Controllable optical resonances and unidirectional scattering by core-shell nanoparticles

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Abstract. Nanoparticles supporting a distinct series of Mie resonances have enabled a new class of nanoantennas and provide efficient ways to manipulate light at the nanoscale. The ability to flexibly tune the optical resonances and scattering directionality are particularly essential for various applications ranging from biosensing to nanolasers. In this paper, we investigate the core-shell nanoparticles that support both electric and magnetic Mie resonances and for the first time systematically reveal the mode evolution from a pure high-index dielectric nanosphere to its plasmonic counterpart. Abrupt mode transition and hybridization of Mie resonances are found in Ag-dielectric core-shell spheres when core-shell ratio increases from 0.4 to 0.5. Furthermore, by engineering the electric and magnetic resonances, we demonstrate the unidirectional forward and backward scattering in such a system and reveal its tunability via geometric tuning.

Keywords: Mie scattering, electrical and magnetic resonance, Kerker condition, unidirectional scattering.

1. Introduction

Light-matter interaction is one of the most prominent fields in the scientific research, as it lies at the core of enormous applications in our modern society today ranging from smartphone's screen, signal processing, aerial to solar panel and nanolasers [1-5]. Starting from the seminal work of Mie [6], light scattering by small particles has attracted great amount of attention in many fields of physics [7] throughout the decades. Together with the recent development in nanoscience and nanofabrication, it further spurs the emerging field of meta-optics [8], where various new physical phenomena such as optical magnetism, anapole state, and unidirectional scattering [9-11] are discovered by delicately designing nanostructures and manipulating the multipolar Mie resonances as well as their mode interferences. Despite the tremendous research effort investigating metallic, dielectric, and hybrid nanostructures respectively [12-15], there is still a lack of systematic studies on how the optical resonances and associated physical phenomena change within such a system.

In this paper, we systematically investigated the mode evolution of multipolar Mie resonances within an Ag/dielectric core-shell nanospheres. The physical origins of the electric dipole (ED) and the magnetic dipole (MD) resonances in such a hybrid system are revealed and found directly correlated to the core-shell ratio. By applying the new systematic understanding of the system, we further demonstrate
the intriguing nonradiating state and unidirectional scattering within the Ag/dielectric nanostructure and rigorously discuss how these phenomena could be realized via geometric tuning.

2. Results and discussion

2.1. Isolated dielectric and plasmonic nanospheres

We start our analysis by investigating the optical resonances within homogeneous dielectric and plasmonic nanoparticles positioned in the vacuum. We set the radius of the spheres to 80nm and calculate their scattering response via analytical Mie theory [7] (See methods). As a representative example, the refractive index of the dielectric particle is set to 3.5 while the optical constants of the plasmonic particle is taken from Ag [16].

![Figure 1](image)

Figure 1. Scattering spectra and their multipolar contribution of (a) the dielectric and (b) Ag nanospheres with a radius $r = 80$nm in the wavelength range of 0.2-1μm including the total scattering (black solid line), the electric dipolar (ED) contribution (red solid line), the magnetic dipolar (MD) contribution (blue solid line), the electric quadrupolar (EQ) contribution (red dotted line), and the magnetic quadrupolar (MQ) contribution (blue dotted line). The intersections between ED and MD contribution at incident wavelength $\lambda = 538$ and 642 nm are marked.

Figure 1 depicts the scattering efficiency of the dielectric and plasmonic nanospheres. Here the scattering efficiency $Q_{sca}(\lambda)$ is defined as the ratio between the scattering cross section $\sigma_{sc}(\lambda)$ and the geometric cross section $\sigma_{geo}$, which could be written as $Q_{sca}(\lambda) = \sigma_{sc}(\lambda)/\sigma_{geo}$. For the high-index dielectric nanoparticle, its scattering response clearly manifests the contribution from all individual Mie resonances, i.e., the electric dipole (ED), magnetic dipole (MD), electric quadrupole (EQ) and magnetic quadrupole (MQ). These intriguing resonances are attributed to the matching frequencies between the incident light and the oscillating bounded electrons [17]. For instance, when the incident wavelength satisfies $\lambda \approx 2nr$, a circulating displacement current is formed and thus introduces the MD resonance. By contrast, the optical magnetism in the dielectric particle does not occur in the plasmonic nanoparticle, as showed in Figure 1(b). This illustrates the intrinsic physical differences in the scattering processes of the two particles, where the metallic nanoparticles exhibit oscillating free electrons and thus can only supports ED and EQ resonances introduced by the localized surface plasmons. Such behaviors have been widely used in nanoantennas and surface-enhanced Raman scattering [18-19].

