第 3 卷　第 1 期　　中国光学与应用光学　　Vol. 3　No. 1
2010 年 2 月　　Chinese Journal of Optics and Applied Optics　　Feb. 2010

文章编号　1674-2915(2010)01-0001-10

Tailoring the properties of optical metamaterials

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Abstract: In this contribution we review our latest achievements of combined experimental and theoretical studies to tailor the properties of optical metamaterials (MMs) at will. We give three examples of metamaterial designs that have been realized by means of electron-beam lithography and whose spectroscopic characteristics have been comprehensively investigated. In every case, our experiments are complemented by rigorous numerical simulations. Particular emphasis is put on the significance of such tailored effective properties of optical MMs.

Key words: optical metamaterials; lithography; negative-index

1　Introduction

Metamaterials (MMs) are manmade media in which the propagation properties of electromagnetic radiation are significantly governed by their artificially structured geometry rather than by the natural materials they are composed of. When the electromagnetic waves interacting with these media stem from the optical or near-infrared spectral domain, they are called optical MMs[1,2]. They allow in principal for the control of propagation properties of an optical wave field; like refraction, diffraction, dispersion, phase- and group-velocities. Thus, the knowledge and access to these optical control mechanisms *via* optical MMs enable the realization of optical components with comprehensive functionalities[3-7]. Moreover, by means of MMs the interaction between light and matter can be extended into domains where nature doesn′t provide any equivalent. Accordingly, although still at the stage of fundamental research, MMs are expected to elicit a boost in the field of modern optics[8,9].

In recent years, a large part of the effort to explore optical MMs was aimed at the derivation of comprehensible design guidelines to realize naturally

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Received date: 2009-10-21; Revised date: 2009-12-24.

unattainable optical functionalities. In this paper we consider a class of MMs whose unit cells have a spatial extent much smaller than the wavelength of the interacting optical radiation. If this condition is met, the light propagation in the MM will be governed by the normal modes of a homogeneous medium. Such MMs are commonly described as assuming effective properties, which, and this is an important statement, can be deliberately tailored. Usually, an effective electric permittivity and an effective magnetic permeability are introduced that can be utilized to derive formally an effective index of refraction.

The usage of these effective properties greatly facilitates the description of light propagation through MMs because details of the correct unit cell geometry forming the MM can be neglected. Despite their unquestionable usefulness for MM designers, it must be carefully borne in mind that the effective material properties are an intuitive yet simplified approximation in order to model light propagation through MMs instead of accurately describing the MM itself. The critical issues in deriving meaningful effective properties were recently discussed in [10]. However, to have such properties at hand, the common approach relies on a retrieval algorithm of effective properties by means of the inversion of the scattering problem of a homogenized finite slab at normal incidence[11]. This algorithm was recently generalized for oblique incidence including the influence of a substrate[12] as it is necessary for the practical realization of any MM.

In this contribution we review our latest achievements to tailor the properties of optical MMs. We show combined experimental and theoretical studies of three exemplary MM designs. All these samples have been realized by means of a Vistec SB350OS electron-beam writer and lift-off techniques. Their optical responses have been investigated spectroscopically. In each case our experiments are complemented by numerical simulations applying either the Fourier Modal Method[13] or Finite-Difference Time-Domain simulations[14]. For the rigorous treatment both the exact geometry of the respective unit cells and the spectral dependence of the material properties as documented in the literature were appropriately taken into account.

The paper is organized as follows: Section 2 addresses a near-infrared, negative-index MM composed of two distinct unit cell elements. This approach permits an independent tuning of the geometry of the unit cell components. Section 3 deals with a novel MM design that releases the constraint of polarization dependency. The Swiss-cross MM was recently shown to have a polarization independent optical response for normal incidence and a negative index of refraction at $\lambda = 1.4$ μm. Moreover, we study the optical properties of the Swiss-cross MM at oblique incidence and reveal its angle-dependent effective properties.

