Near-field optical scattering from plasmonic objects: the way forward

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(Dated: February 8, 2019)

We present freestanding metasurfaces operating at optical frequencies with a total thickness of only 40 nm. The metasurfaces are fabricated by focused ion beam milling of nanovoids in a carbon film followed by thermal evaporation of gold and plasma ashing of the carbon film. As a first example, we demonstrate a metasurface lens based on resonant V-shaped nanovoids with a focal length of 1 mm. The second example is a metasurface phase-plate consisting of appropriately oriented rectangular nanovoids that transforms a Gaussian input beam into a Laguerre-Gaussian $LG_{-1,0}$ mode.

The metasurface concept has set up a new paradigm for the design and fabrication of ultrathin optical devices[1–3]. The underlying idea is to modify the wavefront of a light beam by scattering from a dense array of subwavelength building blocks. These so-called meta-atoms act as antennas, whose scattering properties are controlled by their geometry and composition. Meta-atoms can be categorized based on their functional principle into two large groups: (i) resonant meta-atoms based on plasmonic-[1] or Mie-resonances[4] and (ii) nonresonant meta-atoms based on the Pancharatnam-Berry phase concept[5]. For both types of meta-atoms, the desired phase and amplitude profile of the scattered wavefront is encoded through the appropriate lateral variation of their properties on the metasurface. In this fashion, one can create, for instance, phase gradients leading to anomalous refraction[1, 6], ultrathin lenses[7–9], phase plates for the generation of vortex beams[10–13], and holograms[14–16].

Even though metasurfaces are by definition much thinner than the relevant design wavelength, the corresponding metasurface devices are typically considerably thicker. The reason for this is that in most cases the metasurfaces are fabricated on a supporting substrate, which again introduces some bulkiness to the device. The obvious solution to overcome this limitation is either to shrink the thickness of the substrate to sub-wavelength dimensions [17] or to remove the substrate completely in a freestanding metasurface design.

In this letter, we report on the fabrication and optical characterization of freestanding metasurfaces operating at optical frequencies. The metasurfaces consist of dense arrays of nanovoids in a freestanding gold film with a thickness of only 40 nm. The two examples discussed below demonstrate that our fabrication method is equally suitable for resonant metasurfaces as well as for geometric metasurfaces based on the Pancharatnam-Berry phase concept.

The sample fabrication process is illustrated in Fig. 1. We start with a commercially available carbon film grid (Quantifoil Cu 200 mesh, Quantifoil Micro Tools GmbH) for application in transmission electron microscopy (TEM). The approximately 10 nm thick carbon film is patterned by focused ion beam (FIB) milling with Gallium ions at 30 keV with a current of 10 pA. The footprint of the metasurfaces is 90 $\mu$m $\times$ 90 $\mu$m. For resonant metasurfaces (see example 1 below), we mill V-shaped nanovoids in the carbon film while for geometric metasurfaces (see example 2 below) rectangular nanovoids are produced in this step. Afterwards, the structured carbon film is metallized by thermal evaporation of a 40 nm thick layer of gold. As a last step, the carbon film is removed by a plasma-ashing process with an 80:20 argon-oxygen
gas mixture for 420 s at a pressure of 2.6 μbar.

As a first example, we consider a free-standing metasurface lens composed of V-shaped nanovoids. It is designed for the wavelength \( \lambda = 780 \) nm with a focal length \( f = 1 \) mm. The nanovoids are arranged on a quadratic lattice with a period of 500 nm. Their axes of symmetry are oriented parallel to each other and form an angle of 45° with the \( x \)-axis. Each of the meta-atoms supports a series of plasmonic resonances, whose resonance frequencies can be controlled by the respective nanovoid geometry, i.e., the opening angle and the arm length. For our design, only the two first plasmonic modes play a role. The fundamental resonance can be excited with light linearly polarized along the axis of symmetry while the second plasmonic resonance requires a linear polarization orthogonal to this. Upon illumination with a laser beam, which is linearly polarized along the \( x \)-axis, both resonances can be excited and contribute to the scattered signal that is polarized along the \( y \)-direction and hence orthogonal to the incident light. The phase shift introduced by one of the V-shaped nanovoids to this cross-polarized scattered component depends on the spectral detuning of the two resonances with respect to the laser frequency. Thus, by tailoring the geometries of the nanovoids in the metasurface, one can imprint the desired phase profile on the cross-polarized scattered wave.

Before fabrication of the metasurface, the commercial finite-element-solver COMSOL was used to calculate the phase shift introduced by different nanovoid geometries. All in all, 94 different combinations of opening angle and arm length were evaluated. From this set, the geometries of the nanovoids were chosen such that for each position \((x, y)\) the calculated phase shift of the selected nanovoid fits best to the phase-shift profile of the converging lens (see Fig. 2a) with the design parameters given above:

\[
\phi(x, y) = \frac{2\pi}{\lambda} \left[\sqrt{x^2 + y^2} + f^2 - f\right].
\]

Figure 2b) depicts a scanning transmission electron micrograph of a section of the metasurface lens. The nanovoid arm length varies between 120 nm and 240 nm and the opening angle ranges from 45° to 180°.