In addition to the optical resonances, we also consider the radiation patterns of the scattering in the far-field, as displayed in Figure 2. Representative far-field scattering graphs of dipolar resonances are showed in Figure 2(a) and (b), corresponding to the MD of the dielectric nanosphere and the ED of the Ag nanosphere at $\lambda = 0.538$μm. Figure 2 (e) shows the schema of incident light, which is considered to
have propagation vector (k) along x–direction, electric (E) and magnetic (H) field vectors in y– and z–directions, respectively, with θ and φ being the azimuthal angle and the polar angle.

Figure 2. 3D far-field scattering patterns of the dielectric and plasmonic nanospheres at (a) MD of the dielectric nanoparticle at λ = 0.538 μm. (b) ED of the Ag nanoparticle at λ = 0.538 μm. (c, d) First (λ = 0.642 μm) and second (λ = 0.538 μm) Kerker conditions of the dielectric nanoparticle, corresponding to the enhanced unidirectional forward and backward scattering. (e) Model depicting scattering of incident plane wave by a spherical nanoparticle.

Fig.2(c) represents the first Kerker condition [17,20], which happens with indicating a strong forward scattering and a complete suppression of backward scattering, and this can be described as complete azimuthal symmetry [21]. Given the negligible EQ and MQ contribution, here we only consider the first-order resonances (ED and MD), so the total electric field is [22]

$$E_{far}(r) = \frac{k^2}{4\pi \varepsilon_0} p_x e^{ikr} \left(-\sin\varphi\hat{\phi} + \cos\theta \cos \varphi \hat{\theta}\right)$$

$$- \frac{Zk^2}{4\pi} m_y e^{ikr} \left(\cos \theta \sin \varphi \hat{\phi} - \cos \varphi \hat{\theta}\right)$$

where r, φ, θ are spherical coordinates, p_x is electric dipole moment and m_y is magnetic moment. Using Eq.1, the backward scattering efficiency is

$$\sigma_{back} = \lim_{r \rightarrow \infty} 4\pi r^2 \left| \frac{E_{far}(\varphi = 0, \theta = \pi)}{E_{inc}} \right|^2$$

$$= \frac{k^4}{4\pi \varepsilon |E_{inc}|^2} \left| p_x - \frac{\sqrt{\varepsilon} m_y}{c} \right|^2$$

where $E_{inc}$ is the incident electric field. From Eq.2, one can find that if $p_x - \frac{\sqrt{\varepsilon} m_y}{c} = 0$, then $\sigma_{back} = 0$, and this is the first Kerker condition [17].

While, fig.2(d) represents the second Kerker condition, showing a strong backward scattering and an incomplete suppression of forward scattering, derived from incomplete azimuthal symmetry[21], and leaves forward scattering efficiency to [23]

$$\frac{d\sigma_{sca}}{d\Omega}(0^\circ) = \frac{16k^{10}}{9} \left| \frac{\alpha_e}{\varepsilon_s} \right|^4$$

where $\alpha_e = p_x / (\varepsilon_0 |E_{inc}|)$, $\varepsilon_0$ and $\varepsilon_s$ are the permittivity of the nanosphere and surrounding medium (vacuum for our research).
It is worth noting that such phenomena of vanishing backward or forwards scattering could also be realized at shorter wavelengths when quadrupole terms are considered, naming the generalized Kerker condition [22].

