Interplay Between Optical Biaxiality and Magnetism in Plasmonic Metamolecules

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ABSTRACT: The smallness of natural molecules and atoms with respect to the wavelength of light imposes severe limits on the nature of their optical response. For example, the well-known argument of Landau and Lifshitz and its recent extensions that include chiral molecules show that the electric dipole response dominates over the magneto-electric (biaxial) and even smaller magnetic dipole optical response for all natural materials. Here, we experimentally demonstrate that both these responses can be greatly enhanced in plasmonic nanoclusters. Using atomic force microscopy nanomanipulation technique, we assemble a plasmonic metamolecule that is designed for strong and simultaneous optical magnetic and magneto-electric excitation. Angle-dependent scattering spectroscopy is used to disentangle the two responses and to demonstrate that their constructive/destructive interplay causes strong directional scattering asymmetry. This asymmetry is used to extract both magneto-electric and magnetic dipole responses and to demonstrate their enhancement in comparison to ordinary atomistic materials.

KEYWORDS: Plasmonics, nanoparticle, biaxiality, optical magnetism, meta-molecular

Due to the symmetry of Maxwell’s equations, the electric and magnetic fields, $\vec{E}$ and $\vec{B}$, play equivalent roles in light waves propagating in a vacuum. However, their equivalence no longer holds after light enters a medium comprising atoms or molecules. In fact, the most important part of the medium response can be described in the dipole approximation that assumes that the electric field induces an electric dipole moment $\vec{p} = \alpha_{EE}\vec{E}$, where $\alpha_{EE}$ is a polarizability. The accuracy of the dipole approximation rests on the textbook argument by Landau and Lifshitz that the magnetic dipole moment $\vec{m} = \alpha_{MM}\vec{H}$ induced by the magnetic field is negligible because $\alpha_{MM} \approx \frac{d^3}{\lambda^3}$, where $d$ is the typical size of a molecule and $\lambda$ is the wavelength of light. Somewhat less familiar, but crucial for understanding such fundamental phenomena as optical activity of chiral media, is the magneto-electric coupling $\alpha_{ME}$ which describes the excitation of the electric dipole moment by the magnetic field, and vice versa: $\vec{p} = \alpha_{EE}\vec{E} + \alpha_{ME}\vec{H}$. The extension of the Landau–Lifshitz argument to chiral molecular media indicates that $|\alpha_{ME}| \lesssim |\alpha_{ME}| \ll |\alpha_{EE}|$ in the low-frequency regime, i.e., when the light is detuned far from any atomic transitions. Note that the diagonal and off-diagonal components of the magneto-electric coupling tensor are usually referred to as chiral and biaxial, respectively. For a fully symmetric molecule possessing all three mirror symmetry planes $\alpha_{ME} = 0$ and breaking of at least one reflection, symmetry is required for at least some components of $\alpha_{ME}$ to be nonvanishing.

Recent advances in plasmonics and metamaterials demonstrated that these restrictive scalings can be lifted for plasmonic nanoclusters. While recent experiments demonstrated the excitation of magnetic resonances by observing characteristic Fano interference features in the scattering spectrum, distinguishing between magneto-electric (ME) and magnetic moment (MM) coupling has not been feasible. This difficulty mainly arises from the complicated tensorial nature of the magneto-electric coupling in chiral media.
former, and the fact that the simplest manifestations of the two coupling mechanisms, such as Fano resonance in the scattering cross section, are essentially the same. In this Letter, we use a precise nanomanipulation technique to assemble four nearly identical spherical nanoparticles (NPs) into an isosceles trapezoid geometry. The resulting metamolecule possesses a very simple ME coupling tensor with a single dominant component $\alpha_{ME} \equiv \alpha_{ME} \mathbf{I}$. We show experimentally and theoretically that the combination of the ME and MM coupling mechanisms has an unambiguous experimental manifestation: highly asymmetric scattering that strongly depends on the polarization and incidence direction of light.\(^1\) Finally, we use angle-resolved scattering spectroscopy to disentangle these two contributions to the magnetic resonance and to demonstrate that the Landau–Lifshitz inequality $|\alpha_{MM}| \ll |\alpha_{ME}|$ is broken in the metamolecule.

