Scattering properties of meta-atoms

C. Rockstuhl,[1] C. Menzel,[1] S. Mühlig,[1] J. Petschulat,[2] C. Helgert,[2] C. Etrich,[2] A. Chipouline,[2] T. Pertsch,[2] and F. Lederer[1]
[1]*Institute of Condensed Matter Theory and Solid State Optics*
[2]*Institute of Applied Physics, Abbe Center of Photonics, Friedrich-Schiller-Universität Jena, Max-Wien-Platz 1, D-07743 Jena, Germany*
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Metamaterials consist of a periodic or aperiodic arrangement of so-called meta-atoms. Usually their optical properties are derived from the collective response of this ensemble. However, it is highly desirable to deduce them from the scattering properties of individual meta-atoms because frequently the periodic arrangement has a spurious effect on the desired functionality. Moreover, understanding the scattering properties of individual meta-atoms permits introducing guidelines for their design and predicting effective properties of amorphous metamaterials. To achieve this we introduce a genuine approach to quantify the properties of individual meta-atoms. To this end we evaluate spectrally resolved the composition of the rigorously calculated scattered field in terms of contributions of electromagnetic multipoles, such as electric and magnetic dipoles, quadrupoles and, in principle, arbitrary higher order moments. Beyond its direct application to metamaterial's design and characterization, the approach will be significant in the entire field of nanooptics as, for example, for optical nanoantennas.

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Metamaterials (MMs) composed of micro- or nanostructured materials emerged as a novel kind of artificial matter that allows affecting the light propagation in a manner inaccessible with natural media.[1] With the aim to observe dispersive regimes beyond those achievable by a mere averaging of its constituents, resonances are the key ingredient one aims to exploit. Two distinctive kinds of resonances are usually at the focus of interest. The first one requires a strict spatially periodic arrangement of the unit cells, for convenience called the meta-atoms. A resonance may be encountered if light scattered at consecutive periods interferes constructively. The structure acts then as a photonic crystal and it cannot be homogenized. Such a homogenization can be understood as one of the leitmotifs in the research on MMs.[2] Consequently, it requires that the resonance is associated with the meta-atoms itself and not with their arrangement. This is the second type of resonances to be exploited. Besides Mie resonances in dielectric particles,[3] localized plasmon polaritons allow observing such a resonant response. Accordingly, most of the current optical meta-atoms are metallic nanostructures.[4]

Nevertheless, despite such understanding, in most cases the properties of MMs are evaluated for periodically arranged meta-atoms. This simplifies both their fabrication and foremost their theoretical analysis. Concepts such as normal modes (Bloch modes) with the respective dispersion relation can be applied and dedicated numerical schemes may be exploited.[5] However, such approaches will not allow deriving the scattering properties of individual meta-atoms. But just this information is required to distinguish between properties induced by the lattice or by the meta-atoms themselves. In most cases this distinction is even pointless since it will be rather a combination of both. Dropping the periodicity and analyzing the properties of individual meta-atoms is presently urgently required for two reasons.[6]

At first, it would allow revealing whether the involved optical response that prohibits the assignment of effective material properties to MMs is either due to the periodic arrangement of meta-atoms or to the excitation of higher order electromagnetic multipoles in the meta-atoms.[7] The use of simple constitutive relations requires that the scattering properties of the individual meta-atom are evoked by electric and magnetic dipoles, induced along the polarization of the incident field, and an electric quadrupole.[8] The latter may introduce certain ambiguities since it may be accounted for in the wave equation as either a spatially dispersive permittivity or an effective permeability.[9,10]

Second, self-organized MMs that are fabricated by bottom-up approaches are emerging as a promising option to get rid of cost intensive top-down techniques which are largely restricted to a two-dimensional patterning of thin films, making it practically impossible to fabricate bulk MMs.[11,12] Although stacking of such functional layers may extend the options of top-down technologies,[13] it cannot be regarded as an ultimate route toward bulk MMs. Metamaterials fabricated by bottom-up approaches might be potential candidates for future bulk MMs.[14] However, the fabrication of perfectly periodically arranged meta-atoms is unlikely to be achieved with such methods which provide only a short-range order of the meta-atoms. The assignment of effective properties to such fully disordered MMs constitutes a challenging task. It can potentially be achieved by analyzing the scattering properties of individual meta-atoms in terms of electromagnetic multipole contributions and a succeeding incorporation of the actual dipole moments into averaging procedures that do not require a certain spatial arrangement.

