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Flexible metamaterials at visible wavelengths

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Abstract. We report on the fabrication and characterization of plasmonic structures on flexible substrates (Metaflex) and demonstrate the optical properties of a single layer of Metaflex. The layer exhibits a plasmonic resonance in the visible region around 620 nm. We show experimental and numerical results for both nano-antennas and fishnet geometries. We anticipate the use of Metaflex as a building block for flexible metamaterials in the visible range.

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1. Introduction

Metamaterials (MMs) typically consist of periodic distributions of metal resonators that can be engineered to yield both negative electric permittivity and magnetic permeability, exhibiting effective responses not present in naturally occurring materials. At present, most of the applications based on MMs are limited by their planar fabrication approach. Contemporary MMs are hard and stiff.

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Here, we propose and demonstrate plasmonic structures on flexible substrates (Metaflex) and show their optical properties in the visible region around 620 nm.

The response of MMs is determined by a tailored distribution of meta-atoms (typically periodically arranged and metallic), such as split ring resonators or, more recently, fishnet lattices [1]. If the MM operates at optical wavelengths, the size of the meta-atoms has to be scaled down to a few tens of nanometers [2]. For this reason, traditional fabrication techniques rely on approaches inherited from nanotechnology, e.g. electron beam lithography, which typically requires rigid substrates such as quartz or silicon.

The unique properties of MMs have led to the demonstration of exciting concepts such as superlensing [3] and invisibility cloaking [4]. Both these concepts, as well as other possible applications of MMs, are constrained by the limitation imposed by the fabrication constraints. For example, a 'real' cloaking device would have to be deformable and extend over a large area, rather than being fabricated on rigid substrates such as silicon [5].

The same is true for the superlens, where the flat geometry only allows for the formation of an image in the near field, while a magnifying superlens that pushes the focal length to more practical distances would require a curved realization. Such a curved realization has recently been demonstrated via a tour-de-force in lithography [6], again on a planar substrate, but it is not obvious how this method would scale up in size.

On the other hand, optical conformal mapping or transformation optics (TO) [7]–[10] offers an alternative approach that aims at fabricating transparent materials, whose dispersion properties are varied spatially [11]. According to TO, a geometry with a desired optical response is mapped onto a target topology, which is easier to realize.

While using TO it is possible to generalize some of the concepts behind MM applications, such as perfect invisibility and negative refraction (e.g. by creating folded coordinate transformations) [12]; TO does not strictly require traditional MMs (i.e. periodic arrays of nanosized metallic structures), and recent results suggest that negative refraction itself is not necessary either [13]. TO is a very powerful tool for designing advanced optical devices, and it would be useful for developing a technique that would naturally allow the implementation of curved topology.

In this work, we show that Metaflex can operate in the true optical regime by demonstrating plasmonic resonances down to a wavelength of 620 nm.

Metaflex can be fabricated using standard nanotechnology (e.g. electron beam lithography), but we have developed a fabrication technique to obtain supple and deformable substrates.

Single—individually fabricated—layers of Metaflex can be conveniently used to assemble 3D stacks, with an effective tailored response. The layer-by-layer approach to 3D MMs has the inherent advantage over traditional techniques of decoupling the transversal resolution (of the meta-atom) to the final size of the stack.

In principle, the approach is similar to the flexible printed circuit board technology widely used in the computer industry. It is therefore not surprising that the idea has recently been applied to obtain flexible MMs over the THz range [14], but not yet in the optical regime. A related technique for the creation of small metallic nanofeatures on curved surfaces has also been demonstrated, but no characterization was shown [15].

Clearly, realizing a desired circuitry directly on a flexible substrate is very desirable, as also shown by recent advances in electronic foldable paper, flexible displays or smart fabrics [16, 17]. We believe that Metaflex will be relevant not only to the physics and applications of MMs. Every

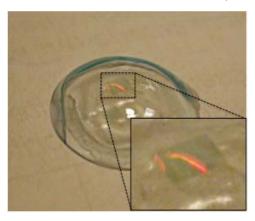


Figure 1. Photograph of a Metaflex membrane placed on a disposable contact lens and illuminated with office light. The inset is an enlargement of the membrane.

application where flexibility is required can benefit from the Metaflex paradigm. As a simple example of the versatility of the flexible approach, we have placed a layer of Metaflex onto ordinary contact lenses (figure 1).