The experiment schematically illustrated in Figure 1a utilizes an asymmetric nanocluster comprising identical nanospheres assembled into an isosceles trapezoid geometry and illuminated by a broadband light source either in normal or oblique incident direction. The resulting metamolecule supports three plasmonic resonances schematically shown in Figure 1b: two electric dipole resonances pointing in $x$- and $y$-directions (black arrows) and one magnetic dipole resonance in the $z$-direction (black circulating arrows). It has been previously demonstrated that the magnetic dipole resonance arising from the circulating displacement current can be tuned in wavelength by simply changing the gap size between the NPs.\(^17,26\) When the magnetic and electric dipole resonances spectrally overlap, Fano resonance features at the magnetic resonance frequency are observed in the scattering spectra for either the normal or oblique incident light as illustrated in Figure 1. However, the details of the Fano feature strongly depend on the incident light’s polarization state and incidence angle. We exploit the angle and polarization-dependent scattering spectra to disentangle the ME and MM excitation mechanisms.

The role of the ME excitation of the magnetic resonance is particularly transparent for normally incident light (Figure 1b) because it does not contain the $z$-component $H_z$ of the magnetic field. The symmetry of the metamolecule prohibits the coupling between the $x$-oriented electric dipole and the magnetic dipole moment, and no magnetic dipole excitation takes place. Thus, no spectral Fano features are observed in the scattering spectra shown in Figure 1b (case I, dashed line) for $x$-polarized light. For all other polarizations, the $y$-oriented electric dipole resonance of the two unequal dipole pairs of NPs (left and right) is excited. The near field ME coupling of the above electric dipole (with the amplitude $a_x$) to the $z$-oriented magnetic dipole (with the amplitude $a_y$) is enabled by the broken mirror symmetry of the metamolecule. The corresponding characteristic Fano feature, which is maximized for the $y$-polarized light (Figure 1b, case II, solid line), can be used for extracting $|\alpha_{ME}|$ (see Methods for details). However, to separately quantify the MM-coupling schematically illustrated in Figure 1c (i.e., to extract the $\alpha_{MM}$ coefficient) it is imperative to use high precision nanomanipulation techniques to assemble four nearly identical spherical nanoparticles (NPs) into an isosceles trapezoid. The near-field coupling rate between the two resonances given by the biaxial magneto-electric coefficient $\chi$ is proportional to the asymmetry of the gaps’ sizes and is related to the magneto-electric coefficient $\alpha_{ME}$ (see Methods for details). The far-field coupling strength of the cluster to the incident field is proportional to the external coupling coefficients $\kappa_{EF}^{(p)}$, which determine the radiative lifetimes of the resonances according to $\gamma_{p(\mu)} = \gamma_{p(\mu),r} + \gamma_{p(\mu),d}$. Within the CMT framework, the electric and magnetic dipole moments $p$ and $\mu$ are given by $P = \kappa_{EF}^{(p)} p$ and $\mu = \kappa_{EF}^{(\mu)} \mu$, respectively. If the full set of coupling coefficients $\gamma_{p(\mu)}$ and

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Figure 1. Effect of electric and magnetic dipole resonance coupling on angle-dependent scattering spectra. (a) Illustration of angle-resolved scattering measurements. The incident light may come from either normal or oblique incident angles and may approach the metamolecule from different sides. (b) Illustration of mode coupling and scattering spectra for normal incident light. Top panel: illustration of the absence or presence of ME coupling between the electric dipole (black arrow) and magnetic dipole (black circle) in the asymmetric four-particle metamolecule with the external E-field (blue arrow) parallel (case I) or perpendicular (case II) to the symmetric axis of the isosceles trapezoid. Bottom panel: scattering spectra with normal incident light corresponding to case I and II. (c) Illustration of mode coupling and scattering spectra for oblique incident light. Top panel: illustration of constructive or destructive interference between ME and MM coupling terms with different magnetic field phases when light approaches the metamolecule from the side with a larger gap size (case III) and a smaller gap size (case IV). Bottom panel: scattering spectra with oblique incident light corresponding to cases III and IV.
κM can be extracted from the scattering spectra, then the set of low-frequency polarizabilities (αEE, αEM, αMM) are expressed (see Methods for details) as follows:

\[ \alpha_{EE} = \frac{k_E^2}{\omega^2}, \quad \alpha_{ME} = -\frac{i k_M k_S}{\omega^2}, \quad \alpha_{MM} = \frac{k_M^2}{\omega^2} \] (3)

Their relative magnitudes can then be compared with the Landau–Lifshitz scalings, which are valid for atomic systems in the optical (low frequency) regime.

