Optical activity in chiral media composed of three-dimensional metallic meta-atoms

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We show that the chirality of artificial media, made of a planar periodic arrangement of three-dimensional metallic meta-atoms, can be tailored to cause a strong optical activity. The meta-atoms support localized plasmon polaritons and exhibit a chirality exceeding that of pseudoplanar chiral metamaterials by an order of magnitude. Two design approaches are investigated in detail. The first is the canonical example for a chiral structure, namely a Möbius strip. The second example is a cut wire-split-ring resonator geometry that can be manufactured with state-of-the-art nanofabrication technologies. Driven into resonance these meta-atoms give rise to a polarization rotation of 30° per unit cell.

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Metamaterials are artificial structures usually composed of periodically arranged unit cells that allow the control of light propagation.[1] Frequently, the rigorous description of light propagation in such media on the basis of the dispersion relation of the respective eigenmodes (Bloch functions[2]) can be simplified by treating the medium as effectively homogenous.[3] The properties usually at the focus of interest are permittivity and permeability or refractive index and impedance. However, to obtain more complex optical functionalities, efforts have been undertaken to extend this concept toward other optical properties, as, for example, chirality.[4]

By definition, a structure is termed *chiral* if the unit cell cannot be mapped onto its mirror image by proper rotations. Consequently, only a bulk medium with three-dimensional (3D) unit cells can exhibit this property. Chiral media attract much interest because the optical response of these structures is different for right-handed and left-handed circularly polarized light. Thus, these media are optically active and the state of polarization of light changes upon traversing such media. The observation of this phenomenon by using appropriately structured metallic thin films on substrates sparked significant research interest on artificial chiral media.[5–9] These structures were termed planar chiral metamaterials (PCMs) as, at a first glance, the unit cell is a thin film only with no structural variation in the propagation direction. An ensemble of gammadions constitutes one geometry of such a PCM. Although the term PCM is an oxymoron, one usually argues that the presence of the substrate breaks the mirror symmetry and saves the three-dimensional character of the unit cell.[10] However, the optical activity of these tiny pseudo-PCMs is small, leaving much space for further studies. To date potentially the largest optical activity was observed for a gammadion bilayer where the gammadions in subsequent layers are rotated by 15° with respect to each other.[11] Although the rotation per unit cell of this pseudo-PCM at the resonance wavelength was orders of magnitude larger than that of any naturally available substance, it is still rather small amounting to 0.37°. The questions arise whether there are feasible approaches toward larger optical activity and which enhancement can be achieved if the pseudo-PCMs are replaced by appropriately designed 3D unit cells. It is the aim of this paper to disclose effective design principles and to investigate the performance of this unique class of 3D chiral meta-atoms.

Before further discussions, we stress that the chirality explored here is related to the symmetry of the unit cell rather than to an appropriate rotation of adjacent crystal planes, as, for example, naturally observed in quartz or in cholesteric liquid crystals. In the latter structure, being potentially the most optically active naturally available substances, optical activity can be as large as $3\times10^{4}\,°/\mathrm{mm}$.[12] Such structures can also be mimicked by fabricated spiral type photonic structures[13,14] or chiral sculptured thin films (CSTFs).[15,16] Also stacked pseudo-PCMs where coupling between subsequent layers is explicitly evoked were recently optimized using genetic algorithm, and a polarization rotation of up to 21° was observed.[17] Moreover, the final aim will be to construct an effective chiral metamaterial from these meta-atoms. Therefore, we exclude geometries that show the desired optical response exclusively for a specific angle of incidence, a specific incident polarization state, or even in a higher diffraction order.