Here we aim at providing the theoretical and numerical means for the spectrally resolved expansion of the scattered field of meta-atoms into multipole contributions. We furthermore demonstrate that the strength of the electric and magnetic dipole moments can be exploited as an explicit criterion for the design of meta-atoms where undesired electromagnetic multipole moments may be fully suppressed. We finally show that effective properties of amorphous MMs can be retrieved with sufficient precision using such a multipole expansion.

To outline the approach we start the analysis with a referential meta-atom, the split-ring resonator (SRR). At first we calculate spectrally resolved the scattered field from the SRR upon plane wave illumination [Fig. 1(a)]. Illumination

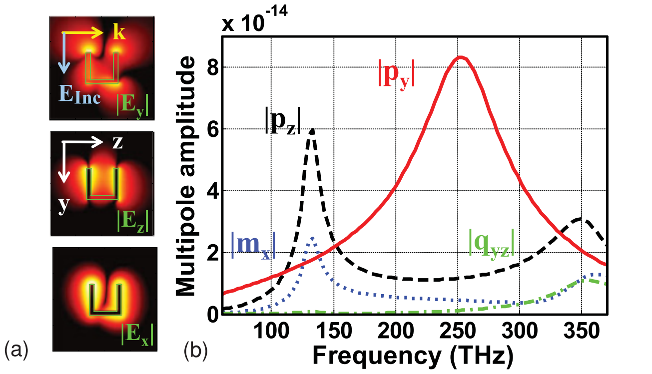

FIG. 1. (Color online) (a) Electric components of the field scattered by an individual meta-atom at $x = 10$ nm in the y-z plane upon plane wave illumination. (b) Moduli of the dominant electromagnetic multipole contributions to the scattered field.

direction and polarization are chosen in accordance with the sketch in Fig. 1. Both the SRR arm and base lengths were 300 nm and the wire that forms it has a geometrical cross section of 40 × 40 nm. The SRR consists of gold surrounded by air. Using the finite-difference time domain (FDTD) method[15] the simulations were performed at each frequency individually to take into account the documented material dispersion.[16] The computational domain was entirely surrounded by perfectly matched layers. For illustration, selected field distributions for all three polarization components at a frequency of 132 THz in a selected cross section are shown in Fig. 1(a).

To extract the amplitude of all electromagnetic multipole contributions, the field was expanded in terms of vector spherical harmonics ($\mathbf{N}(\mathbf{r},\omega)$, $\mathbf{M}(\mathbf{r},\omega)$) at the surface of a virtual sphere containing the entire SRR,[17]

$$\mathbf{E} = \sum_{n=1}^{\infty} \sum_{m=-n}^{n} a_{nm}\mathbf{N}_{nm}(\mathbf{r},\omega) + b_{nm}\mathbf{M}_{nm}(\mathbf{r},\omega). \quad (1)$$

The expansion is identical to that in Mie theory and, except for some prefactors, it corresponds to a multipole expansion in spherical coordinates.[17] The expansion coefficients a_{nm} and b_{nm} are calculated by solving the overlap integral with the scattered field as simulated by the FDTD and the normalized multipole modes. In the expansion the lowest order corresponds to a dipole, the second order to a quadrupole, and so on. Thus, contrary to other existing approaches here it is possible to simultaneously reveal all complex multipole coefficients in spherical coordinates.[18] This is essential if effective properties shall be assigned to the medium. The origin of the expansion was chosen to be the center of the square as formed by the SRR with the same dimensions. It is clear that the expansion coefficients will depend on the respective origin. Nevertheless, this does not affect the conclusions to be drawn.[19] A comparison of the multipole expansions in Cartesian and spherical coordinates allows one to transform the expansion coefficients in Eq. (1) into Cartesian multipole coefficients. Such multipolar coefficients could have been retrieved as well directly from the current.[20]