2. Results

In the following, we describe the fabrication procedure required for obtaining a single Metaflex layer for operation in the near infrared (NIR) and the visible wavelength range.

2.1. Fabrication

The membranes were fabricated using a commercially available polymer (SU8, Microchem), which can be spun in thicknesses varying from a few nanometers to hundreds of microns. The fabrication procedure is summarized in figure 2. A sacrificial layer (XP-SU8 release layer, from Microchem) is spun on a host substrate (typically silicon) and baked for 1 min at 230 °C (figure 2(a)). A suitable blend of different SU8 membranes is then spun onto the sample to obtain a membrane of the desired thickness, which is cured at 100 °C for 5 min (figure 2(b)). At this stage, the lateral extent of the membrane can be defined photolithographically if desired.

An Au layer of 40 nm thickness is then deposited by evaporation (figure 2(c)). A further thin layer of SU8 (100 nm) is finally spun onto the sample to act as an electron-beam resist. The patterns are defined using a modified LEO-RAITH ebeam lithography system with a positioning accuracy of 2 nm and a maximum voltage of 30 kV. Following a post-exposure bake (2 min at $100\,^{\circ}$ C), the sample is developed (EC, Shipley) (figure 2(d)) and the pattern is then transferred onto the gold layer via argon-assisted reactive ion etching (figure 2(e)). Finally, the membranes are released from the substrate via immersion in N-methylpirrolydone (NMP) for \sim 2 h (figure 2(f)).

This last step is critical and its success depends on the geometry of the membrane. For example, for large and very thin membranes, lift-off is problematic, because the edges of the membrane might collapse and prevent the complete infiltration of the solvent [18]. So far, we have been able to realize membranes as thin as $4 \mu m$ and extending over an area of $5 \times 8 mm^2$.

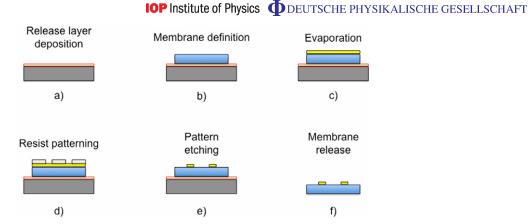


Figure 2. Fabrication process. (a, b) Substrate pre-treating and membrane definition. (c–e) Gold evaporation and pattern definition. (f) Membrane release.

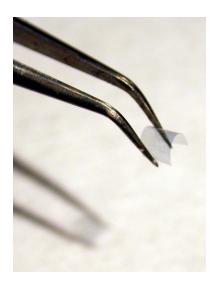


Figure 3. Photograph of hand-held tweezers holding a typical Metaflex membrane.

Figure 3 shows a picture of such a membrane, which is easily handled with laboratory tweezers.

We tested the electromagnetic response of Metaflex by defining two different plasmonic structures, namely nanoantennas and a 'fishnet' lattice, both of which are well known and characterized on rigid structures [19]–[22].

2.2. Characterization

Samples were characterized optically with a polarization-controlled white light source (Ocean Optics tungsten halogen light source).

The source was collimated on to the sample and the transmitted signal was collected by a $100\times$ objective and then spectrally analyzed (Ocean Optics USB 2000; $500-1000\,\mathrm{nm}$). In the measurements, the samples were probed at normal incidence. The effect of curvature on the Metaflex response is beyond the scope of this work, and will be addressed in forthcoming publications.

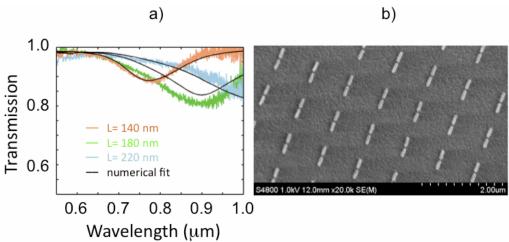


Figure 4. (a) Transmission curves for three different nano-antennas, geometries and numerical modelling. (b) SEM image of a typical sample.