When the upward-scattered light is collected by a small numerical aperture (NA) lens as illustrated in Figure 1a, the amplitude of the signal collected in the direction normal to the in-plane dipole moment: \( S(\theta, \omega) \propto \sqrt{k_M^2} \), Therefore, the magnetic dipole radiation is not directly measured. Instead, by depolarizing the electric dipole moment \( a_p \) according to eq 1, the magnetic dipole \( a_M \) indirectly affects the scattering spectra and manifests itself as a Fano feature emerging in the \( S(\theta, \omega) \) spectrum at \( \omega = \omega_p \) as illustrated in Figure 1b,c. The reason for the sharpness of the Fano feature is that the magnetic dipole resonance plays the role of the “dark” mode because \( f_{p,R} \ll f_{p,B} \). This hierarchy of the decay rates is caused by the weaker coupling of the magnetic dipole moment to the external field, i.e., \( k_M^2 \ll k_E^2 \). Importantly, both pathways for the excitation of the magnetic dipole \( a_M \), the direct excitation by the magnetic field \( H_p \) and the indirect near-field bianisotropic excitation by the electric dipole, \( a_p \), are fully accounted for by the CMT model. From eqs 1 and 2 we obtain a simplified expression for the Fano spectrum for the upward scattered light:

\[ S(\theta, \omega) = \frac{[\gamma_p^2 + (\omega - \omega_p)^2] + 2\gamma_p \sin \theta(\kappa_M/k_E) + \gamma^2 \kappa_M^2}{\gamma_p^2 + (\omega - \omega_p)^2} \] (4)

Three key observations can be made from analyzing eq 4. First, in the absence of the bianisotropic coupling (\( \gamma = 0 \)) no Fano feature emerges for any illumination angle \( \theta \) despite the direct excitation of the magnetic moment according to eq 2. This property of fully symmetric nanocavities was demonstrated earlier, where a combination of a cross polarizer (to suppress the electric dipole’s contribution to scattering) and large numerical aperture (to collect the scattered signal at large angles from the normal) was used to observe the magnetic dipole’s excitation. Second, for normally incident light (\( \theta = 0 \)) the \( k_M \) term does not contribute to the scattering spectrum. Therefore, it is possible to experimentally extract the value of \( \chi \) by measuring the magnitude of the Fano feature for the normally incident \( y \)-polarized light. Note that the \( y \)-direction, defined by cluster’s orientation as being perpendicular to the symmetry axis of the isosceles trapezoid (Figure 1b), can be experimentally established by maximizing the Fano spectral feature in polarization-resolved dark field scattering experiments. Finally, the normalized scattering cross section \( S(\theta, \omega) \neq S(-\theta, \omega) \) is, in general, asymmetric with respect to the incidence angle.

Qualitatively, the scattering asymmetry defined as \( \Delta S(\theta) = S(\theta, \omega_p) - S(-\theta, \omega_p) \) is caused by the constructive/destructive interference between the ME and MM pathways of the magnetic dipole moment’s excitation. A simplified expression for \( \Delta S \) can be obtained in the proximity of the magnetic dipole resonance (\( \omega = \omega_p \)):

\[ \Delta S(\theta) = \frac{4 \sin \theta \gamma_p \kappa_M}{k_E^2 E_0^2} = \frac{4 \sin \theta \gamma_p \kappa_M}{\sqrt{k_M^2 + (\omega - \omega_p)^2}} \] (5)

It is evident from eq 5 that both pathways of the magnetic dipole excitation (\( \kappa_M \neq 0 \) and \( \chi \neq 0 \)) are necessary for nonvanishing scattering asymmetry. Therefore, the quantitative measurement of \( k_M/k_E \propto \alpha_{MM}/\alpha_{EE} \) as seen from eq 3, which is not accessible from the normal-incidence experiments, can be derived from the experimentally measured scattering asymmetry.