2.2. Ag-Si core-shell nanoparticles

2.2.1. Mode evolution of electric and magnetic dipolar resonances. After examining the fundamental scattering characteristics of isolated dielectric and plasmonic nanospheres, we then move our attention to the core-shell nanoparticles where the two systems co-exist and the interplay of all Mie resonances emerges. Following up on our previous analysis, we study the Ag-core-dielectric-shell sphere with a fixed radius of 80 nm. The core-shell ratio is then continuously varied from 0 to 1 to demonstrate the transition from a pure dielectric nanosphere to a plasmonic one, and the interaction between the two co-existed constituents. Here, a core-shell ratio of 0 means a pure dielectric nanoparticle and a core-shell ratio of 1 means a pure Ag nanoparticle. The refractive index for Si and Ag is the same as those in the previous section. Figure 3 shows the scattering response of ED and MD resonances varied with core-shell ratio and the incident wavelength.

![Figure 3](image.png)

**Figure 3.** Scattering contribution from ED and MD resonances in Ag-dielectric nanospheres \((r = 80 \text{ nm})\) with a varying core-shell ratio.

Figure 3(a) shows an evident ED resonance emerging around wavelength 0.4-0.5μm when the core-shell ratio is below 0.5. Such an ED resonance is located at the similar spectral position of a pure dielectric sphere as showed in Fig. 1(a). Therefore, we can deduce that it is due to the high-index dielectric shell. With the increase of the core-shell ratio, its wavelength and intensity are both decreasing, and finally vanish when core-shell ratio is over 0.5 where the Ag core starts to become dominating. This is further proved by the ED resonance appearing at the longer wavelengths, whose scattering efficiency is marginal when core-shell ratio is less than 0.4, but soon increases greatly and reaches its maximum when the core-shell ratio is approximately 0.7. The scattering efficiency of such an ED resonance located at the longer wavelengths is much stronger than ED resonance made by Si at the shorter wavelengths. Moreover, the bandwidth of ED resonance at the longer wavelengths also increases synchronously with the core-shell ratio, which can be explained by the increasing Ohmic loss of the system when the lossy Ag-core starts to grow. It is also interesting to point out the spectral location of the ED resonance at the longer wavelengths first exhibits a noticeable red-shift with the increasing core-shell ratio below 0.4 and then undergoes blue-shifts with the further increasing core-shell ratio afterwards. We attribute this intriguing evolution process is to the mode coupling and hybridization between the Ag and Si constituents.

In detail, we find that ED resonance is mainly determined by the dielectric shell when core-shell ratio is less than 0.4, but it quickly transits to an Ag-determined ED when the core-shell ratio increases from
0.4 to 0.5. To capture this process more precisely, Figure 4(a) shows the total scattering efficiency varied with core-shell ratio and wavelength, and Figure 4(b-d) shows the scattering efficiency with core-shell ratio equal to 0.4, 0.45, 0.5 in more detail with corresponding multipole decomposition. At first, we note that the sharp lines in wavelength of 0.4μm or shorter are not considered because they correspond to MQ and EQ resonances.

Figure 4. (a) Total scattering efficiency of the Ag/Si varied with core-shell ratio and the corresponding scattering spectra with multipole decomposition when core-shell ratio is (b) 0.4, (c) 0.45 and (d) 0.5.

From Figure 4 (b-d), we find that when core-shell ratio increases from 0.4 to 0.5, the plasmonic ED originated from Ag quickly increases at the longer wavelengths while the dielectric Si-determined ED remains relatively stable at the shorter wavelengths. The co-existence of the two ED resonances and their interplay with the MD resonances offer us new degrees of freedom to manipulate and exploit the scattering of the particle. For instance, the ED (dominated by Ag) at the longer-wavelengths and MD resonances are now spectrally well-separated and thus allow us to switch between an ED and an MD scattering easily within one physical system. Meanwhile the ED (dominated by dielectrics) at the shorter wavelengths could still interfere with the MD resonances, making the Kerker conditions still accessible. What’s more, at approximately $\lambda = 0.8 \text{ μm}$, we notice that the total scattering efficiency is nearly zero due to the anapole phenomena where the scattering contribution of the electric dipole almost vanishes [24-25], meaning our particle is nearly invisible and transparent at this spectral regime. To further investigate this phenomenon, figure 5 shows the zone where the total scattering efficiency is less than 0.03, corresponding to figure 4 (b-d).
Figure 5. Spectral region when the anapole-induced optical transparency of the system (scattering efficiency < 3%) occurs and the corresponding core-shell ratio.