The design rules for the 3D chiral meta-atoms that we suggest follow simple physical principles. Large optical activity requires the meta-atom to exhibit two resonances in two different elements of the meta-atom. The first resonance should be excitable by the incoming light, providing sufficiently strong coupling between the external field and the meta-atom. The second resonance should appear for orthogonally polarized light in another element of the meta-atom serving to generate a radiated field that is orthogonally polarized with respect to the incident field. Both structural elements have to be sufficiently strongly coupled to allow for an efficient polarization conversion. The absence of any mirror symmetry in the unit cell ensures that coupling between the two modes sustained by the structure is not prohibited due to symmetry constraints. Within the quasistatic approximation this leads to the conclusion that the meta-atom has to be composed of two resonant structural elements mimicking a dipole type scattering response. These two elements can be either two electric dipoles or an electric and a magnetic dipole. A structural element having an electric-dipole resonance is, for example, a metallic wire of finite length[18] whereas a split-ring resonator (SRR) (Ref. 19) exhibits a magnetic-dipole resonance.

We will study meta-atoms that follow either design principles. This approach toward the understanding of optical activity of meta-atoms provides an extremely intuitive explanation of the fundamental physical effect, as will be shown below. In this context it might be interesting to apply this approach also in other fields, as, e.g., in chemistry, for understanding chirality and optical activity of various naturally available molecules and macromolecular structures.[20] The meta-atoms we consider have a 3D geometry. In building a genuine 3D material, they can be periodically arranged in succeeding x-y planes without any twist between adjacent planes. Thus optical activity based on that twist is safely excluded. Because the meta-atom does not exhibit any mirror symmetry, optical activity is expected to occur for any propagation direction and its strength is only determined by the particular geometrical arrangement within the unit cell. Without loss of generality, we assume the light to propagate in z direction.

The asymmetry of the unit cell requires a general bianisotropic description of the effective medium composed of the meta-atoms.[21] Hence, the rotation of the polarization is a consequence of the joint action of optical activity and birefringence of the structure. Both effects may be distinguished if the polarization rotation is averaged over all possible linear polarization states of the incident light. The averaged rotation is then a measure of optical activity and chirality of the meta-atoms. This is intuitively clear if a statistical ensemble of meta-atoms, arbitrarily rotated around the z axis, is considered where a light field experiences a bi-isotropic rather than a bianisotropic medium.

To validate our concept we study light interaction with a planar periodic arrangement of two different 3D meta-atoms. The optical activity of the first, a Möbius strip[22] [see Fig. 1(a)], relies on the interaction of two resonant electrical dipoles, and the second, a rationally designed cut wire-split-ring geometry [see Fig. 2(a)], takes advantage of the resonant interaction of an electric with a magnetic dipole.

The terminating surface of the Möbius strip is characterized by the parametric equations

$$x(u,v) = \frac{v}{2}\sin\left(\frac{u}{2}\right),$$

$$y(u,v) = \left[1 + \frac{v}{2}\cos\left(\frac{u}{2}\right)\right]\cos(u),$$

$$z(u,v) = \left[1 + \frac{v}{2}\cos\left(\frac{u}{2}\right)\right]\sin(u), \qquad (1)$$

with $0 \le u \le 2\pi$ being an angle, and $-1 \le v \le 1$ being the normalized radius and the width of the strip. In the numerical analysis we assumed a gold strip (dielectric function from Ref. 23), where the radius and the width are identical (166 nm) and the thickness is 33 nm, surrounded by air. The Möbius strips are periodically arranged in the x-y plane with a period of 500 nm in both directions. The chosen geometry ensures that the meta-atoms are subwavelength in the spectral domain of interest. Only a zero-order reflected and transmitted wave will be observed. The orientation of the strip

