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# Optical multipole resonances of non-spherical silicon nanoparticles and the influence of illumination direction

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## ABSTRACT

In this work we theoretically study spectral multipole resonances of parallelepiped- and pyramid- silicon nanoparticles excited by linearly polarized light waves. We apply the numerical finite element method to calculate the scattering cross-sections as a function of the nanoparticles geometrical parameters. We use the multipole decomposition approach to explore optical resonances in silicon nanoparticles and the influence of second and third order multipoles to scattering diagrams. In contradistinction to our previous investigations, now we explore effects in near-IR spectral range. Apart from basic study we also obtained non-symmetrical combination of multipole contributions due to illumination from top and bottom sides of pyramids. Our work provides important information about the role of high-order multipoles in the light scattering by non-spherical and non-symmentrical nanoparticles. Our results can be applied, for example, for development of metasurfaces and metamaterials in near-IR spectral range.

**Keywords:** Manuscript format, template, SPIE Proceedings, LaTeX

## 1. INTRODUCTION

Optical properties of single silicon nanoparticles is the interesting and relecant topic nowadays.[1–6] Such scatterers can support the excitation of both electric and magnetic multipole resonances[1–7] and we can control it be changing size, geometry and material properties of particles.[7,8] Recently we investigated optical properties of single nanoparticles in optical[9,10] and terahertz[11] range. In this work we expand our investigations to the near-IR range up to wavelength $\lambda = 1900\ nm$.

Silicon is one of the most suitable materials for optical applications in nanoscale.[1–3,8,12] This material provides high refractive index and neglectable absorption in near-infrared spectral range. However, in visible spectral range absorption increases. In this work we consider scattering of light by nanoparticles, which show resonant optical properties in near-IR spectrum. Moreover, particles' size is significantly smaller compared to incident wavelength, and in this case absorption can be be neglected even in visible range. Therefore, the light extinction is mainly determined by the light scattering effect.

In this work we investigate multipole resonances of cubical and pyramidal nanoparticles excited by linear polarized light waves. Here we pay attention to excitation of high-order multipoles (up to third order) and their role in scattering pattern configuration in near-infrared range. The information about multipole resonances of non-spherical particles is important, e.g., for development of multifunctional nanoantennas. Our theoretical study is based on numerical finite element method (COMSOL Multiphysics) and on the semianalytical multipole decomposition method.[13,14]

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## 2. THEORETICAL BACKGROUND

In this work we are using the semi-analytical method as in Ref.[10,13] However, according to recent theoretical suggestions, multipole expressions beyond the long-wavelength approximation can be considered.[15] In this approach total electric dipole (TED) moment of a scatterer can be calculated using following equation:

$$\mathbf{D} = \int \varepsilon_0(\varepsilon_p - \varepsilon_d)\mathbf{E}(\mathbf{r}')j_0(kr')d\mathbf{r}' + \frac{i\omega}{2}\int \varepsilon_0(\varepsilon_p - \varepsilon_d)\{\mathbf{r}'^2\mathbf{E}(\mathbf{r}') - 3(\mathbf{r}'\cdot\mathbf{E}(\mathbf{r}'))\mathbf{r}'\}\frac{j_2(kr')}{(kr')^2}d\mathbf{r}'. \tag{1}$$

where $\varepsilon_0$, $\varepsilon_p$, $\varepsilon_d$ are the vacuum dielectric constant, relative dielectric permittivity of the nanoparticle, and relative dielectric permittivity of the surrounding medium (here we consider $\varepsilon_d = 1$), respectively, $\mathbf{E}$ is the total electric field inside the nanoparticle, $\mathbf{r}'$ is the radius-vector of a volume element inside the scatterer, $k$ is the wavenumber, $j_n$ is the spherical Bessel function of n-th order. This equation includes a combination of electric dipole (ED) moment and toroidal dipole (TD) moment considered in Ref.[9–11,13] Magnetic dipole (MD) moment of a scatterer in this case is

$$\mathbf{m} = -\frac{3i\omega}{2}\int \varepsilon_0(\varepsilon_p - \varepsilon_d)[\mathbf{r}'\times\mathbf{E}(\mathbf{r}')]\frac{j_1(kr')}{(kr')}d\mathbf{r}', \tag{2}$$