For MD resonance, MD resonance’s wavelength and scattering efficiency peak remains stable when core-shell ratio is less than 0.4, but after that the wavelength and efficiency peak decrease and finally vanishes when core-shell ratio reaches over 0.95 (Figure 6). From the discussion of ED, we think for MD when core shell ratio is less than 0.4, Ag does not hold the position and $\lambda \approx 2nr$ is approximately available, and then Ag’s mode quickly evolves into main point, due to the decreasing of Si shell, the imaginary part of the refractive index for this core-shell nanoparticle increases rapidly and suppresses MD.

Figure 6. Scattering efficiency when core-shell ratio is 0.95.

2.2.2. Tuning scattering directionality via varying core-shell ratio. Figure 7 shows the Kerker conditions varied with core-shell ratio and incident light’s wavelength. We use ED and MD’s scattering efficiency as our first criterion: if they are equal, then the points meet our criterion satisfied for Kerker conditions. (For each result as core shell ratio increased from 0, 0.1 to 0.8, it really corresponds to its Kerker condition.) To check, we would find the amplitudes and phases of $a_1$ and $b_1$, where $a_1$ and $b_1$ are
the first-order electrical and magnetic scattering efficiency, corresponding to these points. If the amplitudes are equal and the phases are equal or opposite, then these points satisfy Kerker conditions. Here, we only consider first order Kerker conditions, so to avoid EQ and MQ’s interference, Kerker conditions that match λ < 0.4 μm are not considered.

Figure 7. Kerker conditions varied with core-shell ratio and incident light’s wavelength λ. (a) the difference of ED and MD’s amplitude, with black and grey marks for far-field graph in figure 8.; (b) the sum of the phases of a₁ and b₁, with black marks for far-field graph in figure 8; (c) the difference of the phases of a₁ and b₁, with grey marks for far-field graph in figure 8.
From figure 7(a), we can determine the wavelength that matches Kerker condition is close to 0.4 and 0.65 μm with core-shell ratio less than 0.8, corresponding to the result of Figure 1(b) and 2, and its evolution is very similar to MD. From figure 2, we suppose that 7(b) shows the second Kerker condition and 7(c) shows the first Kerker condition. To check our conjecture, we first intermittently find Kerker conditions for core-shell ratio increases from 0.1 to 0.8, step by step with stride 0.1, and we draw all these results’ far-field graphs (first Kerker condition is marked in black in figure 7(a) and 7(c), while second Kerker condition is marked in grey in figure 7(a) and 7(b), see in figure 8) with table 1 (see in Appendix, corresponding to all marks) that should show the results of Kerker conditions in our criteria. Here, if far-field graph for different situations is similar, we combine them to one single figure.

Figure 8. Far-field scattering graph for points that satisfy Kerker conditions. (a), core-shell ratio (cr) = 0.1, \( \lambda = 536 \) nm and cr = 0.5, \( \lambda = 468 \) nm; (b), cr = 0.1, \( \lambda = 642 \) nm; cr = 0.2, \( \lambda = 646 \) nm; cr = 0.3, \( \lambda = 650 \) nm; cr = 0.4, \( \lambda = 650 \) nm; cr = 0.5, \( \lambda = 644 \) nm; cr = 0.7, \( \lambda = 628 \) nm; (c), cr = 0.2, \( \lambda = 532 \) nm and cr = 0.4, \( \lambda = 500 \) nm; (d), cr = 0.3, \( \lambda = 520 \) nm; (e), cr = 0.6, \( \lambda = 412 \) nm; (f), cr = 0.6, \( \lambda = 648 \) nm; (g), cr = 0.8, \( \lambda = 500 \) nm; (h), The relationship of core-shell ratio and scattering efficiency for the first Kerker condition and the second Kerker condition, respectively.
Table 1. Kerker conditions varied with core-shell ratio (for figure 8).