Results of the expansion for the SRR can be seen in Fig. 1(b). The moduli of the contributions of the most dominant multipoles are shown in the frequency interval where the SRR sustains its three lowest order eigenmodes. Clearly, the scattered field of the lowest order resonance at 132 THz is related to a magnetic dipole (m_x), that is, the SRR acts as an artificial meta-atom showing a strong magnetic response. Contrary to previous observations it turns out that the scattered field at this resonance frequency is also dominated by the contribution of a z-polarized electric dipole represented by an oscillating current in the base wire. Since it does not radiate into forward or backward direction upon arranging the SRRs in a lattice, it is not possible to probe for its existence in a subwavelength SRR lattice. Nonetheless, for randomly arranged SRRs as currently pursued,[12] the presence of the z-oriented dipole needs to be considered and suggests much more involved constitutive relations. The second resonance at 250 THz is dominated by an electric dipole that is oriented parallel to the incident electric field. The scattered field at the third order resonance at 350 THz is composed of contributions from various multipoles. Only the strongest are shown here, though a few other higher order multipoles have non-negligible amplitudes as well. At this frequency the SRR cannot be mimicked by a magnetic or electric dipole, hence it is unlikely that a MM made of SRRs can be homogenized at these frequencies.

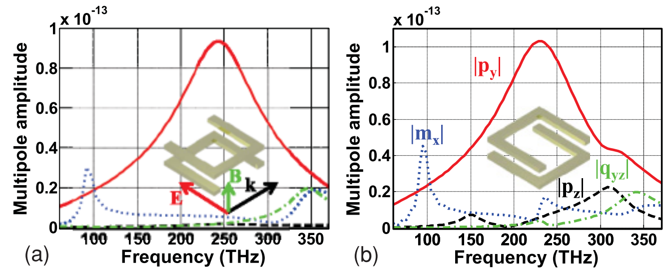

FIG. 2. (Color online) Amplitudes of the dominant electromagnetic multipole contributions that form the scattered field for a meta-atom made of two SRRs. In (a) the two SRRs exhibit an out-of-plane separation of 40 nm; in (b) the two SRRs are separated in-plane by 40 nm.

To suppress the undesired excitation of higher order multipoles, which may appear in addition to the electric and magnetic dipoles aligned in illumination direction, meta-atoms can be appropriately composed of various functional elements which can be suitably arranged. The multipole coefficients on their own may serve as an explicit design guide. Here, some obvious examples are shown which clearly demonstrate the strength of the approach. Most notably, to suppress the electric dipole coefficient in the longitudinal (z) direction of the lowest order resonance, a meta-atom can be considered that consists of two SRRs but with gaps on opposite sides. This approach of nesting SRRs resembles ideas established in GHz or radio frequency domain.[22] Two possible arrangements (out-of or in-plane separation) are proposed and the moduli of the contributions from the excitable multipoles are shown in Fig. 2 along with the SRRs arrangement. Again, only the moduli of the most dominant contributions are displayed.

At the expense of additional higher order multipolar resonances at higher frequencies, the z-polarized electric dipole coefficient at the lowest order resonance is suppressed. The scattering response of the meta-atom comprising nested

SRRs at the two lowest order resonances is dominated, as desired, by a single electric and a single magnetic dipole at normal incidence. This seems to be favorable to achieve dispersion in permittivity and permeability upon amorphously arranging the meta-atoms. It should be mentioned that the multipolar excitations are only probed at normal incidence here. Thus any *ad hoc* conclusions regarding the excitation strength of the multipoles for deviating illumination conditions are impossible.

The possibility to predict effective properties of amorphous MMs is shown in the following. Such amorphous MMs are characterized by a lack of periodicity in meta-atom arrangement.[23,24] To allow yet for a comparison between effective properties as retrieved by established methods as well as having a direct experimental implementation in mind, in a first step we impose here two further restrictions which will be dropped later for genuine bottom-up MMs. At first, and in agreement with previous arguments, all meta-atoms are assumed to have an identical orientation. Second, the meta-atoms are required to be equally spaced in the principal propagation direction; hence disorder manifests itself only in the lateral dimensions. These assumptions make the analysis suitable for an experimental realization using stacking technologies where subsequent functional layers are defined in serial.[25,26] Moreover, the equal separation suppresses longitudinal coupling in excess which would lead to an undesired splitting of resonances. But to be more general we show below that these constraints can also be lifted and that the performance of the method is preserved in predicting the effective properties of a truly amorphous MM. The limitations imposed here at first simply allow a clear and unambiguous analysis since the system is well defined.

