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Hybridization of electromagnetic multipoles in the presence of another nanoscatterer

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Abstract
Coupling between multipolar modes of different orders has not been investigated in depth, despite its fundamental and practical relevance in the context of optical metamaterials and metasurfaces. Here, we use an electromagnetic multipole expansion of both the scattered fields and the oscillating electric currents to reveal the multipolar excitations in a nanoparticle positioned close to another nanoparticle. The considered single-particle multipoles radically differ from multipoles excited in a pair of nanoparticles. Using the expansion, we reveal the multipole character of the electric currents and the contributions of the multipole moments to the scattering cross section of each particle, including the effect of their interaction. We find that light scattered by the particles plays the role of an inhomogeneous incident field for each of the particles, leading to hybridization of the originally independent orthogonal multipole resonances. For an incident plane wave polarized along the nanoparticle pair, the hybridization of the dipole and quadrupole resonances gives rise to a significant narrowband resonance in the spectrum of the dipole scattering, which can be of interest for various applications, e.g. in surface-enhanced fluorescence and Raman spectroscopy. In general, this work shows that the multipole-multipole interaction between nanoparticles must be treated by taking into account also such hybridized multipole resonances.

1. Introduction

Multipole expansion is a useful tool for characterization of electromagnetic excitations in localized systems of particles interacting with light, because the field scattered by such a system can be expanded into orthogonal spherical harmonics, each corresponding to a certain electromagnetic multipole [1, 2]. The system can then be considered as a set of point multipoles located at a point that is usually chosen to coincide with the center of mass of the system. If the system is small, only few lowest-order multipoles have significant contributions to the scattered field. In such a case, the corresponding expansion coefficients are sufficient to describe the field in detail. The classical multipole expansion, however, does not always provide a clear picture of the actual electric charge oscillations in the system. Some of them, e.g. those of toroidal multipoles, can be hidden from the classical expansion, because it expands the radiated field rather than the radiating electric currents. These oscillations, however, can be revealed by using another expansion, called the electric-current multipole expansion. It expands the electric current density in the scatterer and is written in Cartesian rather than spherical coordinates [3]. The two expansions (the classical and the electric-current multipole expansions) are interconnected. For several lowest-order multipoles, the moments of these expansions depend on each other in accordance with simple analytical relations [3]. In contrast to the classical multipoles, the electric-current multipoles are not divided into electric and magnetic multipoles, because they form a complete orthogonal basis for the electric current configurations rather than the patterns of the radiated electromagnetic fields. In addition, they do not produce orthogonal scattered fields. However, they form a complete set of multipoles, including the toroidal ones, and provide a clear intuitive picture of the charge oscillations in the scatterer. It has also been shown that the expansion can be applied to nanoparticles interacting with other nanoparticles or with spatially extended objects [3]. We emphasize,
however, that interacting nanoparticles (e.g. forming dimers, trimers, etc.) are usually characterized in terms of their dipole–dipole interaction [4–8]. As a system, they can show a variety of different multipole excitations, but individually, each nanoparticle is still considered as a point dipole. In contrast, higher-order multipole resonances of individual interacting particles have not been considered much, although they can be non-negligible and significantly affect the scattering properties of the system.