| Core-shell ratio | λ/nm | Amplitude of $a_i$ | Amplitude of $b_i$ | Phase of $a_i$/rad | Phase of $b_i$/rad |
|------------------|------|--------------------|--------------------|--------------------|--------------------|
| 0                | 538  | 0.3480             | -0.9398            | 0.9398             |                    |
|                  | 642  | 0.1017             | -1.2462            |                    |                    |
| 0.1              | 536* | 0.3338             | -0.9549            | 0.9549             |                    |
|                  | 642**| 0.1016             | -1.2464            |                    |                    |
| 0.2              | 532***| 0.3135            | -0.9764            | 0.9764             |                    |
|                  | 646**| 0.0854             | -1.2742            |                    |                    |
| 0.3              | 520  | 0.2777             | -1.0151            | 1.0151             |                    |
|                  | 650**| 0.0661             | -1.3098            |                    |                    |
| 0.4              | 500***| 0.2618            | -1.0323            | 1.0323             |                    |
|                  | 650**| 0.0491             | -1.3448            |                    |                    |
| 0.5              | 468* | 0.2711             | -1.0209            | 1.0209             |                    |
|                  | 644**| 0.0334             | -1.3819            |                    |                    |
| 0.6              | 412  | 0.3048             | -0.9835            | 0.9835             |                    |
|                  | 648  | 0.0083             | -1.4224            |                    |                    |
| 0.7              | 628**| 0.5114             | 0.5587             | 1.5461             | -1.5619           |
| 0.8              | 500  | 0.0212             | 0.0206             | 1.3457             | -1.4081           |

Note: all situations marked * are combined in figure 8 (a), ** are combined in figure 8(b), and *** are combined in figure 8(c).

After contrasting figure 7 with figure 8, we find that with the increasing of core-shell ratio, the first Kerker condition’s wavelength remains stable with small changes but not corresponds to figure 7(a) and (c) exactly, while the second Kerker condition’s wavelength decrease quicker and quicker, its evolution really like MD resonance, but correspond to figure 7(a) and 7(b) exactly. For scattering efficiency, the first Kerker condition’s scattering efficiency gets weaker continuously, while the second Kerker condition’s scattering efficiency first gets weaker and then stronger (figure 8 (h)). For core-shell ratio more than 0.9, we can’t find Kerker condition in our criteria because Si’s ED resonance is too weak to compare with total’s MD or Ag’s ED resonance, the same as figure 7 (a) shows, to reach Kerker condition, EQ and MQ must participate, which corresponds to $\lambda < 0.4$ $\mu$m. Moreover, when core-shell ratio is 0.7 and 0.8, there are only one point matches our first criterion, respectively, and they represent the second Kerker condition. In other words, our nanoparticle can only reach one Kerker condition, neither like dielectric nanoparticle nor similar to plasmonic one, yet their scattering efficiencies are much below than 1, so the scattering light is too weak to use for practical purpose. There is also something that does not match figure 7 in figure 8, like figure 3 (e), which is not a far-field graph of Kerker condition, indicating that high-order scattering participates and couples with it; as figure 8 (g) and (h) show, sometimes a nearly completely suppression of inverse scattering happens in the both sides, not always like figure 2.

3. Conclusion

We demonstrate the evolution of ED, MD resonance and unidirectional scattering with different core-shell ratio in a single Ag-Si nanoparticle, and we find that ED resonance is mainly determined by Ag and MD resonance is completely determined by Si. They couple with each other, switch the mode in a short range and both their properties can appear in a single nanoparticle, in a positive or negative superposition, like high ED resonance with unidirectional scattering in different wavelengths and directions, and nonradiating state in a small range of core-shell ratio and wavelength which represents conditional invisibility. This means we can control the scattering properties of a core-shell nanoparticle.
by adjusting the core-shell ratio, which has a further interest in nanotechnology with high-level machining, like optical sensing, nanoantenna, nonlinear active media with modulable metasurfaces.