It is shown that the spectral and angular domains of the negative refractive index as well as its magnitude are closely connected to the propagation direction and the polarization state of the illumination. Generally speaking, the optical response is dominated by spatial dispersion, as it is expected for any thin film MM that has been published to date. The implications for the notion of effective properties of common MMs are discussed. In Section 4 we attempt to evaluate experimentally the requirement of a periodic arrangement of the unit cells in optical MMs. The answer to this question is urgently needed since the serial fabrication methods of today′s MMs are expected to be replaced by faster and less costly self-assembling or chemically randomized fabrication schemes. We investigate a model MM system by gradually increasing the degree of positional disorder with respect to its unit cells. The observable spectral features occurring upon this transition and the impact of the effective properties of the MM are revealed. Most importantly, we confirm that the magnetic properties of common MMs are hardly affected by an arbitrarily high degree of positional disorder of the unit cells. We elucidate the encouraging conclusions to be drawn with respect to negative index materials and potential devices composed of them.

2 Double-element negative-index structure

In the framework of an effective medium approach, the issue of providing an effective magnetic permeability different from unity, *i. e.* a magneto-optical activity, was commonly employed by the excitation of localized plasmon polariton eigenmodes in metallic nanostructures[15]. In the optical domain, the double cut-wire structure has attracted particular attention[16]. A magnetic moment arises from an anti-symmetric localized plasmon polariton that can be excited if the illuminating electric field is polarized parallel to the wires. Combining this structure with continuous metallic wires decreases its effective plasma frequency and provides control of the effective electric permittivity of the medium. In the well-known fishnet structure, these two components merge into a single unit[17]. Accordingly, the double cut-wire structure is of interest due to the possibility of tailoring the geometry of both structural elements[18].

The geometry of the unit cell is shown in Fig. 1(a). The periods of the structure are P_x = 500 nm and P_y = 600 nm. The width of the continuous wires is W_1 = 130 nm and of the cut-wires W_2 = 100 nm, the length of the cut-wires is L = 430 nm, and the thicknesses of the metal layer d_{Me} = 40 nm and the dielectric spacer d_s = 40 nm. The SEM micrograph in Fig. 1(b) shows a fabricated sample revealing the vertical structure by an FIB-slice. In the experiment we consider normal light incidence and an electric field polarization parallel to the wires. In this configuration the cut-wire structure

