter, J.P. Woerdman
Huygens Laboratory, Leiden University, P.O. Box 9504, 2300 RA Leiden, The Netherlands

P.F.A. Alkemade
Faculty of Applied Science, Delft University of Technology, Rotterdamseweg 137, 2628 AL Delft, The Netherlands

A. van Zuuk, E.W.J.M. van der Drift
DIMES, Delft University of Technology, PO Box 5053, 2600 GB Delft, The Netherlands
(Dated: November 6, 2018)

We report polarization tomography experiments on metallic nanohole arrays with square and hexagonal arrays, i.e. arrays that, for symmetry reasons, cannot modify the SOP for plane-wave illumination at normal incidence. As we will show, depolarization occurs when two (quite common) conditions are fulfilled: (i) the response of the array is nonlocal due to SP propagation, and (ii) the input beam is not a plane wave (but e.g. a Gaussian beam, with a finite numerical aperture (NA)).

In general, depolarization occurs when an optical system acts non-uniformly on polarization within the (spatial or temporal) bandwidth of the incident wave, thereby coupling polarization to other degrees of freedom. The two most widely used formalisms to describe the polarization properties of a linear optical system are due to Jones and Mueller. In both cases, the input and output SOPs are represented by column vectors, which are connected by either a 2 × 2 Jones matrix or a 4 × 4 Mueller matrix, describing the action of the optical system. The Jones formalism is applicable only to situations in which the light is temporally coherent and spatially-uniform polarized. The Mueller formalism deals with the Stokes parameters, which represent (spatial or time) averages of the polarization properties of the light, and as such, is also capable of handling partially polarized and incoherent waves. Experimentally, a study of depolarization requires therefore a measurement of the Mueller matrix by a tomographic method. We report here such polarization tomography experiments on nanohole arrays and interpret the results in the context of SP propagation.

We start by recapitulating the essence of our theoretical model. The input and output optical fields of the array are related via a non-local linear response as $\vec{E}_{\text{out}}(\vec{r}, \omega) = \int t(\vec{r} - \vec{r}', \omega) \vec{E}_{\text{in}}(\vec{r}', \omega) \, d^3 \vec{r}'$. In the far-field, or Fourier domain, this is equivalent to $\vec{E}_{\text{out}}(\vec{k}_t, \omega) = t(\vec{k}_t, \omega) \vec{E}_{\text{in}}(\vec{k}_t, \omega)$, where $\vec{k}_t$ is the transverse wavevec-
tor component; the output $\vec{k}_t$ is equal to the input $\vec{k}_t$ for the zeroth-order transmission. If the non-local response depends on polarization, the elements of the $2 \times 2$ transmission tensor $t(\vec{k}_t, \omega)$ exhibit a different angular dependence and the output field $\vec{E}_{out}(\vec{k}_t, \omega)$ can have a spatially-dependent polarization even for a polarization-pure input field. This necessitates a Stokes vector description, which considers only spatially-integrated intensities. The four components of the Stokes vector are $S_0 = I_{00} + I_{90\circ} = I_{45\circ} + I_{135\circ} = I_\sigma + I_\sigma\overline{\sigma}$, and $S_1 = I_{00} - I_{90\circ}$, $S_2 = I_{45\circ} - I_{135\circ}$ and $S_3 = I_\sigma - I_\sigma\overline{\sigma}$, with $I_i$ the intensity of the polarization component denoted by the subscript $i$ [11]. After spatial integration, the transmission process can be captured in a simple relation $S_{out} = MS_{in}$, which relates the input and output Stokes vectors through the $4 \times 4$ Mueller matrix $M$. For ideal square and hexagonal arrays the Mueller matrix has been predicted to be diagonal (no mixing of Stokes parameters) [11]. For hexagonal arrays, the additional symmetry relation $M_{11} = M_{22}$ holds.

The magnitudes of the diagonal elements $M_{00}$, $M_{11}$, $M_{22}$ and $M_{33}$ depend on the product of the SP propagation length and the wave vector spread of the input beam, $\ell_{SP}\Delta k_t$. A full theoretical description thereof would require a microscopic model; however, from physical considerations it can be seen that an appreciable deviation of $M_{ii}/M_{00}$ from 1 requires $\ell_{SP}\Delta k_t \gtrsim 1$. In any case, there will be no depolarization if either there are no propagating waves ($\ell_{SP} = 0$) or there is plane-wave illumination ($\Delta k_t = 0$); both propagation and wave vector spread are necessary.

Figure 1 shows the experimental setup used to measure the Mueller matrices. A linearly polarized Titanium:Sapphire laser at a wavelength of 810 nm, being the approximate resonance wavelength of the hole arrays (cf. Fig. 2), illuminates the input lens ($L$ or $L'$) of a symmetric telescope. After transmission through a hole array, positioned at the focus of the telescope, the light is imaged onto a photodiode. The SOP of the incident light is set by a rotatable quarter-wave or half-wave plate in front of the first lens. The Stokes parameters at the output are measured with a rotatable quarter-wave plate and polarizer positioned in front of the photodiode.