The meta-atom we consider consists of cut-plate pairs made of Au-MgO-Au [see Fig. 3(a)]. The lateral dimensions are 180 nm. The thickness of gold and MgO layers was 30 and 45 nm, respectively. The surrounding medium is air. The moduli of the dominant multipole coefficients are shown in Fig. 3(b). Clearly, at the low frequency resonance, corresponding to an antisymmetric localized plasmon polariton in the adjacent cut-plates, a strong magnetic dipole is excited, albeit an electric quadrupole as well. At the high frequency resonance a pure electric dipole is excited.[27] To extract the effective properties of a medium made of randomly arranged meta-atoms, the Clausius-Mosotti formula is applied.[28] It links the effective properties to the respective dipole moments by

$$\epsilon_{\text{eff}} = \frac{3 + 2V\alpha_e}{3 - V\alpha_e}, \quad \mu_{\text{eff}} = \frac{3 + 2V\alpha_m}{3 - V\alpha_m},$$

with $\alpha_{e/m}$ being the polarizabilities that are obtained by normalizing the dipole moments p_y and m_x to the dipole moments of the incident field. The volume filling fraction V enters as the only free parameter. In order to reproduce effective properties as retrieved from the rigorously calculated reflection and transmission data in a referential simulation, the volume filling fraction had to be set a factor of 1.3 larger than it actually was. This referential simulation of such a slab and the way the random arrangement was generated is identical to that described in Ref. 21, with the only exception that four amorphous functional layers with different implementations of nominally identical disorder were stacked to get the

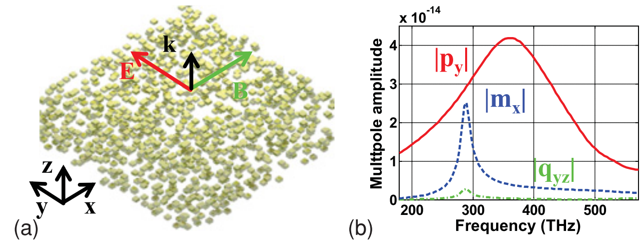

FIG. 3. (Color online) (a) Schematic sketch of an amorphous MM that is composed of four stacked layers of randomly arranged cut-plate pairs. (b) Moduli of the dominant multipols.

bulk properties.[29] The spatial extension along the principal propagation direction of a single functional layer was chosen to be 150 nm and the meta-atoms were placed in a central plane. A sketch of such a MM is shown in Fig. 3(a). Effective properties for the slab at normal incidence were assigned by inverting reflection and transmission coefficients.

For completeness, the moduli of the reflection and transmission coefficient as simulated with the FDTD are shown in Fig. 4. It can be seen that in the transition from a periodic to an amorphous layer, the resonance becomes broader and weaker but the resonance frequency itself persists.[21] For the stacked structure with the design as described above, no further significant modifications emerge except an overall reduction of transmission. This, however, is natural due to losses inside the structure, being both radiative and non-radiative. A further broadening of the resonance can also be noticed but otherwise all spectral features remain to be preserved and the optical properties of the bulk may be already essentially predicted from the single layer data.

Effective properties as retrieved both from the complex reflection and transmission coefficients of the slab and from the electric and magnetic dipole coefficients of the individual meta-atoms are shown and compared in Fig. 5. Good agreement is observed both with respect to resonance positions and the amplitudes. For the magnetic resonance the linewidths are also identical. The only deviating feature is the slightly increased linewidth of the electric resonance for the scattered field approach. A strong change in the linewidth of that effective property was already observed at the transition from a periodic to an amorphous arrangement in a single functional layer and it can be anticipated that the broadening is even stronger for a fully amorphous arrangement also in the third dimension.[21] However, we may safely draw the conclusion