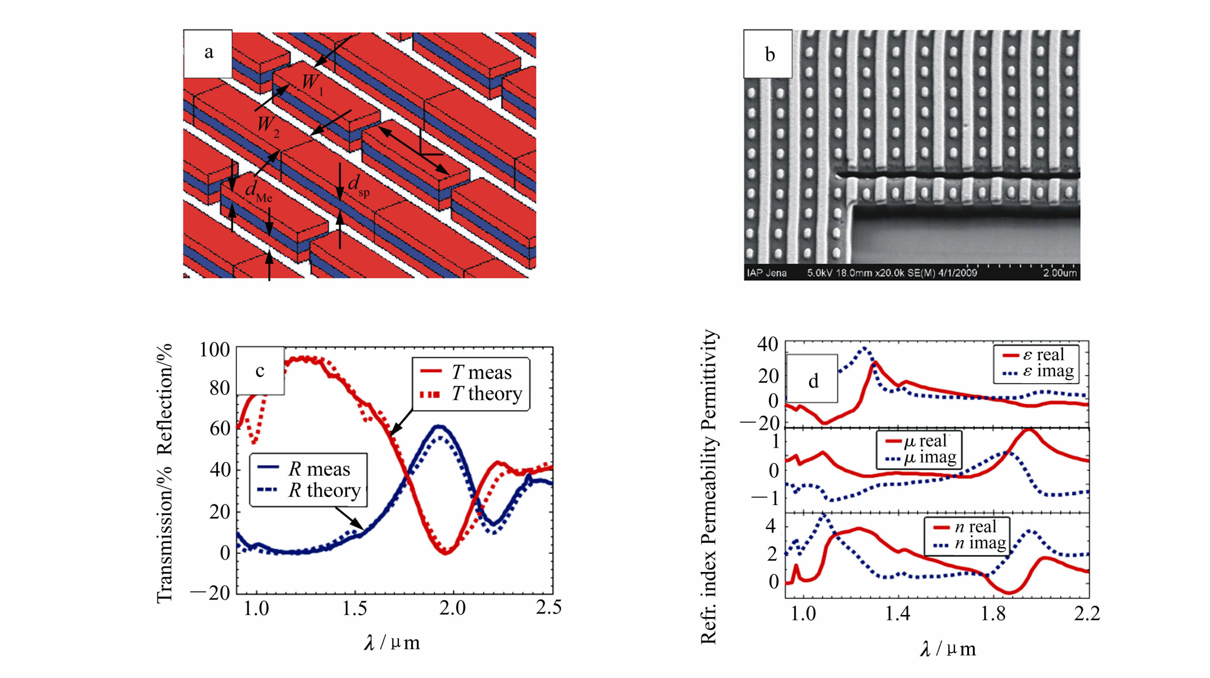

Fig. 1 (a) Schematic of the double cut-wire MM with structural design parameters, (b) tilted electron micrograph view of a fabricated double-cut wire sample sliced with a focused ion beam to visualize the vertical structure, (c) transmission at normal incidence (0, grey lines) and reflection (8°, black lines) spectra for the resonant polarization. Solid and dotted lines represent measured and calculated spectra, respectively, (d) real (grey solid lines) and imaginary (black dotted) parts of the electric permittivity ε, magnetic permeability μ and refractive index n of the sample derived from the calculated spectra shown in c).

supports two plasmonic eigenmodes with different eigenfrequencies. The excitation of an anti-symmetric mode corresponds to anti-phase current oscillations in the cut-wires and evokes the appearance of a permeability resonance. In Fig. 1(c) the measured transmission and reflection spectra are compared to numerical simulations. Regarding the multiple spectral features, we concentrate on the transmission minimum near $\lambda = 2.1$ μm. Here the anti-symmetric eigenmode is most excited. This is confirmed by the calculation of the effective permeability[12], where a resonance with a Lorentzian line shape is observed at $\lambda = 2.1$ μm. With respect to the effective refractive index, we conclude that $n = -0.5 + 1.9\mathrm{i}$ at $\lambda = 2.1$ μm can be formally attributed to our structure, as shown in Fig. 1(d). The options to further tailor the efficiency of the structure are manyfold due to free design parameters. For instance, increasing the strength of the anti-symmetric resonance goes along with a decrease of the period in the x-direction or alternatively with an increase of the width of the cut-wires. Another possibility is to break the vertical symmetry of the cut-wire[19].

3 Polarization-independent Swiss-cross structure and its angular response

The optical MM presented in the former section exhibits its particular optical property, a negative refractive index, for normal incidence and for one polarization state of the electric field component only. This dependency applies to most prototypical unit cells of currently published optical MMs and can be reduced only by a novel design approach. It can be anticipated that for future applications of MMs, a polarization-insensitive response is highly desirable. In addition to that, the angular response of any thin film MM must be known explicitly if it is to be employed in imaging concepts[20]. Here we present a first practical approach to address these issues. The Swiss-cross structure[21] was recently shown to have a polarization-independent optical response for normal incidence. The principle design of the unit cell is illustrated in Fig. 2(a) and the fabricated sample is shown in Fig. 2(b). The structure has a lattice constant of 410 nm in both lateral dimensions. The width and length of the arms of the Swiss cross were designed to be $x_s = 80$ nm and $x_l = 310$ nm, respectively. The thicknesses of the gold films and the intermediate dielectric magnesia film were set to be $d_{\mathrm{Au}} = 30$ nm and $d_{\mathrm{MgO}} = 37.5$ nm, respectively. Remarkably, the metamaterial extends over an area of 9 mm^2.