We realized and characterized a number of different nanoantenna geometries with different arm lengths ranging from 100 to 200 nm and gap spacing ranging from 20 to 100 nm. The nanoantennas are arranged in a square lattice with periodicity $p = 1 \mu m$ in both directions.

Figure 4 shows the transmission spectra for nanoantennas with three different parameters.

The curves show the typical absorption peak, which shifts with the overall length of the nanoantenna. The optical field was selected to have the electric field polarized along the nanoantenna long axis. Figure 4(b) is an SEM picture of the sample.

To verify the result, we performed numerical calculations using finite-element simulations (COMSOL), taking the dispersive properties of SU8 (http://www.microchem.com) and gold [23] fully into account. As shown in figure 4(a), the experimental and the numerical curves match rather well.

Both the numerical and experimental transmission curves presented in figure 4(a) were normalized with respect to a membrane without the gold patterns.

The experimental curves were spectrally filtered to remove the oscillations due to Fabry–Perot resonances at the interface of air/membrane and membrane/gold.

We took advantage of the symmetry of the system and simulated only a quarter of the unit cell of the periodic pattern. Slight differences between the experimental and numerical data are likely to be due to the roughness of the membrane, caused by the ashing of the photoresist, after the gold etching step.

In the simulations, the SU8 membranes were assumed to be uniform and hence with higher transmission. The black curves in figure 4 are best fits of the experimental ones, based on SEM images of the measured samples, with an accuracy of ± 10 nm. For each sample, we considered a gold thickness of 40 nm and arms width of 70 nm. The short nanoantennas (L=140 nm) had a gap of 60 nm. The long ones (180 and 220 nm) had a gap of 90 nm. The period of the nanoantenna distribution was 950 nm.

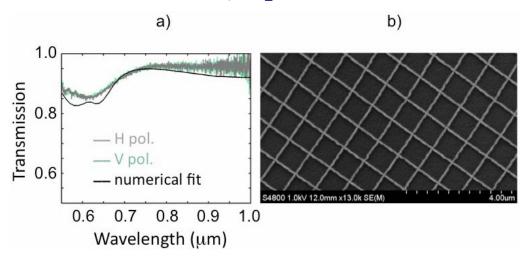


Figure 5. (a) Transmission curves for two orthogonal polarizations (parallel to the fishnet axes) and numerical modeling. (b) SEM image of a typical sample.

2.3. Towards 3D flexible metamaterials

Arguably, one of the most exciting applications of Metaflex is to fabricate three-dimensional flexible MMs in the optical range, which can be achieved by stacking several Metaflex membranes on top of one another.

The natural choice for the corresponding metallic nanostructure is the fishnet, as it has become the preferred building block for MMs in the visible range [24]. To achieve this goal, we realized a fishnet structure with a period of $p = 1 \mu m$ and gold wires of width equal to $\sim 100 \text{ nm}$.

Figure 5(a) shows the corresponding transmission spectrum with the electric field aligned along either one of the main axes of the pattern and the numerical simulations are shown for comparison. Again, the experimental and calculated resonance frequencies are in good agreement and indicate the resonance dip correctly. Figure 5(b) shows an SEM micrograph of the patterned membrane.

For the numerical curve in figure 5, we considered a period of $1 \mu m$, a gold thickness of 40 nm and a gold width of 85 nm.

3. Discussion

While the experiment was realized on a flat membrane and for normal incidence, we believe that this sort of structure is the ideal candidate for addressing an advanced implementation of bulk MMs that consist of a multilayer stack.

Assembling such a stack requires an inter-layer distance of the order of a few hundreds of nanometers [25], which can be done using the Metaflex approach.

The critical factor then becomes the thickness of the membranes. SU8 can indeed be spun at thicknesses down to 100 nm, but there may be practical limitations in terms of the possible membrane area; a quantitative study of these limitations is currently in progress.

We have fabricated and characterized plasmonic nano-structures that were realized on flexible polymeric substrates. We studied both nanoantennas with varying geometrical parameters and fishnet structures, and demonstrated their operation in the NIR and the visible wavelength range, respectively. The experimental curves agreed well with the numerical calculations. These results confirm that it is possible to realize MMs on flexible substrates and operating in the visible regime, which we believe are ideal building blocks for future generations of three-dimensional flexible MMs at optical wavelengths.

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