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Bifacial Metasurface with Quadrupole Optical Response

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We design, fabricate, and characterize a metasurface, whose multipole optical response depends significantly on the illumination direction. The metasurface is composed of gold-nanodisc dimers embedded in glass. In spite of their nanoscale size, the dimers exhibit a dominating electric-current-quadrupole response in a wide range of wavelengths around 700 nm when illuminated from one side, and a primarily electric-dipole response when illuminated from the opposite side. This leads to two consequences. First, the reflection coefficient of the metasurface considerably differs for the two sides of illumination. Second, quadrupole excitation results in a significant local enhancement of both electric and magnetic fields around the dimers. Our experimental spectroscopic data are in good agreement with simulations obtained using a multipole expansion model.

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I. INTRODUCTION

Metamaterials are artificial electromagnetic media composed of subwavelength-sized structural units that are called metamolecules [1,2]. In contrast to ordinary molecules, metamolecules can be designed to exhibit pronounced multipole excitations beyond the electric dipole, such as magnetic dipole and electric quadrupole, at optical frequencies. This gives the materials a wider tunability of the refractive index and an additional degree of freedom in terms of wave impedance that can be changed independently of the refractive index. As an example, a metamaterial can be designed to have a negative refractive index in order to provide “perfect imaging” [3–7] and simultaneously the impedance equal to that of vacuum in order to minimize optical reflection from the material’s surface. Other extraordinary applications include optical cloaking devices [8,9], elements enhancing optical density of states and energy transfer [10,11] as well as coherence [12–14], and a variety of optical elements composed of dispersive and nonlinear materials [15–17].

Although metamaterials promise a rich spectrum of novel functionalities, their practical realization at optical frequencies still faces some technological and even fundamental problems. For example, optically magnetic metamaterials are, as a rule, spatially dispersive [18,19], which is unwanted for many proposed applications, such as a perfect lens and an optical cloak. Furthermore, often-used metal metamolecules show too-high optical absorption, and

due to unavoidable fabrication errors, also scattering loss. *Metasurfaces*, on the other hand, contain only a single layer of metamolecules, and hence, they are easier to design, fabricate, test, and adjust for specific purposes [20–22]. Metasurfaces are therefore more likely to find widespread technological applications. Recently demonstrated examples include holographic elements [23], lenses and axicons [24], antireflection coatings [25], mirrors [26], and polarization converters [27,28]. To our knowledge, however, a metasurface or a metamaterial with an electric-dipole-free optical response [29] and with a strong dependence of optical reflection on the illumination side [19] has not yet been demonstrated experimentally in the visible spectral range.

In this work, we demonstrate a type of metasurface in which optical multipole excitations substantially depend on the illumination side, resulting in side-dependent optical reflection. We also show that the excited higher-order multipoles can prevail over the electric dipoles. Furthermore, we discover that the electric-current quadrupole excitation, while producing less scattering, yields a stronger near field compared to the electric-dipole excitation obtained for the opposite illumination side. The metamolecules are pairs of gold nanodiscs arranged in a periodic two-dimensional array embedded in glass. The disc-dimer geometry (see the inset of Fig. 1) is chosen because it is a generic asymmetric configuration for controlling the two most significant lowest-order multipole moments, the electric-current quadrupole and electric dipole moments, independently of the polarization of the incident field [19,29–31]. Similar interaction effects, such as asymmetric optical scattering and reflection, have been reported in regard to asymmetric nanoscatterers [32–35]

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FIG. 1. The scanning electron micrograph shows a view of a section of the metasurface. The two layers are aligned almost perfectly. The inset shows the design of the metamolecules and the nominal dimensions of the structure.

and metasurfaces in the infrared [36–38] and microwave [39] spectral ranges.

II. GENERAL CONSIDERATION

If the structural units of a metasurface are small compared to the excitation wavelength, one can truncate the multipole expansion after the electric-current quadrupoles, which are the multipoles combining the classical electric quadrupoles and magnetic dipoles [29,40]. In this case, the normal-incidence reflection and transmission coefficients ρ_0 and τ_0 of the metasurface can, for a linearly polarized optical wave, be written as [19]

$$\rho_0 = \frac{k}{2\epsilon\Lambda^2} (i\alpha_x - k\beta_{xz}), \quad (1)$$

$$\tau_0 = \frac{k}{2\epsilon\Lambda^2} (i\alpha_x + k\beta_{xz}) + 1. \quad (2)$$

Here, we assume that at normal incidence the response of the metasurface is polarization independent. The parameters $\alpha_x = p_x/E_0$ and $\beta_{xz} = q_{xz}/E_0$ are, respectively, the electric-dipole and current-quadrupole polarizabilities connected to the electric-dipole and electric-current quadrupole moments p_x and q_{xz} excited by an x -polarized plane wave with an electric-field amplitude E_0 . The wave propagates in the positive z direction. The period of the metasurface in the xy plane is Λ , and k and ϵ are, respectively, the wave number in and the electric permittivity of the surrounding medium. The polarizabilities α_x and β_{xz} are complex quantities that describe the response of the metamolecules to incident plane waves. For individual nanodimers, the properties of these polarizabilities are described in Ref. [31]. If the metamolecules are not symmetric with respect to the xy plane in the middle of the metasurface, α_x

and β_{xz} can depend not only on the light polarization, but also on the illumination side.