In this work, we focus on the multipole excitations in individual nanoparticles when they interact with other nanoparticles. We apply both the classical and electric-current multipole expansions to characterize the excitations in such systems. Considering an example of a plasmonic nanoparticle interacting with another identical nanoparticle, we find that the multipole excitations in the former are hybridized in the vicinity of the latter. We observe a narrowband peak in the broadband dipole resonance spectrum of the nanoparticle due to a hybridization of the dipole with an electric-current quadrupole excitation. The effect should be distinguished from the widely known excitation of bright in-phase and dark out-of-phase dipole oscillations in two interacting nanoparticles [9–11]. The dark oscillation in such a dimer is characterized by higher-order multipoles of the dimer. Here, we consider multipole excitations of each of the interacting particles and find that the spectrum of the dipole moment can exhibit a feature characteristic to a higher-order multipole, improving both the brightness and the spectral narrowness of the scattered field. We introduce a figure of merit (FOM) reflecting this improvement and study its dependence on the separation distance of the particles. We find that the FOM reaches a maximum at a certain, relatively small distance between the particles. We also show that the excitation of the hybridized multipole resonance strongly depends on the polarization of the incoming light. Our work can be extended to study particles with different geometries [12, 13], material composition [7], and numbers of surrounding particles, e.g. in nanoparticle arrays [14–16]. The presented results can be of interest for researchers working in the fields of nano-optics [17–19], optical spectroscopy [20–23], as well as metamaterials and metasurfaces used to shape optical fields in both the near- and far-fields [24–30]. As an example, in surface-enhanced fluorescence and Raman spectroscopy, the highest enhancement factors are achieved in nanogaps between two metal nanoparticles interacting via their near-fields [19, 23, 29, 30]. Narrowband and bright hybrid resonances could be used to improve spectroscopic sensing and detection of molecular species based on their surface-enhanced optical transitions and refractive-index-induced spectral shifts. Furthermore, when designing optical metamaterials and metasurfaces, special attention is paid to the excitation of magnetic dipoles and other higher-order multipoles in the meta-atoms. For example, the wave impendence becomes an additional degree of freedom for a metamaterial, if magnetic dipoles or electric quadrupoles are excited in the meta-atoms in addition to electric dipoles. This allows making a material reflection-free without any antireflection coating [31]. Similarly, Huygens metasurfaces can be created by carefully balancing the excitations of the electric and magnetic dipoles in the same spectral region [32]. Hybrid multipole resonances of both electric and magnetic nature could play an important role in the creation of metamaterials and metasurfaces of these types.

2. Multipole excitations in a nanoparticle

The electromagnetic field scattered by a nanoparticle can be expanded into normalized vector spherical harmonics, $X_{lm}$, and spherical Hankel functions of the first kind, $h_{l}^{(1)}$, using the following equations:

\[
E_s(r, \theta, \phi) = E_0 \sum_{l=1}^{\infty} \sum_{m=-l}^{l} \delta^l \left[ \frac{\pi}{2} \left( 2l + 1 \right) \right]^{1/2} \left\{ \frac{1}{k} a_E(l, m) \nabla \times \left[ h_{l}^{(1)}(kr) X_{lm}(\theta, \phi) \right] + a_M(l, m) h_{l}^{(1)}(kr) X_{lm}(\theta, \phi) \right\},
\]

\[
H_s(r, \theta, \phi) = \frac{E_0}{\eta} \sum_{l=1}^{\infty} \sum_{m=-l}^{l} \delta^{l-1} \left[ \frac{\pi}{2} \left( 2l + 1 \right) \right]^{1/2} \left\{ \frac{1}{k} a_M(l, m) \nabla \times \left[ h_{l}^{(1)}(kr) X_{lm}(\theta, \phi) \right] + a_E(l, m) h_{l}^{(1)}(kr) X_{lm}(\theta, \phi) \right\},
\]

where wavenumber $k$ and impedance $\eta$ are those in the surrounding dielectric. The electric and magnetic multipole expansion coefficients, $a_E(l, m)$ and $a_M(l, m)$, can be calculated from the distribution of the scattering electric current density in the particle defined as

\[
I_s(r) = -i \omega \varepsilon_0 \left[ \varepsilon_r(r) - \varepsilon_{rd} \right] E(r).
\]
Here, $\epsilon_{rd}$ is the relative electric permittivity of the surrounding dielectric, and $\epsilon_r(\mathbf{r})$ is the relative electric permittivity as a function of coordinate $\mathbf{r}$. Outside the scatterer, we have $\epsilon_r(\mathbf{r}) = \epsilon_{rd}$. The multipole coefficients for a particle consisting of non-magnetic, isotropic, and linear materials can be calculated as

\[
a_e(l, m) = \frac{(-1)^{l+1} k^2 \eta O_{lm}}{E_0 [\pi (2l+1)]^{1/2}} \int \exp(-im\phi) \left\{ \left[ \Psi_l(kr) + \Psi'_l(kr) \right] P^m_l(\cos\theta) \hat{\mathbf{r}} \cdot \hat{\mathbf{J}}(\mathbf{r}) + \frac{\Psi'_l(kr)}{kr} \left[ \tau_{lm}(\theta) \hat{\mathbf{r}} \cdot \hat{\mathbf{J}}(\mathbf{r}) - im_{lm}(\theta) \hat{\mathbf{r}} \cdot \hat{\phi}(\mathbf{r}) \right] \right\} d^3r, \tag{4}
\]