We have characterized the arrays with two types of illumination. Almost plane-wave illumination, with a numerical aperture of $NA = 0.01$ ($\Delta k_t \approx 0.08 \mu m^{-1}$), was provided by focussing the 1 mm diameter laser beam with a lens $L$ of $f = 50$ mm. Focussed illumination (up to $NA \approx 0.15$ or $\Delta k_t \approx 1.2 \mu m^{-1}$) was obtained by focussing the laser beam on a 10 $\mu m$ diameter mode-cleaning pinhole (not shown) to homogeneously illuminate a lens $L'$ ($f = 15$ mm at 40 cm from the pinhole) through a variable aperture $A$. The polarization isotropy of all optical components was checked by measuring the Mueller matrices of both setups in the absence of hole arrays. These matrices were practically equal to the identity matrix, with individual elements deviating by not more than 0.02 (typically 0.008).

Our arrays were fabricated in Au films on glass substrates. We used a square array made with electron-beam lithography, identical to the one used in Ref. [13], with a lattice spacing of 700 nm and a nominal hole diameter of 200 nm, and a hexagonal array made with ion-beam milling, with a lattice spacing of 886 nm and a nominal hole diameter of 200 nm. SEM pictures and transmission spectra of both arrays under almost plane-wave illumination at normal incidence are shown in Fig. 2. Both arrays show a resonance wavelength of 810 nm, which is marked by a dashed vertical line; the polarization experiments
were performed at this wavelength. The resonances at 810 nm correspond to SPs propagating in the $(\pm 1, \pm 1)$ direction at the metal-glass interface for the square array, and the (six-fold degenerate) $(1, 0, 0)$ direction at the metal-air interface for the hexagonal array (the labeling is with respect to the reciprocal lattice vectors). The linewidths are 40 nm and 25 nm, for square and hexagonal arrays, respectively, from which we estimate $\ell_{SP}\approx 2 \mu m$ and $\ell_{SP}\approx 4 \mu m$, respectively\textsuperscript{12}.

Figure 3 shows the dependence of the diagonal elements of the Mueller matrix on the NA of the incident light for both arrays (the plotted elements $M_{11}$, $M_{22}$ and $M_{33}$ are normalized with respect to $M_{00}$). The figure shows that, for the case of almost plane-wave illumination (NA $\approx 0$), the $M_{ii}$ values are close to 1 for both arrays. There is no depolarization, as $\Delta k_t \approx 0$. However, for increasing wavevector spread, or decreasing spot size on the array, the depolarizing effect of the arrays quickly increases. Furthermore, the depolarization is clearly anisotropic, i.e. the amount of depolarization depends on the input SOP. For the hexagonal array the depolarization sets in faster upon increasing NA than for the square array, due to its smaller resonance linewidth.

The effect of the different array symmetries on the curve shapes are prominent. For the square array (Fig. 3a), the observed inequality $M_{22} > M_{11}$ shows that there is less depolarization for an input polarization along either of the array diagonals than for a polarization along the main axes. This observation is consistent with the $(\pm 1, \pm 1)$ propagation directions of the resonantly excited SPs on the metal-glass interface; as SPs are mainly longitudinally polarized, they preserve polarization along their propagation direction. However, the deviation of $M_{22}$ from 1 indicates that the $(\pm 1, \pm 1)$ SPs are not the only SPs involved in the transmission process; other (non-resonant) SPs on both surfaces apparently contribute\textsuperscript{12}. For the hexagonal array (Fig. 3b) the theoretical equality $M_{11} = M_{22}$ also holds quite well. For both arrays, $M_{33}$ is the smallest diagonal element. This shows that depolarization is most dramatic for circularly polarized light, a case that has not been studied before. We note that $M_{33} \approx M_{11} + M_{22} - 1$ for both arrays, as follows from an extension of the symmetry-based theory used here by explicit modelling of SP propagation; this will be discussed elsewhere\textsuperscript{14}.

The full Mueller matrices for both arrays are shown in the Table. They were measured with nearly plane-wave illumination (NA $= 0.01$ or $\ell_{SP}\Delta k_t \approx 0.2 - 0.3 \ll 1$), and with illumination at NA $= 0.15$ ($\ell_{SP}\Delta k_t \approx 2$) for the square and NA $= 0.10$ ($\ell_{SP}\Delta k_t \approx 3$) for the hexagonal array (indicated by the dashed vertical lines in Fig. 3). The diagonal elements (marked with boxes in the Table) conform to the discussion given above. The off-diagonal elements of a perfectly symmetric square or hexagonal array should theoretically be zero. For our square array they are indeed relatively small and do not show any systematics behavior. For the hexagonal array however, these elements are much larger both for plane-wave and focussed illumination; this array apparently does not have perfect hexagonal symmetry. Furthermore, the off-diagonal elements have a clear pattern and similar values in both cases, with the “odd” off-diagonal elements $M_{62}$, $M_{20}$, $M_{13}$ and $M_{31}$ (underlined in the Table) are substantially larger than the others. This pattern was checked to be present also for an intermediate NA of 0.03. The pattern is compatible with a birefringent/dichroic 45° axis, which is apparently due to array errors, such as a spatially variant lattice spacing or ellipticity of the holes. These errors could be created by alignment errors or even intrinsic imperfections in the ion beam optics (astigmatism and deflection errors).