In order to test its performance, we illuminate the metasurface lens with a collimated laser beam (780 nm wavelength). A first polarizer in front of the sample ensures that the electric field vector is aligned parallel to the \( x \)-axis. The transmitted light is imaged with a lens \((f_1 = 50 \) mm\) onto a CCD camera. A second polarizer oriented along the \( y \)-axis positioned in front of the camera acts as the analyzer, which only lets pass the cross-polarized light wave scattered by the metasurface. By moving the imaging lens along the optical axis (\( z \)-axis), we can record the intensity distribution \( I(x, y, z) \) in different \( z = \text{const.} \) planes behind the metasurface lens. The corresponding data is presented in Fig. 2c). As intended, the scattered light field is focused 1 mm behind the metasurface lens. Figure 2d) shows the intensity distribution in the focal plane \((z = 1 \) mm\). The measured full width half maximum of the focused light spot is approximately 10 μm. This value is in good agreement with the expected spot size (FWHM) \( 2W_0 \approx \frac{\lambda F_\#}{\pi \sigma^2} \approx 13.3 \) μm of a Gaussian beam with \( \lambda = 780 \) nm wavelength that is focused by a lens with F-number \( F_\# = f/D = 11.1 \), where \( D \) is the diameter of the lens.

In the second example, we use rectangular nanovoids to implement a freestanding metasurface phase-plate that transforms a Gaussian beam, i.e., the Laguerre-Gaussian \( LG_{0,0} \) mode, into the Laguerre-Gaussian \( LG_{-1,0} \) mode. The nanovoids with dimensions of 230 nm \( \times \) 50 nm (length \( \times \) width) are arranged on a quadratic lattice with 500 nm period. Scattering of an incident circularly polarized wave by a rectangular nanovoid creates a wave with orthogonal circular polarization and a phase shift \( \phi = 2\sigma \theta \), where \( \theta \) is the the angle between the long nanovoid axis and the \( x \)-axis and \( \sigma \) characterizes the circular polarization state of the incident beam (right circular polarization...
(RCP): $\sigma = 1$, left circular polarization (LCP): $\sigma = -1$.

In order to generate a Laguerre-Gaussian $LG_{-1,0}$ mode from a left-handed circularly polarized Gaussian input beam, the metasurface has to imprint the phase-shift profile $\phi(r, \varphi) = -\varphi$ onto the scattered wave (see Fig. 3a)). Following the Pancharatnam-Berry phase concept, we design the metasurface phase-plate such that the inclination angle $\theta(r, \varphi)$ of the nanovoid at the position $(r, \varphi)$ relative to the $x$-axis is given by $\theta(r, \varphi) = -\phi(r, \varphi)/2$. Figure 3b) depicts a scanning electron micrograph of the central section of the metasurface phase-plate.

For the experimental characterization of the metasurface phase-plate, we add two quarter-wave plates to the setup described above. The first quarter-wave plate is placed behind the first polarizer and is used to illuminate the metasurface with left-handed circularly polarized light. The second quarter-wave plate is positioned in front of the analyzer and selects in combination with the latter the scattered right-handed circularly polarized wave created by the metasurface. Figure 3c) depicts the intensity distribution at a distance 750 $\mu$m behind the metasurface. As expected for the $LG_{-1,0}$ mode, it features a donut-shaped mode profile with an intensity minimum on the optical axis.

The topological charge $l$ of the generated Laguerre-Gaussian mode[20] can be easily determined by using the setup as a common-path interferometer. For this purpose, the polarization axis of the analyzer is rotated such that the scattered wave and the Gaussian input beam are overlapped with comparable intensities on the CCD camera. The interference of these two contributions creates an intensity pattern with a spiral fringe (see Fig. 3d)). Since the phase fronts of the Gaussian beam do not vary with the azimuthal angle $\varphi$, we can conclude that the topological charge of the generated Laguerre Gaussian mode is $l = -1$.

In summary, we presented a new approach for the fabrication of freestanding metasurfaces operating at optical frequencies. This method is suitable both for resonant and for geometric metasurfaces. We see as a big advantage that such freestanding metasurfaces can be easily transferred to substrates that are not suitable for standard nanolithography, e.g., curved interfaces. Preliminary tests show that the copper grid can be dissolved in a suitable etch without degrading the optical quality of the gold metasurface.

**FUNDING**

S.L. acknowledges financial support by the German Federal Ministry of Education and Research through the funding program Photonics Research Germany (project 13N14150).

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