Super spherical Au NPs were used as the building blocks of the metamolecule. These nanospheres are produced in two steps: the synthesis of octahedral Au NPs is followed by galvanic etching (see Methods for details). The uniformity of the NPs can be observed from the TEM images shown in Figure 2a. The average diameter of the gold nanospheres is estimated to be \( 88 \pm 5.3 \) nm. Following the synthesis procedure, we further improve the uniformity of NPs by selecting a few NPs with nearly identical scattering spectra. The diluted NPs are spread on a patterned quartz substrate with suitable density (a few NPs over a \( 10 \mu m \) by \( 10 \mu m \) area, Figure 2b). After locating the NPs with an atomic force microscopy (AFM) image, scattering spectra from eight different NPs are taken and displayed in Figure 2c. All spectra feature an electric dipole resonance near \( 532 \pm 2 \) nm. The similarity between these scattering spectra supports the claim that these NPs are highly uniform in size and shape as observed in the TEM image. In the following AFM assembly process, we selected four NPs with nearly overlapping scattering spectra, as shown in Figure 2c inset.

We then use the AFM nanomanipulation method (illustrated in Figure 2d) to assemble the designed metamolecule with an isosceles trapezoid geometry (see methods for details). The 3D view of the assembled structure is shown in Figure 2e. The accuracy in controlling the gap size (a few nanometers) between the NPs is limited by the finite resolution in the lateral dimensions of the AFM image due to the tip convolution effect. While the highest achievable resolution may be \( \sim 1 \) nm using super sharp tips, we have chosen tips with relatively large tip radius to reduce the possibility of damage during nanomanipulation. Thus, the actual dimensions of the assembled structure have some deviations from the ideal design. In practice, we repeatedly adjusted the gap size between the NPs until the desired spectroscopy features emerged. The measured scattering spectra exhibit symmetry and properties closely resembling the calculations based on the designed isosceles trapezoid geometry, indicating sufficient positioning accuracy in the assembled metamolecule.

The experimental setup for normally incident light is illustrated in Figure 3a. Linearly polarized white light was focused through a microscope objective with NA = 0.28 onto the sample. A microscope objective with NA = 0.7 was used to collect scattered light. A small opaque screen was placed in the center of the optical path to block the directly transmitted light.
so that only the scattered light is sent through the spectrometer to generate the scattering spectra. Polarization-resolved dark field scattering measurements were performed to determine the spatial orientation of the metamolecule and to extract the bianisotropic coupling coefficient $\chi$. When incident E-field is along the $y$-direction (which we refer to as the Fano axis $\text{fi}$), the Fano resonance arising from the ME coupling is the most pronounced. A series of spectra taken at a sequence of polarizer angles in the $10^\circ < \phi < 170^\circ$ range are shown in Figure 3b, with $\phi = 0^\circ$ direction shown as the white dotted line in Figure 3b inset. A strong scattering peak corresponding to the wavelength of the electric dipole resonance at $\lambda_e \equiv 2\pi/\omega_e \approx 690$ nm is observed in all spectra, while a Fano resonance near $\lambda_m \equiv 2\pi/\omega_m \approx 815$ nm is most strongly manifested for $\phi = 90^\circ$ direction coincident with the Fano axis.

To understand the polarization dependence of the scattering spectra, COMSOL simulations were carried out for $\text{fi}$ (Figure 3c,d) and $\gamma$- (Figure 3e,f) polarized incident light assuming that the metamolecule comprised equal nanospheres with diameters $D = 90$ nm, and gap size $g_{TB} = 0.5$ nm gaps, $g_L = 0.65$ nm, and $g_R = 0.35$ nm. The subscript $T$, $B$, $L$, and $R$ correspond to top, bottom, left, and right, respectively, as labeled in Figure 3c. We note the gap sizes used in the simulation should be considered as an approximated, effective gap size rather than precise parameters reproducing the experimentally assembled metamolecules. Some details were not taken into account in the simulation. For example, the presence of ligand molecules at NP surfaces changes the effective refractive index and prevents the NPs from physically touching each other. Furthermore, the limited lateral resolution of the AFM image does not allow us to extract gap size precisely. We estimate the actual gap sizes are below 1 nm. Such trapezoidal arrangement of the nanospheres corresponds to the symmetry axis along the $\phi = 0^\circ$ direction. The calculated charge (Figure 3c,e) and magnetic energy (Figure 3d,f) distributions clearly demonstrate that the excitation of the magnetic moment occurs only for the $\phi = 90^\circ$ polarization direction. In this case, the interference between the $y$-polarized electric dipole shown in Figure 3e, and the magnetic moment shown in Figure 3f due to bianisotropic coupling (finite $\chi$) manifests itself as a Fano feature in the scattering spectrum. Because no bianisotropic coupling exists between the $x$-polarized electric dipole shown in Figure 3c and the magnetic moment, no Fano features are expected in the spectrum. These experimental and numerical results confirm the analytic prediction of eq 4 that a finite bianisotropy coefficient $\chi$ is necessary for observing the Fano resonance.