electric quadrupole (EQ) tensor of a scatterer is

$$\hat{Q} = 9\int \varepsilon_0(\varepsilon_p - \varepsilon_d)[\mathbf{r}'\mathbf{E}(\mathbf{r}') + \mathbf{E}(\mathbf{r}')\mathbf{r}' - \frac{2}{3}(\mathbf{r}'\cdot\mathbf{E}(\mathbf{r}'))\hat{U}]\frac{j_1(kr')}{(kr')}d\mathbf{r}', \tag{3}$$

magnetic quadrupole (MQ) tensor of a scatterer is

$$\hat{M} = \frac{15\omega}{3i}\int \varepsilon_0(\varepsilon_p - \varepsilon_d)\{[\mathbf{r}'\times\mathbf{E}(\mathbf{r}')]\mathbf{r}' + \mathbf{r}'[\mathbf{r}'\times\mathbf{E}(\mathbf{r}')]\}\frac{j_2(kr')}{(kr')^2}d\mathbf{r}'. \tag{4}$$

To calculate scattering patterns we use far-field scattered power $\mathrm{d}P_{\mathrm{sca}}$ into the solid angle $\mathrm{d}\Omega$,[16] determined by the time-averaged Poynting vector[17]

$$\mathrm{d}P_{\mathrm{sca}} = \frac{1}{2}\sqrt{\frac{\varepsilon_0\varepsilon_d}{\mu_0}}|\mathbf{E}_{\mathrm{sca}}|^2 r^2 \mathrm{d}\Omega\,, \tag{5}$$

where $\mu_0$ is the vacuum permeability, and

$$\mathbf{E}_0^{\mathrm{sca}}(\mathbf{n}) \simeq \frac{k_0^2}{4\pi\varepsilon_0}\left([\mathbf{n}\times[\mathbf{D}\times\mathbf{n}]] + \frac{1}{v_d}[\mathbf{m}\times\mathbf{n}] + \frac{ik_d}{6}[\mathbf{n}\times[\mathbf{n}\times\hat{Q}\mathbf{n}]] + \frac{ik_d}{2v_d}[\mathbf{n}\times(\hat{M}\mathbf{n})]\right) \tag{6}$$

is the scattering amplitude into the direction $\mathbf{n}$ ($\mathbf{n}$ is the unit vector directed along $\mathbf{r}$). Note that tensors $\hat{Q}$ and $\hat{M}$ are symmetric and traceless.[13]

When considering multipole moments up to the magnetic quadrupole moment, the scattering cross-section can be presented as (see[13]for details):

$$\sigma_{\mathrm{sca}} \simeq \frac{k_0^4}{6\pi\varepsilon_0^2|\mathbf{E}_{inc}|^2}|\mathbf{D}|^2 + \frac{k_0^4\varepsilon_d\mu_0}{6\pi\varepsilon_0|\mathbf{E}_{inc}|^2}|\mathbf{m}|^2 + \frac{k_0^6\varepsilon_d}{720\pi\varepsilon_0^2|\mathbf{E}_{inc}|^2}\sum|\hat{Q}_{\alpha\beta}|^2 + \frac{k_0^6\varepsilon_d^2\mu_0}{80\pi\varepsilon_0|\mathbf{E}_{inc}|^2}\sum|\hat{M}_{\alpha\beta}|^2. \tag{7}$$

where $k_0$ is the free-space wave number; $c$ is the light speed in the vacuum; $\mathbf{E}_{inc}$ is the electric field amplitude of the incident light wave. Total scattering cross-section is obtained through the integration of the Poynting vector over a closed surface in the far-field zone and the normalization to the incident field intensity.[16] Total electric fields in scatterers are calculated numerically using COMSOL Multiphysics. Using the calculated electric fields, the multipole moments and their contributions to scattering cross-sections are obtained by a numerical integration.