\[
a_m(l, m) = \frac{(-1)^{l+1} k^2 \eta O_{lm}}{E_0 [\pi (2l+1)]^{1/2}} \int \exp(-im\phi) j_l(kr) \left[ i m_{lm}(\theta) \hat{\mathbf{r}} \cdot \hat{\mathbf{J}}(\mathbf{r}) + \tau_{lm}(\theta) \hat{\mathbf{r}} \cdot \hat{\phi}(\mathbf{r}) \right] d^3r, \tag{5}
\]

where $j_l(kr)$ are the spherical Bessel functions and $\Psi_l(kr) = krj_l(kr)$ are the Riccati–Bessel functions; $\Psi'_l(kr)$ and $\Psi''_l(kr)$ are the first and second derivatives of $\Psi_l(kr)$ with respect to $kr$. Other functions and parameters in equations (4) and (5) are

\[
O_{lm} = \frac{1}{l(l+1)^{1/2}} \left[ \frac{2l+1}{4\pi} \frac{(l-m)!}{(l+m)!} \right]^{1/2}, \tag{6}
\]

\[
\tau_{lm}(\theta) = \frac{d}{d\theta} P^m_l(\cos\theta), \tag{7}
\]

\[
im_{lm}(\theta) = \frac{m}{\sin\theta} P^m_l(\cos\theta), \tag{8}
\]

where $P^m_l(\cos\theta)$ are the associated Legendre polynomials that can be expressed in the form

\[
P^m_l(\cos\theta) = (-1)^m (\sin\theta)^m \frac{d^m}{d(\cos\theta)^m} P_l(\cos\theta), \tag{9}
\]

with $P_l(\cos\theta)$ given by

\[
P_l(\cos\theta) = \frac{1}{2^l l!} \frac{d^l}{d(\cos\theta)^l} (\cos^2\theta - 1)^l. \tag{10}
\]

The multipole coefficients in equations (4) and (5) can be obtained numerically by calculating the electromagnetic field inside the scatterer, e.g. with the help of a commercial software COMSOL Multiphysics. Using these coefficients, one can calculate the scattering cross section of the particle:

\[
C_s = \frac{\pi}{k^2} \sum_{l=1}^{\infty} \sum_{m=-l}^{l} (2l+1) \left[ |a_e(l, m)|^2 + |a_m(l, m)|^2 \right]. \tag{11}
\]

To calculate the multipole moments of the electric-current multipole expansion, we use a set of equations given below. The vector components of the dipole moment are calculated from (see [3] for comparison)

\[
\mathbf{p} = \begin{bmatrix}
0 & 1 & 0 \\
-1 & 0 & 1 \\
0 & 1 & 0
\end{bmatrix} \frac{1}{2C_1} \begin{bmatrix}
a_e(1, -1) \\
a_e(1, 0) \\
a_e(1, 1)
\end{bmatrix}. \tag{12}
\]

The elements of the quadrupole tensor

\[
\hat{\mathbf{Q}} = \begin{bmatrix}
Q_{xx} & Q_{xy} & Q_{xz} \\
Q_{yx} & Q_{yy} & Q_{yz} \\
Q_{zx} & Q_{zy} & Q_{zz}
\end{bmatrix} \tag{13}
\]

can be expressed as

\[
Q_{xx/yy} = \frac{1}{12C_2} \left[ \pm a_e(2, 2) \pm a_e(2, -2) \right] = \frac{1}{2\sqrt{6}C_2} a_e(2, 0), \tag{14}
\]

\[
Q_{xy/yz} = \frac{1}{12C_2} \left[ a_e(2, -2) - a_e(2, 2) \right] = \frac{1}{10\sqrt{2}C_2} a_m(1, 0), \tag{15}
\]

\[
Q_{xz/yz} = \frac{1}{12C_2} \left[ a_e(2, 1) + a_e(2, -1) \right] = \frac{1}{20C_2} \left[ a_m(1, -1) - a_m(1, 1) \right], \tag{16}
\]

\[3\]
where the coefficients $C_1$ and $C_2$ are defined as $C_1 = -i k^4/(6\pi \epsilon E_0)$ and $C_2 = -k^4/(60\pi \epsilon E_0)$; $\epsilon$ is the electric permittivity of the surrounding medium. The current multipoles, unlike classical multipoles, are not divided into electric and magnetic multipoles. The electric current configurations corresponding to some lowest-order representatives of these multipoles are illustrated in figure 1. The octupole and higher-order current excitations are not considered here, since as a rule, their contribution to the scattering is much smaller that that of the dipole and quadrupole in nanoparticles of subwavelength size.