The expansion in the electric-current multipoles we use is described in detail in Ref. [40]. Briefly, we expand the electric-current density distribution $\mathbf{J}(\mathbf{r})$ in a scatterer (a metamolecule in our case) in a series of elementary current excitations, as shown schematically in Fig. 2. Each excitation corresponds to a certain element of the corresponding multipole tensor. For example, the current density $\mathbf{J}_1(\mathbf{r})$ of the first-order multipole in the expansion is the same as that of a point electric dipole, i.e., $\mathbf{J}_1(\mathbf{r}) = \mathbf{C}_1\delta(\mathbf{r})$, where $\mathbf{C}_1$ is a vector. The next-order multipole is the electric-current quadrupole. The tensor elements of this multipole are obtained by splitting the dipole current into two current elements oscillating in the opposite directions. The two possible current configurations are shown in the third box of Fig. 2. In total they form nine elements with mutually orthogonal orientations. The current excitations of the octupole contain four linear current elements obtained by splitting and flipping the quadrupole elements, as shown in the fourth box. An important advantage of this decomposition over the classical multipole expansion is that ours is a complete one. In particular, it also includes the toroidal multipoles that are missing in the classical decomposition (see, e.g., Sec. 4.1 of [41]). As an example, the toroidal dipole is a simple combination of the electric-current octupole elements (see Fig. 2 of Ref. [40]). Furthermore, the excitable electric-current configurations representing the elements of our expansion are connected to the geometry of the nanoscatrerer, which helps tremendously in designing the metamaterial constituents. For example, in order to obtain significant octupole moments, one would need the scatterers to be in the form of quadrimers instead of dimers [41]. The moment q_{xz} represents the oscillation of two opposite electric-current elements, as in the upper

FIG. 2. The multipole expansion of the electric-current density distribution $\mathbf{J}(\mathbf{r})$ calculated for an arbitrary scatterer (S) in a series of electric-current multipoles. The first element of the expansion is the classical point electric dipole. It is given by the current density $\mathbf{J}_1(\mathbf{r}) = \mathbf{C}_1\delta(\mathbf{r})$, where $\mathbf{C}_1$ is a vector. The next multipole is the electric-current quadrupole, represented by $\mathbf{J}_2(\mathbf{r}) = \hat{\mathbf{C}}_2 \cdot \nabla\delta(\mathbf{r})$, where $\hat{\mathbf{C}}_2$ is a dyadic. The electric-current octupole is described by the current density $\mathbf{J}_3(\mathbf{r}) = [\hat{\mathbf{C}}_3 \cdot \nabla] \cdot \nabla\delta(\mathbf{r})$, where $\hat{\mathbf{C}}_3$ is a tensor of rank 3, etc. Each tensor element in the expansion corresponds to a certain elementary electric-current configuration. The boxes contain these configurations, but not all possible orientations of them.

quadrupole configuration of Fig. 2, which are parallel to the x axis and displaced with respect to each other in the z direction. Only this quadrupole-tensor element contributes to the reflection and transmission; e.g., the moment q_{zx} cannot radiate in the considered case.