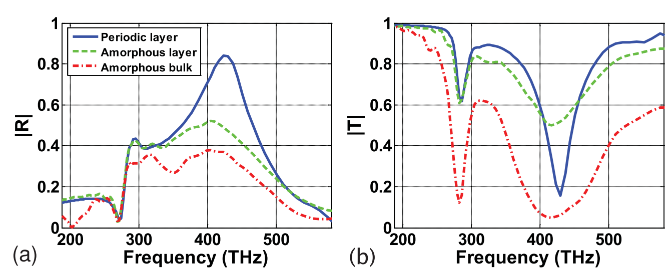

FIG. 4. (Color online) Moduli of the reflection (a) and transmission (b) coefficients at the MM made of cut-plate pairs arranged periodically or amorphously in a single layer and amorphously in an equally separated four-layer stack (bulk).

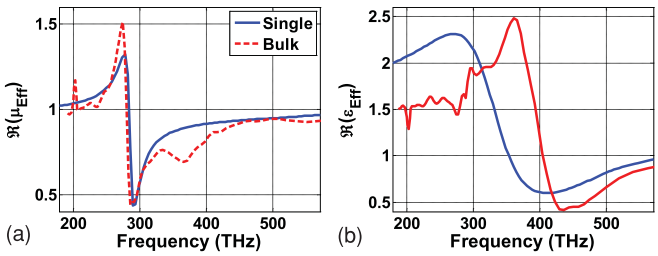

FIG. 5. (Color online) Effective properties as retrieved from the reflection and transmission coefficients of a slab of the MM (red dashed line) and as retrieved from the electric and magnetic dipole coefficients (blue solid line).

that the analysis of the scattered field of a single meta-atom can be used to predict effective properties of amorphous MMs with satisfying precision.

It remains to be mentioned that this peculiar kind of meta-atom arrangement suppresses coupling effects between meta-atoms in adjacent MM layers. However, just these coupling effects frequently evoke interesting effects and promise many applications,[25,26] but on the other hand, they prevent the MM homogenization over a larger spectral domain. This can be recognized from Fig. 6, where the retrieved real and imaginary part of the effective permeability are shown for both a single layer and a four-layer stack of strictly periodically arranged meta-atoms. As can be clearly seen, the tight arrangement of adjacent layers leads to coupling and hybridization of plasmonic excitations in individual functional layers. New resonances emerge at other frequencies consistent with the predictions of hybridization theory. These resonances occur neither in a single meta-atom nor in a single layer of them. The dispersion in the effective properties is strongly affected by such coupling and the resulting effective parameters are not meaningful since they result from a nonlocal response, a response that is induced between adjacent layers. Disorder in the meta-atom arrangement in the individual layers greatly suppresses these interactions since this arrangement is independent in each layer. It has to be stressed that also for an entirely periodic arrangement the effective properties as retrieved from a single layer can correspond to those of multiple layers, but only if the distance of adjacent layers is sufficiently large. However, this enhanced separation decreases the volume filling fraction and thus accordingly the dispersion in the effective properties. This leads to the conclusion that

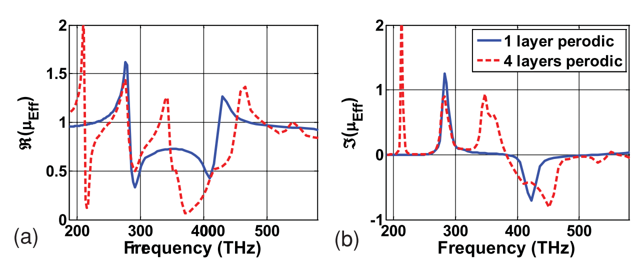

FIG. 6. (Color online) Effective properties as retrieved from the reflection and transmission coefficients of a slab of the MM made from periodically arranged meta-atoms (blue solid line) and as retrieved from four layers of the periodically arranged meta-atoms (red dashed line).

an amorphous meta-atom arrangement permits MMs with a larger volume filling fraction.

Finally we demonstrate that the present approach is also valid if the restrictions regarding the meta-atom arrangement (identical orientation, identical longitudinal spacing) will be dropped (i.e., the MM may be considered as being fully amorphous). To be specific, the MM is now assumed to be made of the same cut-plate pairs but randomly located with a random orientation. To allow for a comparison the number of meta-atoms and the thickness of the film is kept unchanged compared to the four-layer structure discussed above. Only a minimum separation between adjacent meta-atoms as above was enforced to avoid near-field coupling in excess which would lead to a modification of the individual plasmonic properties.