It can be seen that nine elements of the current quadrupole tensor are expressed in terms of eight multipole coefficients. This is in agreement with the fact that a spherically symmetric multipole with $Q_{xx} = Q_{yy} = Q_{zz}$ does not radiate [33], and therefore, one of the diagonal elements can be chosen arbitrarily. In our calculations, light incident on a scatterer is considered to propagate in the $z$-direction and we set $Q_{zz}$ to zero, because in such cases, the charge excitation corresponding to this multipole moment is not expected to be significant.

Next, we calculate the scattering cross sections resulting from each current multipole using equations (11)–(17). Since each multipole coefficient is related to a certain set of the multipole moments, we use the relations provided in appendix to calculate the contributions of all the relevant multipole moments to the scattering cross section. These contributions are

$$C_s(p_{x/y/z}) = \frac{6\pi}{k^2} |C_1 p_{x/y/z}|^2,$$

$$C_s(Q_{xx/yy}) = \frac{120\pi}{k^2} |C_2 Q_{xx/yy}|^2,$$

$$C_s(Q_{xy/yz/zy/zx}) = \frac{240\pi}{k^2} |C_2 Q_{xy/yz/zy/zx}|^2.$$

The total scattering cross sections due to the electric current dipoles, $C_s(p)$, and quadrupoles, $C_s(Q)$, cannot be calculated by summing the contributions of all the individual elements of the dipole and quadrupole moments, respectively, because the fields produced by these multipoles are not orthogonal. Note, however, that the total scattering cross section due to the current quadrupoles can be calculated as a sum of the scattering cross sections by the classical electric quadrupole and magnetic dipole excitations that produce orthogonal fields.

### 3. Hybridization of multipoles by interparticle scattering

Multipoles excitations in an optical nanoscatterer can change due to the presence of another scatterer, because the scattered fields modify the total field ‘seen’ by each of the scatterers. To reveal these modifications, we numerically calculate the multipole expansion coefficients for a silver nanosphere scattering an incident plane wave in the vicinity of another identical nanosphere, displaced along the $x$-axis (see figure 2). The incident wave is assumed to propagate in the positive $z$-direction. The nanoparticle radius is taken to be 40 nm, which is large enough for quadrupole excitations to appear and small enough for octupole and other higher-order multipole excitations to still be negligible. We assume that the surrounding medium has a refractive index of 1.5 (corresponding to that of, e.g. glass) in order to make the system realizable in practice.
Figure 2. Interaction geometry of silver nanospheres with an incident plane wave. The spheres are embedded in a dielectric medium with \( n = 1.5 \), separated by distance \( g \), and have radius \( R \). The plane wave propagates in the \( z \)-direction and is either TE- or TM-polarized.

Figure 3. The spectra of the classical and current multipolar contributions to the scattering cross section of a silver nanosphere illuminated by an \( x \)-polarized plane wave propagating along axis \( z \): (a) the total scattering cross section (red line) and the contributions of the classical electric dipole (ED), electric quadrupole (EQ), and magnetic dipole (MD) represented by the blue, black, and magenta lines, respectively. The values of the scattering cross section due to the magnetic dipole excitations are multiplied by a factor of 100. The current dipole and quadrupole contributions are marked with the blue and black circles, respectively. In (b), we present the non-negligible contributions of the components of the classical electric quadrupole moment (black line) and magnetic dipole moment (magenta line). Case (c) introduces the contributions to the scattering cross section of the non-negligible current quadrupole moments, \( Q_{xz} \) and \( Q_{zx} \).

Since the expected modification of the excited multipole moments must depend on the polarization of the incident light, we consider both the transverse magnetic (TM) polarization and transverse electric (TE) polarization defined with respect to the line connecting the particles (with the electric field being parallel to the \( x \)- and \( y \)-axis, respectively).

Let us first consider a single isolated nanosphere. The spectra of the scattering cross sections due to the excited dipole and quadrupole moments for the nanosphere are presented in figure 3. The optical response is seen to be dominated by the classical electric dipole contribution (see the blue line in figure 3(a)) that is the same as the contribution of the dipole moments of the electric-current multipole expansion (see the blue
Figure 4. The classical and current multipole contributions to the scattering cross section calculated for a single silver nanoparticle in the presence of another nanoparticle: (a) and (b) show the total (red line), electric dipole (blue line), magnetic dipole (magenta line), and electric quadrupole (black line) scattering cross sections for the interparticle gap size of 50 and 10 nm, respectively. The current dipole and quadrupole contributions are shown by the blue and black circles, respectively. In (b), three characteristic points of the spectra are marked by green circles. The current density distributions in the particles corresponding to these points are plotted in figure 6.