From a general perspective, Mueller tomography can give new insight into the physical mechanisms active in hole arrays. It would be interesting to do Mueller tomography on metal hole arrays in the (sub)millimeter-wave regime, as SPs propagate much farther in this part of the spectrum, which is expected to increase the depolarization. Another area of interest is the connection be-

| Measurement | Mueller matrix $M_{ij}$ |
|-------------|------------------------|
| Square array | $M_{ij}$ |
| NA = 0.01 | \[ \begin{array}{cccc} 1.00 & 0.00 & -0.02 & 0.00 \\ -0.02 & 1.01 & -0.01 & 0.00 \\ 0.00 & 0.00 & 1.01 & -0.01 \\ 0.00 & 0.00 & 0.02 & 0.99 \end{array} \] |
| Hexagonal array | $M_{ij}$ |
| NA = 0.01 | \[ \begin{array}{cccc} 1.00 & 0.01 & -0.02 & 0.01 \\ 0.01 & 0.55 & -0.04 & -0.03 \\ -0.04 & 0.02 & 0.84 & 0.01 \\ -0.01 & 0.01 & 0.01 & 0.41 \end{array} \] |
| Square array | $M_{ij}$ |
| NA = 0.15 | \[ \begin{array}{cccc} 1.00 & 0.06 & -0.14 & 0.00 \\ 0.04 & 1.00 & 0.00 & -0.07 \\ -0.12 & 0.01 & 1.01 & -0.03 \\ 0.00 & 0.06 & 0.02 & 0.97 \end{array} \] |
| Hexagonal array | $M_{ij}$ |
| NA = 0.10 | \[ \begin{array}{cccc} 1.00 & 0.03 & -0.11 & 0.00 \\ 0.02 & 0.78 & 0.00 & -0.08 \\ -0.13 & 0.01 & 0.78 & 0.00 \\ 0.00 & 0.06 & 0.02 & 0.51 \end{array} \] |
between classical polarization properties and entanglement degradation\cite{13, 14}. Our work also shows that polarization tomography provides for sensitive diagnostics of array symmetry imperfections.

Finally, one may wonder whether depolarization in optics is reversible or irreversible. In principle, depolarization is reversible, as it is always due to some form of averaging over spatial (or temporal) degrees of freedom\cite{13}; there is no infinite bath that acts as an “information sink”. In the present experiment, “repolarization” would require an element that modifies the array output polarization in a spatially-dependent way; such an element could in principle be constructed based upon a spatial light modulator\cite{16}, provided that it has a sufficient number of degrees of freedom (pixels) available. So, in the end, the difference between reversibility and irreversibility is not absolute but gradual; it depends on the number of degrees of freedom that can be managed in a practical case (The same statement holds of course in statistical mechanics).

In conclusion, we have demonstrated surprising consequences of SP propagation for the polarization behavior of nanohole arrays. The non-locality of the array response forms an essential ingredient of the physics of these intriguing devices.

Acknowledgements

This work has been supported by the Stichting voor Fundamenteel Onderzoek der Materie (FOM); partial support is due to the European Union under the IST-ATESIT contract.

[1] T.W. Ebbesen et al., Nature 391, 667 (1998).
[2] H.F. Ghaemi et al., Phys. Rev. B 58, 6779 (1998).
[3] H. Raether, Surface Plasmons (Springer, Berlin, 1988).
[4] M. Beruete et al., arXiv:cond-mat/0311036 (2003).
[5] J. Gómez Rivas et al., Phys. Rev. B 68, 201306 (2003).
[6] F. Miyamaru and M. Hangyo, Appl. Phys. Lett. 84, 2742 (2004).
[7] J. Elliott et al., arXiv:cond-mat/0310596 (2003).
[8] R. Gordon et al., Phys. Rev. Lett. 92, 037401 (2004). Unfortunately, the authors employ the word depolarization in an improper way, using it to describe a change of a uniform SOP.
[9] R.M.A. Azzam and N.M. Bashara, Ellipsometry and Polarized Light (North-Holland, Amsterdam, 1987).
[10] F. Le Roy-Brehonnet and B. Le Jeune, Prog. Quant. Electr. 21, 109 (1997).
[11] C. Genet et al., arXiv:physics/0311137 (2003).
[12] E. Altewischer, M.P. van Exter and J.P. Woerdman, J. Opt. Soc. Am. B 20, 1927 (2003).
[13] E. Altewischer, M.P. van Exter and J.P. Woerdman, Nature 418, 304 (2002).
[14] E. Altewischer, M.P. van Exter and J.P. Woerdman, to be published
[15] I. Freund, Opt. Lett. 15, 1425 (1990).
[16] R.L. Eriksen et al., Appl. Opt. 42, 5107 (2003).