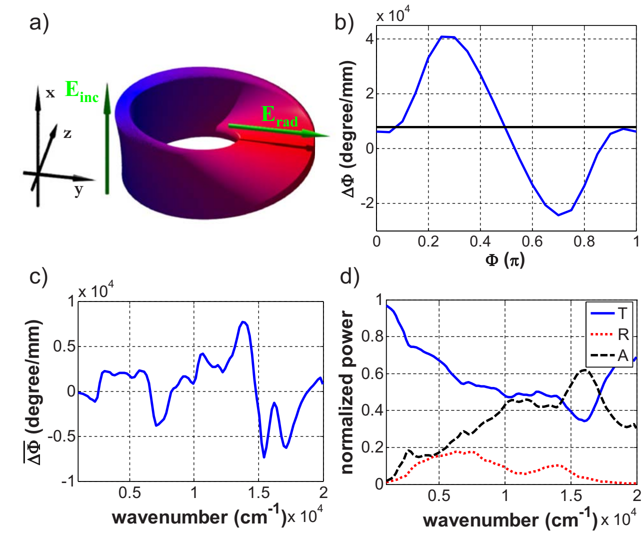

FIG. 1. (Color online) The Möbius strip. (a) Geometry and coordinate system; the arrows indicate the polarization of the electric-dipole resonances. (b) Polarization rotation as a function of the polarization angle of the incident field at $\nu = 1.373 \times 10^4$ cm^{-1}. (c) Averaged polarization rotation as a function of the wave number. (d) Averaged reflection, transmission, and absorption as a function of the wave number.

with respect to the axis as well as the exact geometrical parameters is chosen arbitrarily to a certain extent and shall only serve to illustrate the described approach. We do not intend to optimize the chiral response. Nonetheless, the particular orientation was chosen because certain segment of the strip resemble cut wires in both the x and y directions. As

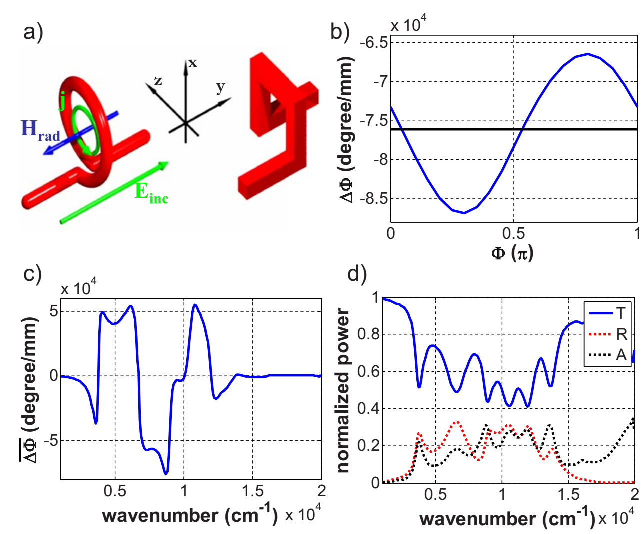

FIG. 2. (Color online) The omega particle. (a) Geometry and coordinate system; left: modified omega particle; right: cut wire-SRR particle used in numerical simulations. The arrows indicate the polarization of the fields associated with the electric and magnetic dipoles. (b) Polarization rotation as a function of the polarization angle of the incident field at $\nu = 0.87 \times 10^4$ cm^{-1}. (c) Averaged polarization rotation as a function of the wave number. (d) Averaged reflection, transmission, and absorption as a function of the wave number.

both wires are orthogonally oriented, we expect that both segments support the excitation of a localized plasmon polariton with an electric-dipole field at a certain frequency although they require an orthogonal polarization for their excitation. The excited electric dipole and the radiating electric dipole are indicated in Fig. 1(a) by arrows.

Since the system is linear, it suffices to calculate the transmission coefficients for two mutually orthogonal incident polarizations, say x and y polarizations. The transmission coefficients T_x and T_y for arbitrary linear polarization characterized by the polarization angle $\phi \in [0, 2\pi)$ of the incident light are given by

$$\begin{pmatrix} T_x \\ T_y \end{pmatrix} = \hat{T} \begin{pmatrix} I_x \\ I_y \end{pmatrix} = \begin{pmatrix} T_{xx} & T_{yx} \\ T_{xy} & T_{yy} \end{pmatrix} \begin{pmatrix} \cos(\phi) \\ \sin(\phi) \end{pmatrix}, \qquad (2)$$

where the matrix $\hat{T}$ is obtained by the two calculations mentioned before.

The average polarization rotation $\overline{\Delta\Phi}$ is then given by

$$\overline{\Delta\Phi} = \frac{1}{2\pi} \int_0^{2\pi} \Delta\Phi(\phi) d\phi, \qquad (3)$$

where the polarization rotation $\Delta\Phi(\phi)$ is given by[24]

$$\Delta\Phi(\phi) = \Re\left\{ a \tan\left(\frac{T_\perp(\phi)}{T_\parallel(\phi)} \right) \right\}, \qquad (4)$$

with $T_\parallel$ denoting the transmitted amplitude parallel polarized to the incident field and $T_\perp$ denoting the transmitted amplitude perpendicular polarized to the incident field.