The functionality of the structure can be understood in terms of a generalized isotropic cut-wire plate combined with orthogonally oriented wires. Like the double-element MM, an anti-symmetric plasmonic eigenmode is excited at a given resonance wavelength in the cut-wires that are now merged in the Swiss-cross structure. Because of the structure′s fourfold rotational symmetry, a polarization-independent optical response at normal incidence is anticipated. Based on spectroscopic measurements in transmission and reflection we provide experimental evidence of this property (Fig. 2(c) and (d)). The measured experimental data is confirmed by comparison to rigorous calculations (Fig. 2(e)). Again, we assign effective permeability, permittivity and consequently a refractive index n to our Swiss-cross structure. For the fabricated sample a value of $n = -1.9 + 2.7\mathrm{i}$ at the resonance wavelength around 1.4 μm is deduced (Fig. 2(f)). The design parameters are chosen primarily because of the experimental constraints imposed by our setup and are not meant to be optimized. The Swiss cross improves a particular aspect of the fishnet design as it eliminates the drawback of a polarization-dependent optical response. The free design parameters are the width x_s and the length x_l of the cross arms and the thicknesses d_{Au} and d_{MgO} of the thin film layers. By changing these values the negative-index domain could be tuned to other wavelengths as well.

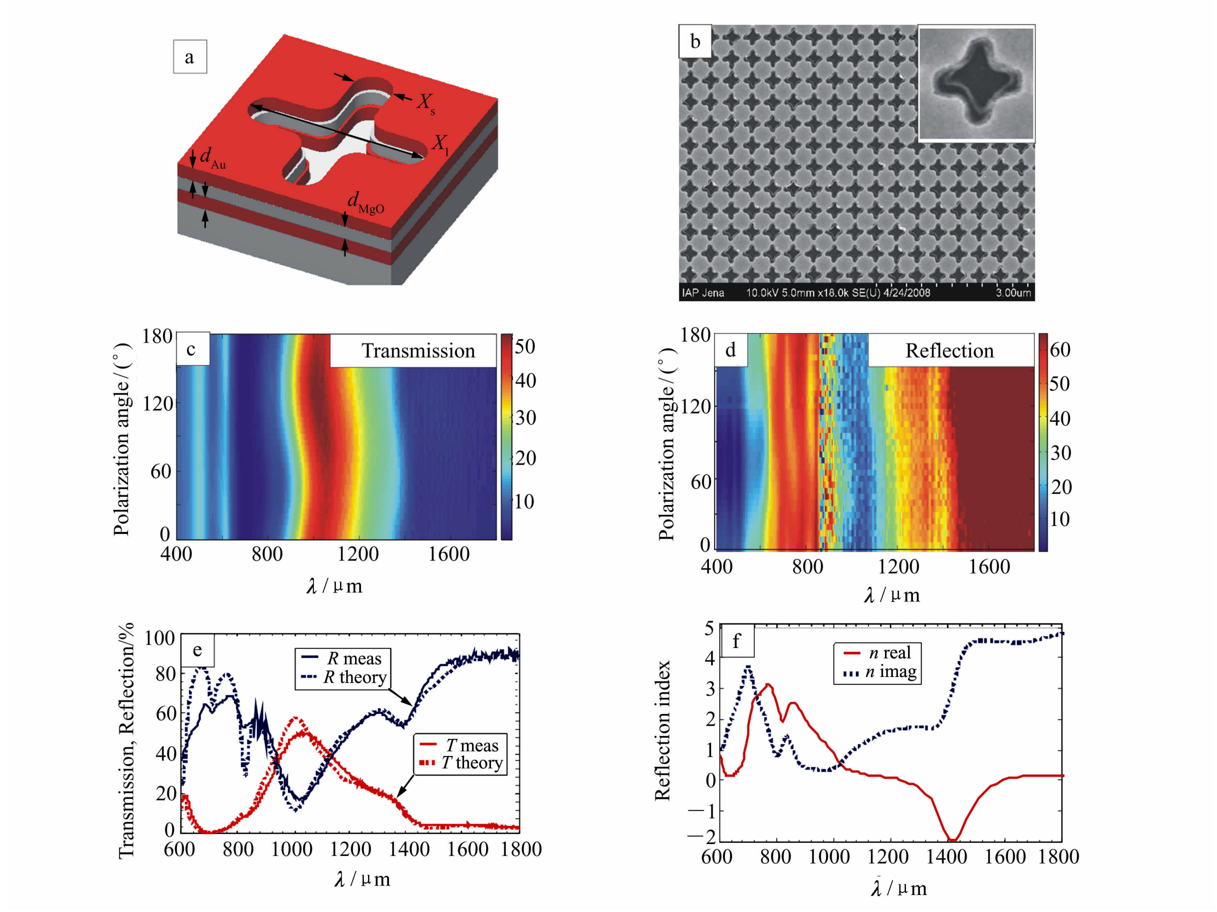

Fig. 2　(a) Schematic of the Swiss-cross MM unit cell with structural parameters, (b) normal view electron micrograph of a fabricated sample (inset: magnification of a unit cell), (c) measured transmission, (d) reflection spectra of the sampl for a complete set of linear polarization states from 0° to 180° in steps of 5° at normal incidence, (e) measured (solid lines) and calculated (dotted lines) transmission (grey) and reflection (black) for 0° polarization, (f) real (solid line) and imaginary (dotted line) part of the effective refractive index n derived from the spectra shown in (e).

Furthermore, we provide insight into the dependence of the effective MM properties of the Swiss cross on the angle of incident light both experimentally and theoretically[22]. The angular and spectral dependent response was measured using a self-built spectroscopic setup for specular transmission and reflectance. We take the azimuth angle ϕ, the elevation angle θ and the state of polarization to describe the plane wave normal. TE