3.1. Multipole excitations by TM-polarized light

Let us now consider the optical response of a single nanoparticle in the presence of another nanoparticle for a TM-polarized incident light. When the interparticle gap, \( g \), is infinitely large, the optical response of the particle is the same as that of an isolated particle. The spectra of the scattering cross sections are therefore the same as in figure 3. By gradually decreasing \( g \), we observe that the presence of the second particle starts to be noticeable when the gap becomes comparable with the size of the particle. In figure 4(a), the scattering cross sections due to the classical and current dipole and quadrupole moments are shown for \( g = 50 \) nm. The calculated electric dipole contribution to the scattering cross section (see the blue line and blue circles corresponding to the classical and electric-current multipole expansions) exhibits a small narrow peak at \( \lambda = 400 \) nm corresponding to the spectral position of the quadrupole resonance (see the black line and black circles resulting from the two expansions). The peak exists independently of the expansion used. The presence of this peak in the dipole response would be impossible for a single isolated nanosphere scattering an optical plane wave, because the dipole and quadrupole excitations are orthogonal to each other. In the presence of the second particle, the quadrupole mode hybridizes with the dipole, which can be explained by the fact that the dipole/quadrupole radiation by the first particle can excite a quadrupole/dipole moment in the second particle. This leads to a significant enhancement of the overall scattering within a narrow band centered at \( \lambda = 400 \) nm (see the red line). The total scattering cross section (red line) that includes all possible multipole excitations in the particle is seen to be nearly exactly equal to the sum of the contributions from the electric dipole (blue line) and electric quadrupole (black line). The next most significant multipole moment, the magnetic dipole moment, can still be neglected (see the magenta line), and therefore, the classical and current quadrupoles have identical total contributions to the scattering. For \( g = 10 \) nm (see figure 4(b)), the influence of the quadrupole excitation on the dipole scattering is even more pronounced. In addition, the quadrupole resonance is slightly red shifted and shows a small side lobe. The hybridization also
shifts the two peaks corresponding to the dipole and quadrupole scattering to 405 and 605 nm (see the blue and black curves).

To obtain a more detailed picture of the multipole hybridization, we calculate the scattering contributions for all the non-negligible components of the excited current multipole moments. Figures 5(a) and (b) show the spectra of the contributions of the dipole and quadrupole moments, respectively. The closeness of the second particle breaks the symmetry of the single-particle interaction with the incident light. As a result, the quadrupole excitation at $\lambda = 400$ nm gives rise to a small longitudinal dipole moment, $p_z$. Moreover, a significant excitation of a new quadrupole moment, $Q_{xx}$, appears. We note that this moment cannot be excited in an isolated nanoparticle due to the symmetry of the interaction. Now, that the overall incident field for the particle is highly inhomogeneous, the moments $p_x$ and $Q_{xx}$ become coupled to each other, resonating together. As a result, a bright and narrowband feature appears in the overall scattering at $\lambda = 400$ nm. When the gap between the particles decreases to 10 nm (see figures 5(c) and (d)), the scattering due to $Q_{xx}$ becomes the most significant among the quadrupolar moments and it strongly hybridizes with $p_x$. We remind that we have set $Q_{zz}$ to zero, which can influence the relative strengths of the calculated quadrupole moments. The dipole scattering peaks in figure 5(c) are observed to be shifted to the red. The spectral changes of the dipole
Figure 6. The distributions of the electric current density (color shows the amplitude and arrows represent the instantaneous electric field vectors) on the surface of silver nanosphere separated from another identical nanosphere by $g = 10$ nm. The excitation wavelengths are 405 nm in (a) and (b), 435 nm in (c) and (d), and 605 nm in (e) and (f). These wavelengths are marked by green circles in figure 4(b). The electric current distributions in the right column are separated from those in the left column by a phase delay of $\pi/2$. The dashed contour in each plot indicates the location of the second nanosphere and quadrupole scattering for a range of values of $g$ are shown in figures 5(e) and (f), respectively. The peak close to $\lambda = 400$ nm barely shifts, while the wide dipole peak moves to the red by more than 10 nm.