The complex transmission coefficients have been computed by using the Fourier modal method (FMM).[25] Overall in the Fourier expansion we have retained 21×21 orders. The number is sufficient to ensure that all numerical results presented herein are converged.

For a wave number of $\nu = 1.373 \times 10^4$ cm^{-1}, the polarization rotation as a function of the polarization angle of the incident light is shown in Fig. 1(b). As usual in the field of chiral metamaterials, the polarization rotation is measured in angle per mm. The average value of the polarization rotation is indicated by a straight line and amounts to $\overline{\Delta\Phi} = 7.711 \times 10^{3\,\circ}$/mm or 3.5° per meta-atom layer. This polarization rotation is evoked by the chirality of the metamaterial and is comparable to that of common PCMs. The averaged polarization rotation as a function of the wave number is shown in Fig. 1(c). Because the structure always meets the symmetry requirements (two orthogonal cut wires), optical activity is present in the entire spectral domain. The strength depends on the respective resonance strength, which depends critically on the wave number. A pronounced maximum exists around $\nu = 1.373 \times 10^4$ cm^{-1}. The angular averaged and spectral dependent optical coefficients (transmission, reflection, and absorption) are shown in Fig. 1(d). Spectral regions of high absorption correspond to spectral regions of high optical activity. This supports our initial statement that strong optical activity is linked to the excitation of a localized plasmon polariton resonance. Nonetheless, it remains noteworthy that even in the spectral domain of the strongest absorption the transmission is as large as about 40%.

The second design approach for a chiral meta-atom uses the combination of structural unit elements that support the excitation of an electric dipole and a magnetic-dipole resonance. The former is again a finite metallic wire element, whereas the latter is provided by a SRR. The idealized geometry we envision is shown in the left of Fig. 2(a).

To a certain extent the structure resembles an omega particle although the metallic wires are perpendicularly arranged with respect to the SRR plane. This structure was already studied in the context of metamaterials but only with regard to controlling the permittivity rather than its optical activity.[26] The interaction of the light with the meta-atom can again be best understood in terms of elementary electromagnetic excitations. A y-polarized incident electric field induces at first an oscillating current in the wire. At the plasmon polariton resonance the charge-density oscillation is in resonance. As the SRR is coupled to the wire, a current will also flow through the wire forming the SRR. The oscillating current will induce a magnetic field that is perpendicular to the SRR from which it will radiate. The radiated magnetic dipole has a dominant electric-field component perpendicular to the incident polarization. This field will cause a strong rotation of the polarization, hence maximizing optical activity of the meta-atom.

Because the fabrication of such a highly curved structure would be seemingly difficult at optical frequencies, we suggest and study a slightly simplified variant [right section of Fig. 2(a)] that maintains, however, all structural features. Basically all curved elements were replaced by their rectangular counterparts. In the subsequent numerical analysis, the cross section of the wire amounts to 50×50 nm^2. The length of the nanowires is 200 nm and the length of the SRR side arms as well as of the SRR base is 300 nm. The SRR is again made of gold, and the period in both the x and y directions is 500 nm.

The polarization rotation as a function of the polarization angle of the incident field at a wave number of $\nu = 0.87 \times 10^4$ cm^{-1} is shown in Fig. 2(b). Two features are important to note. Optical activity is larger by an order of magnitude when compared to the Möbius strip and the structure exhibits only a marginal anisotropy. The averaged polarization rotation is large and varies only slightly (about 1/5 of the averaged rotation) with the polarization angle of the incident field. Note that the variation for the Möbius strip was about four times the averaged value. The average polarization rotation at the pertinent wave number amounts to $\overline{\Delta\Phi} = 7.609 \times 10^{4\,\circ}$/mm; or 30.43° per meta-atom layer. The spectrally dependent average polarization rotation is shown in Fig. 2(c). It shows well pronounced resonances where it is significantly enhanced within spectrally narrow domains. The position of the resonances can be unambiguously correlated with resonances in the absorption or the transmission, shown in Fig. 2(d). It is likewise averaged over all possible linear polarization states of the incident field. Near all resonances steep changes occur for the polarization rotation. There, either the cut wires, the SRRs, or both support the excitation of a localized plasmon polariton. However, it is pointless to study in detail the isolated resonances. One should rather regard the meta-atom as a single complex structure that allows the excitation of localized plasmon polaritons with vari-