4. Methods
We use these formulas derived from Mie theory to calculate Mie scattering coefficients \(a_n\) and \(b_n\) for pure nanosphere [7]:

\[
a_n = \frac{m \psi_n(mx) \psi_n'(x) - \psi_n(x) \psi_n'(mx)}{m \psi_n(mx) \xi_n(x) - \xi_n(x) \psi_n(mx)}
\]

\[
b_n = \frac{\psi_n(mx) \psi_n'(x) - m \psi_n(x) \psi_n'(mx)}{\psi_n(mx) \xi_n(x) - m \xi_n(x) \psi_n(mx)}
\]

And these to calculate Mie scattering coefficients \(a_n\) and \(b_n\) for core-shell nanosphere:

\[
a_n = \frac{\psi_n(y) [\psi_n'(m_2 y) - A_n \chi_n'(m_2 y)] - m_2 \psi_n(y) [\psi_n(m_2 y) - A_n \chi_n(m_2 y)]}{\xi_n(y) [\psi_n'(m_2 y) - A_n \chi_n'(m_2 y)] - m_2 \xi_n(y) [\psi_n(m_2 y) - A_n \chi_n(m_2 y)]}
\]

\[
b_n = \frac{m_2 \xi_n(y) [\psi_n'(m_2 y) - B_n \chi_n'(m_2 y)] - \xi_n(y) [\psi_n(m_2 y) - B_n \chi_n(m_2 y)]}{m_2 \xi_n(y) [\psi_n'(m_2 y) - B_n \chi_n'(m_2 y)] - \xi_n(y) [\psi_n(m_2 y) - B_n \chi_n(m_2 y)]}
\]

\[
A_n = \frac{m_2 \chi_n(m_2 x) \psi_n'(m_2 x) - m_1 \psi_n'(m_2 x) \psi_n(m_1 x)}{m_2 \chi_n(m_2 x) \psi_n(m_1 x) - m_1 \psi_n(m_2 x) \psi_n'(m_1 x)}
\]

\[
B_n = \frac{m_2 \chi_n'(m_2 x) \psi_n(1) - m_1 \psi_n'(m_2 x) \psi_n(1)}{m_2 \chi_n'(m_2 x) \psi_n(1) - m_1 \psi_n'(m_2 x) \psi_n'(1)}
\]

where \(x = k R_1, y = k R_2, k = 2 \pi / \lambda\) (\(\lambda\) is the incident light wavelength in free space), \(R_1\) is core radius and \(R_2\) is the radius of nanosphere; \(m\) is the refractive index for a pure sphere, \(m_1\) is the refractive index of core and \(m_2\) is the refractive index of shell; \(\psi_n\) is spherical Bessel function of the first kind and \(\psi_n'\) is its derivative function, \(\xi_n\) is the spherical Hankel function of the first kind and \(\xi_n'\) is its derivative function, \(\chi_n\) is the spherical Bessel function of the second kind and \(\chi_n'\) is its derivative function, \(n\) is positive internal, represents 2\(^n\) electric or magnetic multipoles.

Finally, we use these to calculate scattering, extinction and absorption cross section:

\[
C_{sca} = \frac{2 \pi}{k^2} \sum_{n=1}^{\infty} (2n+1)(|a_n|^2 + |b_n|^2)
\]

\[
C_{ext} = \frac{2 \pi}{k^2} \sum_{n=1}^{\infty} (2n+1) \Re(a_n + b_n)
\]

\[
C_{abs} = C_{ext} - C_{sca}
\]

where \(C_{sca}, C_{ext}\) and \(C_{abs}\) is scattering, extinction and absorption cross section, respectively.

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