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Negative-Index Metamaterials: Looking into the Unit Cell

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ABSTRACT With their potential for spectacular applications, like superlensing and cloaking, metamaterials are a powerful class of nanostructured materials. All these applications rely on the metamaterials acting as a homogeneous material. We investigate a negative index metamaterial with a phase-sensitive near-field microscope and measure the optical phase as a function of distance. Close to the metamaterial we observe extremely large spatial phase variations within a single unit cell which vanish on a 200 nm length scale from the sample. These deviations of a state-of-the-art metamaterial from a homogeneous medium can be important for nanoscale applications.

KEYWORDS Near-field optics, nanophotonics, metamaterials, negative refractive index

The optical properties of "natural" bulk materials arise from a complex interplay between the electronic orbitals of the constituent atoms, which leads to a certain electronic band structure. An externally applied light field interacts with electrons in this band structure. In general, this results in a dispersion of the refractive index and also leads to absorption-edges and even to sharp Lorentz-like absorption lines. Thus a natural material can be considered as an effective medium in which the role of the individual atoms no longer has to be considered.[1]

Similarly in the field of metamaterials,[2] we like to think in terms of constituent "meta-atoms" that couple with each other to give rise to the optical properties of the bulk material, which is then considered as an effective medium.[3] Several important experiments have been performed to elucidate the interplay between the meta-atoms that constitute a metamaterial. For instance, the response of a single meta-atom was measured and compared with that of an array of meta-atoms.[4] Also, the response of an array of meta-atoms was studied as a function of the periodicity in two directions, to extract both the electric as well as the magnetic coupling between the meta-atoms.[5,6] By now, it is well understood that, as in the case of natural materials, the effective optical properties of a metamaterial are not simply the addition of its constituent meta-atoms, and a monolayer of a metamaterial has not yet developed all the optical properties of the bulk metamaterial. As a result, the properties of a monolayer of meta-atoms that we study in this letter are not necessarily the same as those of a bulk material.[7] Nevertheless, here we use the notion "negative-index metamaterial" for a monolayer to connect to a very large number of corresponding publications.

To fully understand how negative index metamaterials can be applied on the nanoscale as in, for instance, a near-field superlens,[8] it is not sufficient to know the effective medium response of the material that is measured far away from the structure, that is, in the far field. For nanoscale applications, it is crucial to also know the near-field behavior. Hence, we would like to take a look inside the unit cell of a negative index metamaterial. A recent pioneering effort gained the first glimpse of the meta-atoms in an array of so-called split-ring resonators that exhibits positive effective refractive index and operates in the infrared regime.[9] While these measurements showed spatial variations of the field, the crossover behavior to the distance at which the structure behaved as a homogeneous medium remained unexplored.

In this Letter, we present a phase-sensitive near-field investigation of negative index metamaterials within a single unit cell.[10] We find that the optical phase varies strongly within the unit cell and that, surprisingly, the maximum of the detected field lies behind the metal, instead of behind the glass as one might have intuitively expected. We find that the large variations in the field on the scale of a unit cell exponentially decrease as the distance to the structure is increased. For distances larger than 200 nm, the wavefront of the transmitted light is flat and the material can be treated as a homogeneous medium.[10,11] However below that dis-

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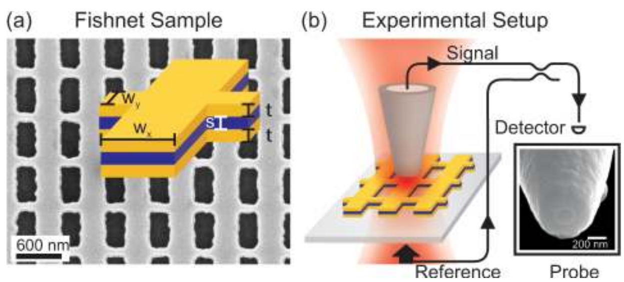

FIGURE 1. (a) Top-view electron micrograph of the investigated double-fishnet negative index metamaterial structure. The inset shows a 3D sketch of metamaterial unit cell. (b) Scheme of the experimental setup and the metamaterial sample. The inset shows an electron micrograph of the coated probe used in this investigation.

tance, the metamaterial can no longer be considered as homogeneous since large optical phase and amplitude variations are observed.