To verify the results presented in figures 4 and 5, we have plotted the instantaneous distributions of the electric current density on the surface of a single nanosphere separated from the other nanosphere by a 10 nm gap at the wavelengths of 415, 435, and 605 nm marked by the green circles in figure 4(b). The plots projected onto the $xz$ plane are shown in figure 6. The amplitude and instantaneous local vector of the current density are represented by colors and arrows, respectively. At $\lambda = 415$ nm (see figures 6(a) and (b)), the current density exhibits a mixture of multipole excitations $p_x$, $Q_{xx}$, $Q_{xz}$, and $Q_{zx}$. The former two are clearly seen in figure 6(a), while the latter two can be observed in figure 6(b) (the two distributions are separated by a phase delay of $\pi/2$). At $\lambda = 435$ nm (see figures 6(c) and (d) that differ by a phase of $\pi/2$), the multipole excitations are weaker and exhibit moments $p_x$ and $Q_{xx}$ clearly observed in figure 6(c) and moments $p_z$, $Q_{xz}$, and $Q_{zx}$ observed in figure 6(d). At $\lambda = 605$ nm, the excitation had to show a nearly pure dipole character, and this is what is indeed observed in figures 6(e) and (f) (in the first profile, the instance of time corresponds to the maximum of the average current density, and the second profile is phase shifted by $\pi/2$).

The strong and narrow peak in the scattering spectrum caused by the hybrid dipole-quadrupole resonance can be characterized by a FOM that compares it with the dipole resonance. The FOM is a product of three factors: (1) the factor, by which the peak is narrower than the dipole-resonance peak, i.e. $\Delta \nu_d/\Delta \nu_h$, (2) the factor, by which the peak is larger than the dipole-resonance peak, i.e. $C_h/C_d$, and (c) the factor showing the difference between the peak value of the hybrid resonance and the minimum value of the scattering cross section between the hybrid and dipole resonances, i.e. $C_h - C_{\text{min,h}} \rightarrow \nu_d$. The last factor
Figure 7. (a) The figure of merit (FOM) calculated using equation (26) for the hybrid dipole-quadrupole resonance as a function of gap \( g \). (b) The total (red line), dipole (blue line), and quadrupole (black line) scattering cross sections for \( g = 32 \, \text{nm} \).

Figure 8. The spectra of the overall scattering cross sections of the system of two nanoparticles with a gap of 10, 32, and 50 nm (red, blue, and magenta lines, respectively) illuminated with a TM-polarized plane wave. The spectrum calculated for an isolated nanoparticle is also included in the figure for reference (see the solid black line).

quantifies the contrast (visibility) of the hybrid resonance peak in the spectrum. Normalizing the last factor such that it is between 0 and 1, we obtain

\[
\text{FOM} = \frac{\Delta \nu d C_h}{\Delta \nu h C_d} \left( 1 - \frac{C_{\min,h-d}}{C_h} \right). \tag{21}
\]

The higher the FOM, the more useful the hybrid resonance can be for spectroscopic applications. Figure 7(a) shows the calculated FOM as a function of the gap between the particles. The function reaches its maximum at \( g = 32 \, \text{nm} \). When the gap becomes smaller than this value, the hybrid dipole-quadrupole peak becomes wider and splits into more peaks. The scattering spectrum corresponding to \( g = 32 \, \text{nm} \) is shown in figure 7(b).

Experimentally, it can be difficult to separate the contribution of one nanoparticle to the scattering when it interacts with another nanoparticle, because the total scattered field is a superposition of the fields scattered by the two nanoparticles individually. Therefore, we have numerically computed the spectra of the scattering cross sections for the nanoparticle pairs considered above with interparticle gaps of 10 nm, 32 nm, and 50 nm (see the dashed lines in figure 8). These spectra can be compared with the spectra in figures 4 and 7(b) obtained for a single nanoparticle in the presence of the other nanoparticle. The hybridized peak at \( \lambda \approx 400 \, \text{nm} \) is clearly seen also in these new spectra. However, its height relative to the dipole peak is reduced due to a constructive interference of the fields scattered by the particles at the dipole resonances (note that
the dipole peak is about 4 times larger than that of an individual particle of the two-particle system, but only 2 times larger that of an isolated particle). By varying the gap between the nanoparticles and measuring the spectra presented in figure 8, one can experimentally verify our predictions. The strong and relatively narrow peak at $\lambda = 400$ nm—especially pronounced at $g = 10$ nm—is accompanied by a high local field enhancement in the gap between the particles, which can be used for spectroscopic molecular sensing.