polarization implies that the incident E-field is tangential to the surface. To exclude the undesired effect of depolarization we consider the four symmetry directions of all possible combinations of $\phi = 0$, $\phi = 45°$, TE- and TM-polarization. In these cases no coupling can occur between the TE-like and TM-like polarized eigenstates of the effective MM, hence the polarization states of in- and out-coming waves are maintained. The resulting spectra were measured for their dependence on θ and λ and have been compared to the numerically simulated data. As an excellent agreement is observed, we can rely in future on the simulated data to retrieve the angular-dependent effective properties of the structure[12].

We note that the effective properties of the Swiss-cross structure suffer from strong spatial dispersion and consequently have to be understood as wave parameters rather than genuine material parameters. Any effective property looses its meaning if it has to be determined for every incidence angle and polarization state separately. For instance, it can be shown that the spectral and angular domains of the negative refractive index as well as its magnitude are closely connected to the propagation direction and the polarization state of the illumination. We can conclude for the given example of the Swiss-cross MM that its description as effectively homogenous and anisotropic is physically inappropriate[22]. However, we dissuade from abandoning the general description of MMs by effective properties at the present stage. Provided that the limits of their applicability are carefully borne in mind, angular resolved effective properties give preliminary insight into the underlying physics and can serve to simplify the description of light propagation inside a Swiss-cross MM.

4 Transition from periodic to amorphous MMs

Almost all fabricated optical MMs to date are composed of meta-atoms or unit cells arranged in periodic lattices. This has been shown to be convenient for numerical treatment since periodic arrangements greatly facilitate rigorous simulations by considering one single unit cell equipped with periodic boundary conditions. Here we provide an intuitive approach to lift the constraint of periodicity in optical MMs by investigating the transition form periodic to truly amorphous MMs[23]. Besides unraveling the significance of periodic arrangements, the quantitative investigation of disordered and amorphous MMs is usually regarded to be essential for the realization of isotropic MMs.