Next, we perform scattering experiments using obliquely incident ($\theta \approx 60^\circ$) $s$-polarized illumination as schematically shown in Figure 4a. The spectra corresponding to the light incident from four different directions (defined as the top, bottom, left, and right as illustrated in Figure 4b) were collected and plotted in Figure 4c–f. For all four incident directions, a finite $H_y$ component excites the magnetic dipole resonance. In the experiment, the incidence direction was fixed, and the sample was rotated around the $z$-axis in such a way as to align the electric field with the symmetry axis (see Figure 4c: top and bottom orientations) or with the Fano axis (see Figure 4d: left and right orientations). As expected from eq 4, no Fano feature or any asymmetry between the top and bottom sample orientations were experimentally observed. This experimental result is confirmed by a COMSOL simulation shown in Figure 4e, where the scattering cross sections are calculated by integrating the outgoing Poynting flux over a small solid angle ($\Delta \Omega = 0.6\pi$) around the $\theta = 0$ direction. In contrast, the measured scattering spectra exhibit strong asymmetry at the magnetic resonance wavelength $\lambda_m \approx 860$ nm between the left and right sample orientations as shown in Figure 4d. This asymmetry is due to the combination of the direct magnetic excitation by the $H_y$ field component and by the bianisotropic coupling between electric and magnetic plasmonic resonances. The numerically calculated spectra shown in Figure 4f qualitatively reproduce the experimental data.

We now combine the semianalytic model given by eqs 1–5 with the experimental scattering data for normal and oblique incidence angles to extract the key parameters of the metamolecule that describe the electric and magnetic resonances and their coupling to far-field light and to each other. First, $\omega_{(p)(m)}$, $\gamma_{(p)(m)}$, and $\chi$ are extracted as fitting parameters by fitting eq 4 (with $\theta = 0$) to the experimentally measured spectrum with $y$-polarized ($\phi = 90^\circ$) light (see the Supporting
Information section for more details). These parameters are used in combination with asymmetric scattering spectra in Figure 4d to extract the $\frac{\kappa_M}{\kappa_E} = 0.134$ ratio using eq 5. The details of this fitting and analysis can be found in the Supporting Information section. Using eq 3, the following scaling between electric, magnetic, and bianisotropic responses is obtained:

\[
\frac{\alpha_{EM}}{\alpha_{EE}} = \frac{\chi}{\alpha_m} \left( \frac{\kappa_M}{\kappa_E} \right) = 0.006,
\]

\[
\frac{\alpha_{MM}}{\alpha_{EE}} = \frac{\alpha_{m}^2}{\alpha_m} \left( \frac{\kappa_M}{\kappa_E} \right)^2 = 0.02
\]

Equation 6 demonstrates that the magnetic and bianisotropic optical excitations of a plasmonic metamolecule are no longer negligible and can be considerably larger than in natural atoms and molecules. For example, the ratio of the magnetic to electric polarizabilities is more than 2 orders of magnitude larger than the $\alpha_{MM}^{(mol)}/\alpha_{EE}^{(mol)} \approx (1/137)^2$ scaling based on natural molecules. Furthermore, the MM contribution is actually larger than the ME contribution, clearly breaking the Landau–Lifshitz permeability argument. Remarkably, our experimental extraction of the polarizabilities’ ratio based on collecting vertically scattered light was enabled by the presence of a very small bianisotropic response $\alpha_{EM} \ll \alpha_{MM}$ of the metamolecules. Of course, the magnitude of $\alpha_{EM}$ can be controlled (and further increased) by changing the asymmetry of the metamolecules. For example, the asymmetry can be increased by further reducing the smallest gap size $g_R$. This was not done in our system because the ligand molecules on the surface of the NPs limit the smallest obtainable gap size.