### 3.2. Multipole excitations by TE-polarized light

We have also evaluated the multipole excitations in a single nanoparticle in the presence of another nanoparticle for TE polarized incident light. If the separation between the particles is large, the multipole scattering spectra are the same as those of an isolated particle, as in the previous case (see figure 3(a)). However, the induced multipole moments are now $p_y$, $Q_{yz}$ and $Q_{zy}$. In figure 9(a), the spectra of the scattering cross sections due to the excited multipole moments are shown for $g = 50$ nm. These spectra do not exhibit any significant multipole hybridization. Indeed, the excited currents at this polarization are nearly perpendicular to the line joining the particles. As a result, the enhanced near-field of each particle does not enter anyhow considerably the second particle. Even when the separation is reduced to 10 nm (see figure 9(b)), the influence of the quadrupole excitation on the dipole moment stays insignificant. Instead of hybridization, one can observe a weak Fano-like interference between the excited modes, which is visible at both $g = 50$ nm and $g = 10$ nm. However, this interference does not significantly affect the cross section magnitudes. The overall scattering cross sections for the systems of the two particles are shown in the insets of figures 9(a) and (b). Interestingly, for $g = 10$ nm, the presence of the second particle significantly reduces the dipole contribution to the scattering cross section, making the quadrupole peak slightly more pronounced in the overall scattering spectrum. However, no enhancement of narrowband scattering is observed, as opposed to the case of TM-polarized incident light that was considered in the previous section.

### 4. Conclusions

In this paper, we have studied electromagnetic multipole excitations in a silver nanosphere interacting with another identical nanosphere via scattering of light from an incident plane wave. Each particle experiences an inhomogeneous optical excitation, as a result of which the usually independent orthogonal multipoles loose their independence. We show that a TM-polarized incident wave excites a hybridized dipole-quadrupole resonance that is as bright as the dipole resonance, but with a much smaller spectral width. This may be of interest for applications in spectroscopy and optical sensing. Our results clearly show that, when considering the multipole-multipole interaction of closely spaced nanoparticles, one must take into account also hybridized multipole resonances. To the best of our knowledge, these hybrid resonances have not been addressed in the previous research as significant optical excitations. Their further exploration can be especially important in the analysis of light-matter interactions in nanoparticle arrays and optical nanomaterials based on them. These studies can have a potential for discovering new effects of both practical and fundamental importance.
Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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Appendix

We have used equations (35)–(48) of [3] to obtain equations (12)–(20). By neglecting the octupolar terms, the multipole expansion coefficients can be written in terms of the current multipole moments as

\[ a_E (2, \pm 1) = 3C_2 \left[ Q_{xx} - Q_{yy} \mp i \left( Q_{xy} + Q_{yx} \right) \right], \]

\[ a_E (2, 0) = \sqrt{6} C_2 \left[ 2Q_{xy} - Q_{xx} - Q_{yy} \right], \]

\[ a_E (1, 1) = C_1 \left[ p_x + ip_y \right], \]

\[ a_E (1, 0) = \sqrt{2} C_1 p_z, \]

\[ a_M (1, 1) = 5C_2 \left[ -Q_{xx} + Q_{yy} \mp i \left( -Q_{xy} + Q_{yx} \right) \right], \]

\[ a_M (1, 0) = 5\sqrt{2} C_2 \left[ -Q_{xy} + Q_{yx} \right]. \]

These equations make it possible to obtain the scattering cross sections due to each component of the current multipole moments. For instance, using equation (11), one can calculate the scattering cross section due to \( Q_{xx} \), setting all the other components to zero. The result is

\[ C_s (Q_{xx}) = \frac{5\pi}{k^2} \left[ |a_E (2, 2)|^2 + |a_E (2, -2)|^2 + |a_E (2, 0)|^2 \right]. \]

Each multipole coefficient in this equation can be expressed in terms of \( Q_{xx} \), using equations (A1) and (A3) with all the elements except \( Q_{xx} \) being set to 0. This gives

\[ C_s (Q_{xx}) = \frac{120\pi}{k^2} |C_2 Q_{xx}|^2, \]

which is the same as equation (19). Equations (18)–(20) are derived analogously.

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