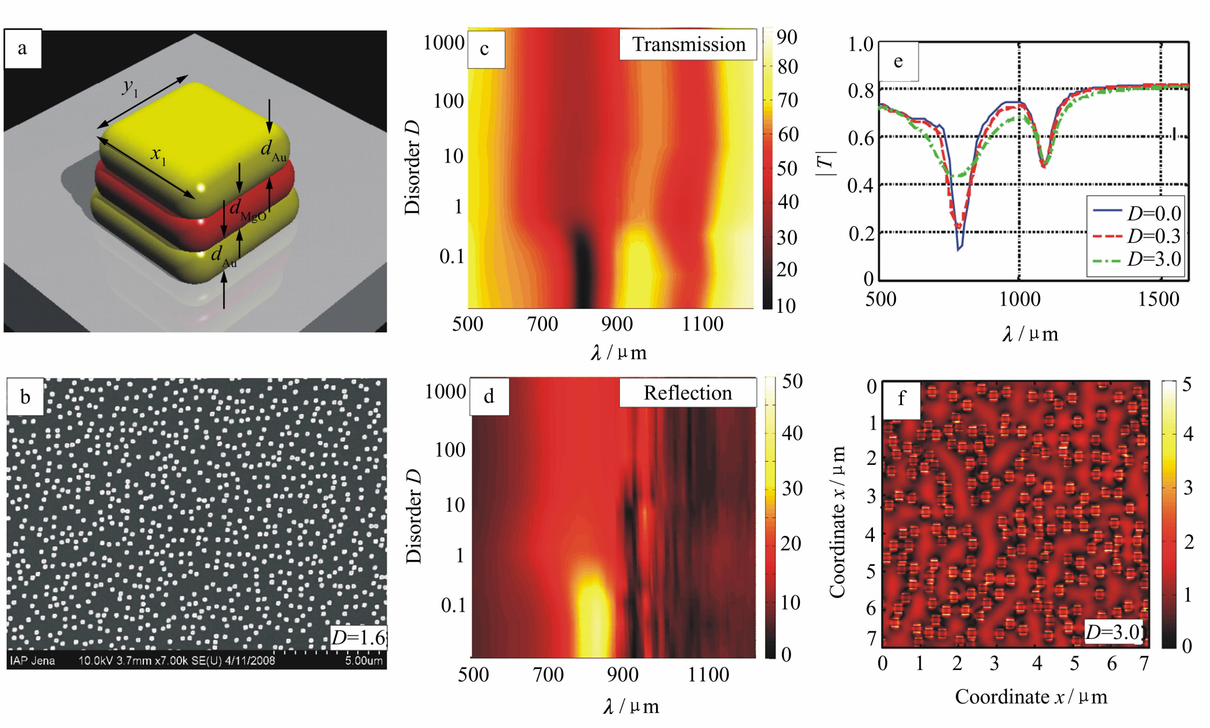

Fig. 3 (a) Schematic of the cut-wire MM unit cell with structural parameters, (b) SEM micrograph of a sample with disorder $D = 1.6$, (c) measured transmission and (d) reflection spectra as a function of the disorder parameter D. Both spectra are recorded for discrete values of D and interpolated to guide the eye, (e) Simulated transmission spectrum for three discrete values of D, (f) FDTD simulation of the absolute E-field amplitude in reflection for $D = 3.0$ at $\lambda =$ 1 050 nm.