The metasurface described with Eqs. (1) and (2) is not bifacial if the quadrupole polarizability β_{xz} is equal to 0. Indeed, optical reciprocity requires the transmission coefficient τ_0 to be the same for the two illumination directions, which according to Eq. (2) implies that α_x is also the same whenever $\beta_{xz} = 0$. In this case, however, ρ_0 given by Eq. (1) must be the same as well. The presence of both multipole excitations, on the other hand, allows each of α_x and β_{xz} , and as a result ρ_0 , to differ for different illumination sides. It is interesting to note that the sum $i\alpha_x + k\beta_{xz}$ must still be the same for the two illumination sides to ensure that τ_0 is also the same. This makes the values of $\alpha_x^{(1)}$ and $\beta_{xz}^{(1)}$ evaluated for one illumination direction depend on $\alpha_x^{(2)}$ and $\beta_{xz}^{(2)}$ obtained for the opposite illumination direction. This type of mutual dependence of the opposite-side polarizabilities will hold independently on the choice of the multipole expansion and the multipole polarizabilities, because the equality $\tau_0^{(1)} = \tau_0^{(2)}$ holds in all cases. Another interesting observation is that the reflection can be completely suppressed by tuning the multipole polarizabilities such that $i\alpha_x = k\beta_{xz}$. This suggests a way to design ultrathin antireflection coatings and perfect absorbers [42]. We point out that in the presence of spatial dispersion, the transmission and reflection coefficients of a metasurface can be written in terms of the so-called bianisotropic polarizabilities, as in Ref. [36]. Equations (1) and (2) do not contain explicitly the traditional magnetic-dipole and electric-quadrupole moments. Neither do they contain the mentioned bianisotropic polarizabilities. The two approaches, however, are equivalent and give exactly the same physical results. The electric-current quadrupole moment in our approach depends on the illumination direction similarly to the electric-dipole moment. Moreover, since the electric-current multipoles are expressible through the classical multipoles [40], the classical multipole moments evaluated by using the two approaches must and will be exactly the same. In our calculations, however, we do not divide each multipole moment into parts excited separately by the electric and magnetic components of the incident field, as done when using bianisotropic polarizabilities.

III. METASURFACE DESIGN AND EXPERIMENTS

The metasurface design is shown in Fig. 1. The meta-molecules are gold-nanodisc dimers with diameters 55 and 110 nm. The discs are separated by a gap of 70 nm and have a nominal thickness of 20 nm. The period of the array is 190 nm. The metasurface is fabricated using two-step electron-beam lithography on a fused-silica substrate.

FIG. 3. The measured and calculated reflectance and transmittance spectra. Around the wavelength of 675 nm the reflectance for light incident from the larger-disc side (green) is significantly higher than from the smaller-disc side (red). The measured data shown by dots are in excellent agreement with the calculated spectra shown by the continuous lines.

First a 100-nm-thick transparent film of photopolymer is deposited by spin coating (PC403, JCR, Japan). Then, 200-nm-thick double-layer polymethyl methacrylate is used as resist with the lower layer being more sensitive than the 50-nm-thick top layer. This results in an undercut of the resist after exposure and development. The sample is exposed with the pattern of the larger discs, and after development of the resist a 3-nm adhesion layer of chromium and 20 nm of gold is deposited by thermal evaporation. The metal in the unexposed areas is removed in a lift-off process resulting in a regular array of nanodiscs. The first layer of the metasurface is then coated with PC403 to planarize the sample and to provide the necessary spacer. The second layer of the structure is then fabricated in the same way as the first layer. The two layers are aligned with an accuracy of ca. 10 nm to each

FIG. 4. The absolute values (top) and phases (bottom) of $ip_x(\lambda)$ and $kq_{xz}(\lambda)$. The colors of the curves correspond to the illumination side (see the red and green arrows in the inset). The stars show the results of direct numerical calculations using the field distributions in the metamolecules. A dominating electric-current quadrupole scattering is observed for the smaller-disc illumination side (red) at $\lambda \in [675 \text{ nm}, 790 \text{ nm}]$. For light incident from the larger-disc side the spectrum is dominated by the electric-dipole moment (see the green curves). E_0 is set to 1 V/m.

other using markers in the first layer [43]. Finally, the sample is covered with a 100-nm-thick layer of PC403 to provide a homogeneous dielectric environment to the structure. The refractive index of PC403 is 1.55. A scanning electron micrograph of the realized metasurface is shown in Fig. 1.

Our key experimental results are shown in Fig. 3. The sample reflectances measured for the two illumination sides [see the green and red dots in (a)] have clearly different values around $\lambda = 675 \text{ nm}$, indicating a significant electric-current quadrupole excitation in the metamolecules. The color of the curves indicates the direction of incidence: incidence from the larger-disc side (green) and from the smaller-disc side (red). Our experiment yields the wavelength dependence of the reflection, but not the absolute reflectivity. Hence, the reflection spectra shown in (a) use arbitrary units. The transmission spectra are identical for light incident from the two sides due to optical reciprocity. The blue dots in (c) show this spectrum measured when the sample is illuminated from the larger-disc side. The green, red, and blue solid lines in (b) and (c) show the spectra calculated using the finite element method (COMSOL Multiphysics). These spectra are seen to be in good agreement

with the measured data. For the simulations we assumed that the chromium adhesion layer had oxidized to Cr_2O_3 and used a frequency-independent refractive index of $n_a = 2.2 + 0.5i$ for the adhesion layer [44,45]. Also, we took the values of the refractive index of gold from [46] and used a 15-nm thickness for the discs to best match the resonant features in the measured curves.