Figure 1a shows an electron micrograph of the investigated metamaterial. We choose the so-called double-fishnet design, which is a well-established structure that has previously been studied in far-field experiments.[10] The far-field optical properties of this type of structure have already been thoroughly investigated.[10,12−15] In the near-field experimental setup, schematically illustrated in Figure 1b, the sample is illuminated under normal incidence from the glass substrate side by a weakly focused beam (the waist of the focus is roughly 60 μm). On the scale of one unit cell of the metamaterial structure, this excitation can be viewed as a plane wave. The nanoscale geometry of the sample (thickness of the Au layer is $t = 25$ nm, thickness of the MgF2 is $s = 35$ and the wire dimensions are $wx = 320$ and $wy = 90$ nm) has been chosen such that incident light with linear polarization parallel to the narrow metal stripes experiences an effective negative phase velocity at wavelengths in the telecom range.[10]

The near-field probe that we use has a 200 nm aluminum coating and an aperture with a diameter of 200 nm (Figure 1b), through which light is collected. The probe is raster scanned at a constant distance of 20 nm above the structure. We retrieve the phase of the collected light by interferometrically mixing it with a reference beam.[16] Figure 2 a,b presents the amplitude and phase images measured in the near field at vacuum wavelength $\lambda = 1500$ nm. The depicted area has a size of roughly 3 × 2 unit cells. It is evident that both the amplitude and the phase exhibit significant spatial variations within a single unit cell. These large spatial variations clearly show that at a distance of 20 nm the structure cannot be treated as a homogeneous medium.

To assign the location of the optical features we observe, it is crucial that we have reliable knowledge of the relative positioning of the optical images and the actual metamaterial structure. Topographic information obtained with the coated aperture probe during the optical measurements is routinely acquired but can sometimes be misleading as the part of the probe nearest to the sample is not necessarily located in the middle of the probe aperture. Therefore, we perform additional experiments with special probes on the same sample. We use focused ion beam milling to fabricate probes that have a nanoscale dielectric protrusion (typical height 100−200 nm), the apex of which is located in the center of the probe aperture. Since the dielectric protrusion is small enough not to affect the optical measurements and its apex is now always the part of the probe that is nearest to the sample, an accurate determination of the position of the optical features with respect to the geometry of the sample can be made. Details are reported in the Supporting Information.

Interestingly, we find that the maxima of the measured amplitude in Figure 2a occur when the probe is located behind the metal−dielectric−metal structure (the drawn rectangles in Figure 2 represent the holes in the structure) rather than behind the air holes. This observation is in marked contrast to the naive expectation of geometrical optics for which one would have rather expected the maxima behind the holes.

We furthermore notice that Figure 2b exhibits a significant spatial variation of the optical phase. Throughout this Letter, the sign of the phase is defined as follows. A plane wave at carrier frequency ω propagates along the z-direction according to $\propto \exp(i(kz - \omega t))$. Thus, upon propagation over a positive distance z (e.g., the sample thickness), it acquires a phase $\phi = kz$. For propagation in a medium with positive (negative) refractive index n, the wavenumber $k = \omega/v = n\omega_0/c$ is positive (negative). Hence, the sign of the phase ϕ is quite intuitive: propagation through a negative-index material leads to a negative phase. For convenience, we choose the phase of the average of the complex interferometric signal in a single unit cell, ϕ_0, as reference for the local phase $\phi(x, y)$ at each wavelength. In the following, we will therefore use the term "phase difference" when discussing the position-dependent phase signal.

The phase difference in Figure 2b locally varies between about $\pm\pi/2$. Our previous far-field experiments on a closely similar sample have shown a negative index of refraction (approximately $n \simeq -1$ at 1500 nm), corresponding to a negative phase acquired while propagating through the sample.[10] For the sample thickness of 85 nm and, for example, a refractive index of $n = -1$ the propagation phase is -0.11π. We can therefore conclude that the local phase variations within a single unit cell in the near field are considerably larger in magnitude than the propagation phase. The largest negative phase differences occur behind the holes and the thin wires of the double-fishnet structure, that is, around the positions marked as "3" and "4″, respectively. This statement also holds true for other wavelengths as can be seen by the local phase difference spectra depicted in Figure 2c.

For every (x, y)-plane at a certain height above the structure, we can determine a phase difference histogram that reflects the distribution function of the phase differences within a unit cell. Figure 2d depicts the measured phase-

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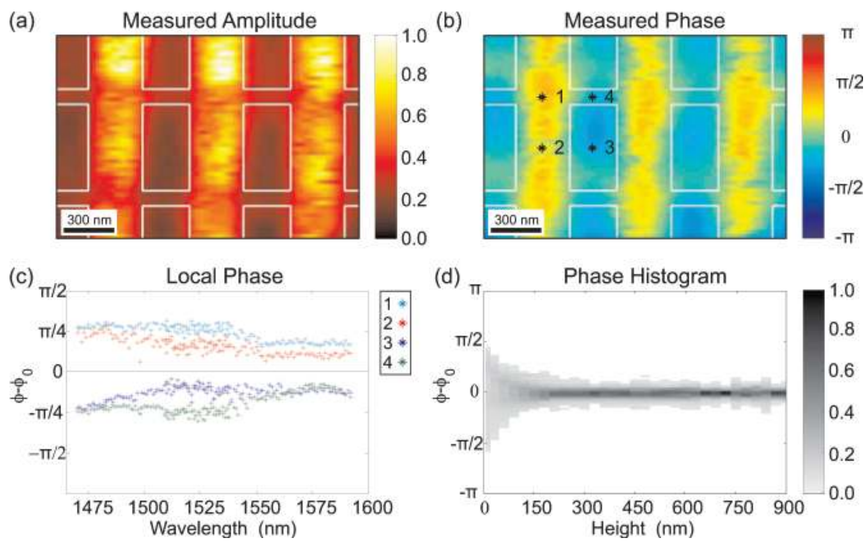

FIGURE 2. (a) Measured amplitude and (b) phase near-field images at a wavelength of $\lambda = 1500$ nm. The superimposed white rectangles indicate the metamaterial's structure. (c) Local spectral phase variation at four different locations within the metamaterial unit cell, as indicated in (b). (d) Histogram of the local phase as in (b) as a function of the height of the probe above the metamaterial surface.