Figure 7(a) shows the moduli of the reflection and transmission coefficients as calculated by FDTD. Most notably, it can be seen that the functional dependency of transmission and reflection is entirely preserved (compare with Fig. 4), however, the strength of the resonances is weaker. This is not surprising since the meta-atom in the quasistatic limit is uniaxial anisotropic and for a random arrangement it cannot be assured that the linearly polarized illuminating plane wave always excites the resonance with maximum amplitude. This is only possible for a favorable orientation of the individual meta-atoms as enforced before. It remains to be mentioned that the third axis of the meta-atom also supports plasmonic resonances. But they occur at much higher frequencies and are of no relevance to the present study.

In the effective properties, shown in Figs. 7(b) and 7(c), we nicely identify the dielectric resonance at approximately 370 THz, where the imaginary part peaks. The induced magnitude in the dispersion is equally slightly weaker when compared to the effective properties for the layered amorphous MM (see Fig. 5). The same observation holds for the effective permeability which sustains again a Lorentzian resonance at the same frequency as before (approximately 270 THz), although at a slightly weaker strength. The reduced strength was safely anticipated since again, not all meta-atoms have the favorable orientation to get a sufficiently strong excitation of the relevant resonance. The only deviating feature which cannot be explained from considering the scattering properties of the individual meta-atom is the antiresonance in the permeability, occurring at the resonance frequency of the permittivity. Nevertheless, and this is important. the functional dependency of the effective properties remains to be largely preserved even for this completely amorphous arrangement.

In perspective and in order to get full access to bottom-up MMs, the calculation of the scattering response upon plane wave illumination at normal incidence does not suffice and more sophisticated illumination scenarios must be considered as well. The ultimate goal has to be to retrieve the entire T matrix of such meta-atoms.[30] Such approach will certainly pave the way toward a new class of bulk MMs fabricated with bottom-up self-organized chemical or biological methods. Moreover, other fields of related research will benefit from such an analysis as well. Examples would be in the design of optical nanoantennas which shall be used to couple

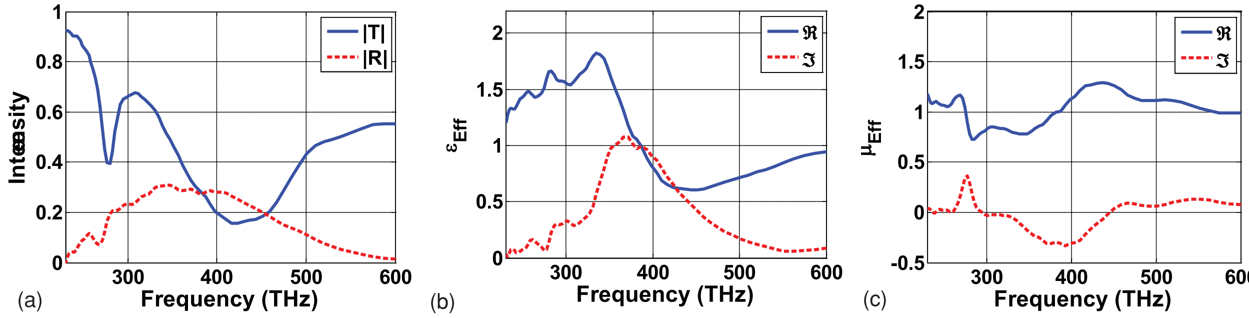

FIG. 7. (Color online) (a) Moduli of the reflection and transmission coefficients of a 600-nm thick, fully amorphous MM slab. It consists of both randomly placed and oriented cut-plate pairs where only a minimal separation between adjacent meta-atoms was enforced. In (b) and (c) the real and imaginary part of the effective permittivity and permeability as retrieved from an inversion of the scattering data are shown.

light (e.g., from localized dipolar scatterers into the far field). With the presented method, the tailoring of optical nanoantennas with a predefined mode content will come into reach.

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