The system we consider is based on the cut-wire pair MM[16]. Fig. 3(a) shows the principle design of the unit cell. Each cut wire pair consists of two gold layers with a thickness of $d_{\mathrm{Au}} = 30$ nm separated by a magnesia spacer with $d_{\mathrm{MgO}} = 45$ nm. The length of the wires is $x_1 = y_1 = 180$ nm and the lattice constant is $P_x = P_y = 512$ nm in the reference sample. Positional disorder is introduced by summing a random displacement to the centre position of each unit cell, independently in both lateral directions. Normalizing this displacement to the period $P_x = P_y$, we obtain an average dimensionless parameter D to quantize the degree of disorder in the system. Keeping the average density of cut-wire pairs and hence the average surface filling fraction constant, several MM samples with D increasing from 0 to 1 000 were fabricated. Fig. 3(b) shows one representative SEM micrograph of a sample with $D = 1.6$. The results of the spectral characterization for transmission and reflection are summarized in Fig. 3(c) and (d). As for the periodic reference sample we note two dips situated at $\lambda = 800$ nm and $\lambda = 1\ 050$ nm in the transmission and a peak at $\lambda = 800$ nm in the reflection spectrum. These two resonances are identified as the symmetric and anti-symmetric plasmonic eigenmodes of the cut-wire pairs. They evolve differently if the degree of disorder D is increased. While the anti-symmetric resonance at $\lambda = 1\ 050$ nm is almost independent of D, the symmetric resonance rapidly decays even for a moderate degree of disorder. This behaviour is confirmed by finite-difference time-domain simulations for a supercell of cut-wire pair MMs with no ($D = 0$), moderate ($D = 0.3$) and high ($D = 3.0$) positional disorder corresponding to a periodic, disordered and amorphous MM, respectively (Fig. 3(e)).

Our key finding is that the anti-symmetric resonance is nearly invariant to positional disorder. It is important to note that this resonance is the key feature in the majority of today's negative-index MMs. Based on a detailed investigation of the eigenmodes supported by near-field calculations for different values of D (Fig. 3(f)), we can explain this result. Basically we argue that the electric quadrupole associated with the anti-symmetric resonance does not have any in-plane component. Thus it makes the interaction among neighbouring particles negligible. Furthermore, with the claim of evaluating the effective properties of amorphous MMs for the first time, similar conclusions can be drawn. The resonance in the effective magnetic permeability that is related to the anti-symmetric eigenmode does not experience any noteworthy changes upon the transition from a periodic to an amorphous MM. Independent of the degree of disorder the line shape, the strength and the width of this resonance remain unchanged. It can be concluded that the magnetic response of any MM based on this particular eigenmode is solely determined by the response of the individual meta-atoms regardless of their arrangement. This is an important finding when it comes to the fabrication of MMs by self-organized approaches. The implications of our finding facilitate the integration of optical negative-index MMs in sub-wavelength imaging applications and relax the constraint of the necessity of periodical arrangements in modern MM designs. For instance, the effective properties of large-scale optical MMs fabricated by quick and reliable bottom-up approaches[19] can be evaluated by considering their periodic equivalents. Moreover, the influence of the structural parameters on the tunability of the effective properties of such amorphous MMs can be revealed[23].

5 Conclusions

Effective properties constitute an intuitive way to gain insight into light propagation in optical MMs. We have demonstrated how structural parameters can be employed to design and tailor the response of highly dispersive, optical MMs. We addressed this approach on the basis of three optical MM designs: the double cut-wire pair structure, the Swiss-cross structure and the amorphous cut-wire pair MM. Combining experimental and theoretical studies it was shown how the operational wavelength of an effective index of refraction smaller than zero and its sensitivity to polarization can be modified. Nevertheless, the valuable concept of effective parameters must always be used in the limits of its applicability due to the scaling of the characteristic lengths of the constituent meta-atoms and the wavelength of the interacting electromagnetic radiation. By retrieving angular-dependent effective properties we show through the example of the Swiss-cross MM that in the vicinity of the resonance with negative refraction it cannot be described as effectively homogeneous. Another way to look at the homogenization of optical MMs is to evaluate the necessity of a periodic arrangement of the unit cells. We investigated the transition from periodic to amorphous MMs and confirm that the opto-magnetic properties of common MMs are virtually unaffected by an arbitrarily high degree of positional disorder. This new degree of freedom in the design and fabrication of optical MMs opens further paths to tailor their effective properties according to the requirements imposed on them.

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