In summary, we experimentally extracted the optical magnetic polarizability of an asymmetric plasmonic metamolecule and disentangled its bianisotropic (ME) response from the magnetic (MM) response. This was accomplished by designing and assembling a metamolecule with a very simple single-component bianisotropic optical polarizability tensor. The excellent agreement between experiments, theory, and simulation has been enabled by the combination of several recent experimental advancements: carefully synthesized and selected NPs, a precise AFM nanomanipulation method, and angle-dependent optical scattering measurements. Remarkably, the presence of even a small ME response enabled us to quantify these two dominant nondipole optical responses using upward-scattered light that was collected by the optics placed directly above the metamolecules. The knowledge and capability of controlling different contributions to the magnetic resonance as presented in our study is expected to stimulate...
exciting future development of metamolecules and metamaterials with exotic optical properties, thus enabling a new generation of photonic materials.

**Methods.** Connection of $\alpha_{EMM,EMM}$ and $\kappa_{EM}$. At low frequency limit, from solving eqs 1 and 2 with the assumption that $\omega \rightarrow 0$,

$$p \equiv \kappa_{EM} a_p = \alpha_{EM} E_z + \alpha_{EM} H_z$$  \hspace{1cm} (7)$$

$$\mu \equiv \kappa_{M} a_\mu = \alpha_{M} E_z + \alpha_{M} H_z$$  \hspace{1cm} (8)$$

By solving eqs 7 and 8, we obtain the expressions for the electric, magnetic, and magneto-electric (bianisotropic) polarizabilities of the metamolecule given by eq 3. Note that the low-frequency regime is directly related to the optical response of atoms/molecules under the assumption that the energy of a single photon is smaller than the energy difference between excited and ground states of a molecule.

**Super Spherical NP Synthesis.** We briefly describe the synthetic process that produces super spherical NPs used as the building block of the metamolecule.\(^{29-31}\) The ultrasmooth Au nanospheres are synthesized in two steps: the synthesis of octahedral Au NPs and the subsequent galvanic etching. The etching step removes the vertices and edges of the octahedral NPs and leads to super spherical NPs. To synthesize octahedral NPs, a reaction solution of polyDADMAC (0.4 mL), H\(_3\)PO\(_4\) (0.8 mL, 1 M), and HAuCl\(_4\) (0.02 mL, 0.5 M) in anhydrous ethylene glycol (20 mL) was prepared in a 50 mL glass vial.

The mixture was stirred at room temperature for 15 min, and then the solution in the glass vial was directly placed in a hot oil bath, which was preheated to 195°C. After 30 min, the resultant solution was cooled down to room temperature. To chemically etch the octahedral gold NPs, 5 $\mu$L of a 0.5 M HAuCl\(_4\) solution was injected into the unwashed octahedral gold suspension at room temperature. The reaction mixture was kept for 20 h and then centrifuged three times with added ethanol to obtain a precipitate. All chemicals were purchased from Aldrich and used without further purification.

**AFM Nanomanipulation.** We used customer-developed automatic manipulation software integrated with the Park Systems (XE-120). The manipulation follows several steps. We first define a target position for a selected NP. The AFM probe tip moves through the center of the NP toward the target position, while z feedback of the AFM is disengaged to push the NP upon contact. The AFM tip then scanned the vicinity of the tip trajectory during the pushing step to relocate the NP, while z feedback is engaged. This push-and-scan process is repeated automatically until the NP reaches the target position. Manual manipulation toward the end of the assembly process is often necessary to finely adjust the NPs’ positions into a precise geometry with the designed gap sizes to observe the Fano resonance. Typical AFM parameters used are integrative gain of 2, proportional gain 20, servo gain 2.0, and scanning speed of 0.35–0.5 $\mu$m s\(^{-1}\) for imaging and 2–20 $\mu$m s\(^{-1}\) for manipulation.

**ASSOCIATED CONTENT**

1. Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.6b01380.

Section covering details of the fitting procedure (PDF)

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**Author Contributions**

L.S. carried out optical measurement and data analysis; J.S. and I.M. assisted in experiment. T.M. performed theoretical and numerical calculations. Super spherical gold NPs were synthesized by D.-K.K., G.L., and G.-R.Y. S.-C.Y. assembled NP metamolecule structure, and L.S. provided assistance. G.S. and X.L. guided theoretical and experimental investigations. L.S., T.M., G.S., and X.L. wrote the manuscript.

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**Notes**

The authors declare no competing financial interest.

**ABBREVIATIONS**

ME, magneto-electric; MM, magnetic moment; NPs, nanoparticles; NA, numerical aperture

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