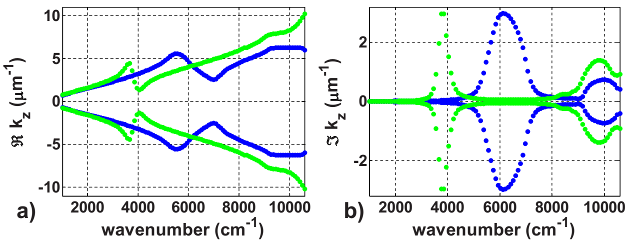

FIG. 3. (Color online) (a) Real and (b) imaginary parts of the propagation constant of the two lowest-order eigenmodes that do propagate in a medium of periodically arranged modified omega particles.

ous distinctive polarization components. The strength of optical activity depends then on the coupling of the incident field to this eigenmode.

As a complementary approach in understanding the occurrence of the polarization rotation, Fig. 3 shows (a) the real and (b) the imaginary parts of the propagation constants for the two lowest-order eigenmodes at wave numbers smaller than $\nu=1\times10^4$ cm^{-1}. For the computation of the eigenmodes, the medium was assumed to be composed of periodically arranged elementary unit cells in the principal propagation direction. Hence, the structure is truly three dimensional. The corresponding wave equation was then solved assuming Bloch-periodic boundary conditions in this principal propagation direction.[27] The approach permits the derivation of the propagation constant as a function of the frequency and the transverse wave vector for all eigenmodes.[28] In the present case we have put the transverse wave vector to zero as light propagation is considered only into the z direction.

Although the final problem of determining the polarization rotation from this dispersion relation is only possible by solving the question of how an external field couples to the eigenmodes, at least it can be seen that spectral positions of vanishing polarization rotation correlate to spectral positions where the propagation constants of the two lowest-order eigenmodes have the same real part. This occurs at $\nu=0.38\times10^4$ and at $\nu=0.65\times10^4$ cm^{-1}. Both spectral positions are in perfect agreement with spectral positions of $\overline{\Delta\Phi}=0°/\text{mm}$ as shown in Fig. 2. Only at these frequencies, the phase advance inside the medium is the same for all eigenmodes that contribute to the transport of light through the medium. Therefore, although the structure is chiral, no polarization conversion can be observed. It remains to mention that the description ceases to be valid at larger spectral frequencies because higher order eigenmodes gain importance and more than two modes contribute to the light transport.

In conclusion, we have systematically analyzed the optical activity of planar periodic arrangements of three-dimensional chiral meta-atoms. The working principle of these meta-atoms was explained in terms of elementary excitations of the structure. A large optical activity was predicted to occur if the meta-atom contains two structural elements, which may exhibit polariton plasmon resonances with orthogonal polarizations. If these structural elements are much less than the wavelength of interest, the excitations represent either electric or magnetic dipoles. These considerations permit the rational design of 3D meta-atoms forming the building blocks for a 3D chiral metamaterial. Chiral meta-atoms relying on the coupling of an electric with a magnetic dipole (a cut wire—SRR geometry) proved superior to a meta-atom with two coupled electric dipoles. The optical activity for a planar arrangement of the former structure was as large as 30.43° per layer. In this respect these meta-atoms exhibit a giant chirality. Changing the geometry of the essential structural units permits spectral tuning of the chiral properties. Based on this idea this work hosts a methodical aspect as well. It allows both derivation of rational design strategies for chiral unit cells and disclosure of the basic physics of the working principle of highly efficient chiral media.

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