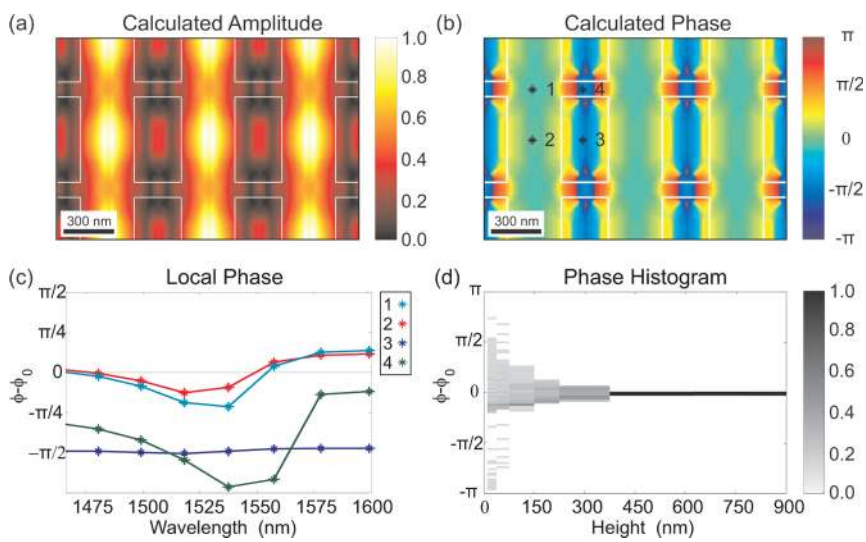

FIGURE 3. Calculated data that can be directly compared to the experimental data shown in Figure 2.

difference histograms for different heights of the probe above the metamaterial surface. We observe an exponential decay of the local phase variations with increasing height. The measured phase variations have vanished for heights larger than 200 nm so that the light beyond this distance from the fishnet structure can be considered a plane wave. All these findings indicate that metamaterials can be correctly considered as homogeneous media only in the far field,[10,11] whereas in the near field this is no longer true.

We complement our experimental findings with numerical calculations obtained by finite-integration technique. The gold is described with the Drude free electron model with parameters identical to those in ref 10. Details on the calculations can be found in the Supporting Information. Given the highly structured field distribution present close to this metamaterial, we may expect the electromagnetic response of the probe to be rather complicated. The probe typically collects the transverse electric field parallel to the incident polarization.[17−19] However, since the spatial gradient of the axial component, that is, the z-component, of the electric field just above the fishnet structure is larger than in previously investigated photonic structures,[17−19] we expect that this component is also collected.[20] More information can be found in the Supporting Information. Following these expectations, we calculated the total field collected by the probe taking into account both the transverse component and the gradient of the axial component. The resulting amplitude and phase maps are shown in Figure 3a,b, respectively. We find a reasonable agreement with the corresponding measurements shown in Figure 2a,b. Similarly, we calculate the local phase difference as a function of wavelengths. Figure 3 (c) shows that the local phase

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variations within one metamaterial unit cell exceed the modulus of the propagation phase, as in the experimental data (Figure 2c). Finally, Figure 3d shows the exponential decay of the strength of the phase variations from the optical near field to the far field on a comparable scale as the experiment in Figure 2d.

In conclusion, by employing near-field phase-sensitive optical microscopy, we have mapped the phase fronts of light within a unit cell of a state-of-the-art negative index metamaterial and investigated its development as a function of distance from the structure. We found that in the near field, the sample cannot be considered as a homogeneous medium. To better understand the optical response of such a metafilm in the near field, its optical properties should be described by optical constants that depend not only on frequencies but also spatial Fourier components.[21] These findings might have important consequences for the use of negative index metamaterials for nanoscale applications, such as superlensing. In such a lens all details of a light source in the near field of one side should be perfectly transferred to the near field of the other side.[8] Given the findings reported in this Letter, the suitability of a metamaterial as perfect lens can depend on the position of the source within a single metamaterial unit cell.

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Note Added after ASAP Publication. There was an error in the title of two panels of Figure 3 in the version of this paper published ASAP June 2, 2010; the correct version published on June 16, 2010.

Supporting Information Available. Optical and topographic imaging, calculation of the detected field distribution, and additional figures and references. This material is available free of charge via the Internet at http://pubs.acs.org.

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