IV. ANALYSIS

Using the simulated reflection and transmission coefficients of the sample we can evaluate the coefficients ρ_0 and τ_0 of the metasurface by excluding the influence of the glass-air interfaces. Then, applying Eqs. (1) and (2), we can obtain the spectra of the polarizabilities $\alpha_x(\lambda) = p_x(\lambda)/E_0$ and $\beta_{xz}(\lambda) = q_{xz}(\lambda)/E_0$. Solid and dashed lines in Fig. 4 show the calculated amplitudes and phases of the contributions $ip_x(\lambda)$ and $kq_{xz}(\lambda)$ to the reflection and transmission coefficients; E_0 is set to 1 V/m. We note that for our dimers, the contribution of the multipole moments of higher orders than electric-current quadrupole to the scattering is negligibly small. This is verified by applying the rigorous multipole expansion of Ref. [40] to the field distributions obtained directly in the numerical calculations (such as those in Fig. 5). The results of the direct numerical calculations of the multipole moments p_x and q_{xz} are shown by stars in Fig. 4. They are in good agreement with

FIG. 5. Distributions of the electric and magnetic field components around the dimers at $\lambda = 715 \text{ nm}$. White arrows show the illumination direction. The colors reflect the normalized absolute values of the fields. The color bars show the field-enhancement factors $|E/E_0|$ in (a) and (c) and $|H/H_0|$ in (b) and (d), where E_0 and H_0 are the incident wave amplitudes.

those calculated from Eqs. (1) and (2), which proves the validity of these equations for the considered metasurface. At λ between 675 and 790 nm, the electric-current quadrupole moment is seen to prevail over the electric-dipole moment for the illumination from the smaller-disc side (see the red dashed line above the red solid line in the upper plot). At about 715 nm, the dipole moment is nearly completely suppressed and the scattering is almost purely of the current-quadrupole nature [29]. For the opposite-side illumination, the situation is reversed and the reflection is dominated by the electric dipole (see the green dashed line below the green solid line). For the smaller-disc illumination side at $\lambda_0 \approx 675$ nm, the multipole contributions $ip_x(\lambda)$ and $-kq_{xz}(\lambda)$ to the reflection coefficient have comparable amplitudes and oscillate nearly out of phase, which leads to a reduced reflectance, as was discussed previously. For the opposite illumination side, the situation changes and the reflectance is high. Hence, the multipole analysis provides a clear and unambiguous explanation of the obtained experimental results.

The distributions of the electric and magnetic field amplitudes corresponding to the nearly pure current-quadrupole scattering at $\lambda \approx 715$ nm are shown in Figs. 5(a) and 5(b), respectively. The dimers are illuminated from the smaller-disc side in this case. The near field of the dimers primarily around the upper disc exhibits quite high enhancement factors $|E/E_0|$ for the electric field and $|H/H_0|$ for the magnetic field; E_0 and H_0 are the incident wave amplitudes. The electric currents in the discs oscillate nearly out of phase and have nearly equal volume-integrated magnitudes. The complex amplitudes of these integrated current elements in the smaller and larger discs are $1 + 0.8i$ and $-1.1 - 0.7i$, respectively. They are normalized with respect to the real part of the first one. For the opposite illumination direction, the field profiles are different, as shown in Figs. 5(c) and 5(d). The electric and magnetic fields are seen to be enhanced also in this case, but not much. This explains a lower optical absorption and higher reflection by the metasurface compared to the small-disc illumination side. The normalized complex amplitudes of the volume-integrated current elements corresponding to this case are $0.2 + 0.6i$ and $-0.8 + 1.5i$ for the smaller and larger disc, respectively.

V. SUMMARY

We have designed and realized experimentally a bifacial optical metasurface that exhibits a dominating electric-current quadrupole excitation in its gold metamolecules in a broad wavelength range between 675 and 790 nm. When the metasurface is illuminated from one side with a light of 675-nm wavelength, the fields radiated by the excited current-quadrupole and electric-dipole moments in the backward direction oscillate approximately out of phase and have equal amplitudes, which reduces the reflectance. Simultaneously, both the electric and magnetic near fields

of the dimers are considerably enhanced. For the opposite illumination direction, the electric-dipole radiation dominates over the quadrupole one and the reflectance is high. Accordingly, the near-field enhancement is low. The observed good agreement between the predicted and measured reflectance and transmittance spectra demonstrates the power of our current-multipole approach to the design of optical metasurfaces and metamaterials. The unusual asymmetric reflection and field enhancement characteristic of such metasurfaces can lead to novel applications, e.g., in interferometric devices, solar cells, optical isolators, spatial filters, and nanofabricated optoelectronic devices including directional light sources and detectors.

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