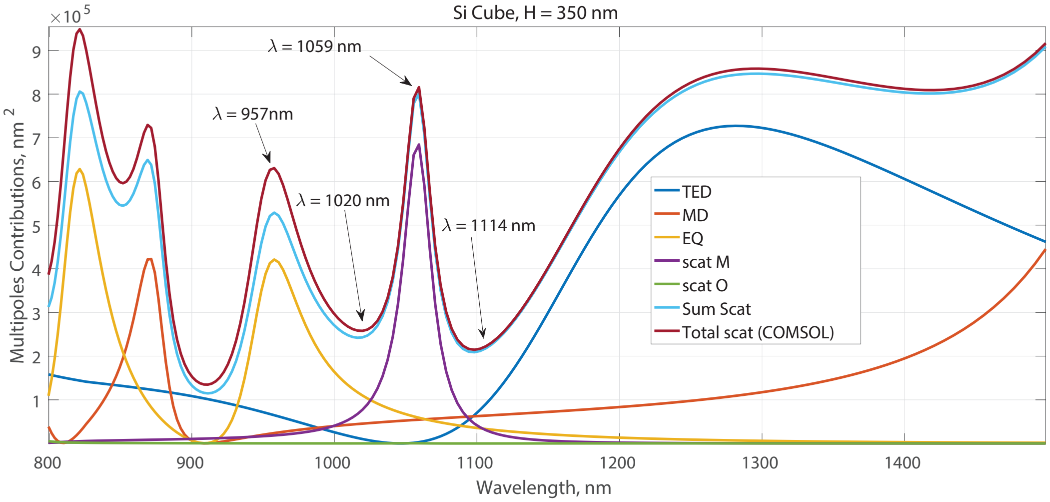


Figure 1. Spectra of the scattering cross sections and corresponding multipoles calculated for the silicon cube (height of H = 350 nm). Sum Scat is the sum of multipoles contributions, Total Scat (COMSOL) is a scattering cross section obtained via direct numerical simulation; other abbreviations can be found in the text. The black arrows in (a) correspond to the spectrum points for which the scattering patterns are presented in Fig. 2.

## 3. LIGHT SCATTERING BY SILICON NANOPARTICLES

In this work we consider two non-spherical shapes of nanoparticles. First of all we study cubical nanoparticle and then expand our research to pyramidal nanoparticles. In contrast to previous works[18–20] here we pay attention to excitation of high-order multipole contributions to the scattering. The dielectric permittivity of silicon was taken from the textbook.[21] In this article we consider that all nanoparticles are placed in a homogeneous surrounding medium with $\epsilon_d = 1$.

### 3.1 Cubical nanoparticles

We start from multipole decomposition of scattering cross-section of cubical silicon nanoparticle with height H = 320 nm. (Fig. 1) Such a particles exhibit interesting resonant properties in optical and near-IR spectral range. There are several special points marked by black arrows; scattering diagrams in these wavelengths are presented in Fig. 2. Figure 1 demonstrates that the higher-order multipole resonances are excited with decreasing the light wavelength. We can also note that for cubical nanoparticle there is no total overlapping of multipole resonances realized. One can clearly see too electric quadrupole moment resonances in wavelengths $\lambda = 821$ nm and $\lambda = 958$ nm. First of them also affected but TED contribution. Narrow MQ resonance appears in wavelength $\lambda = 1059$ nm. Worth to note that according to previous researches[10] of the cube magnetic quadrupole resonant contribution suppresses and even disappears as the symmetry of the cube changes. Well-known dipole resonances can be obtained in wavelength $\lambda > 1150$ nm. So-called second magnetic dipole resonance appears in wavelength $\lambda = 871$ nm between two EQ resonances.

The far-field radiation patterns which correspond to the EQ and MQ resonances are shown in Figs. 2 a,c. One can see that four radiation lobes correspond to forward, backward and side scattering. The direction of side-scattering correspond to the polarization of electric (for EQ) and magnetic (for MQ) fields in incident plane wave.

In Figs. 2 b,d we considered interesting non-resonant properties of silicon nanocube. We can show that for cubical particle in the medium with $\epsilon_d = 1$ we can achieve side-scattering effects. Fig. 2b shows the amplified scattering along the direction of electric field polarization of incident wave. Moreover, in Fig. 2d one can note the symmetric side-scattering effect: interference between dipole and quadrupole moments of both types provide

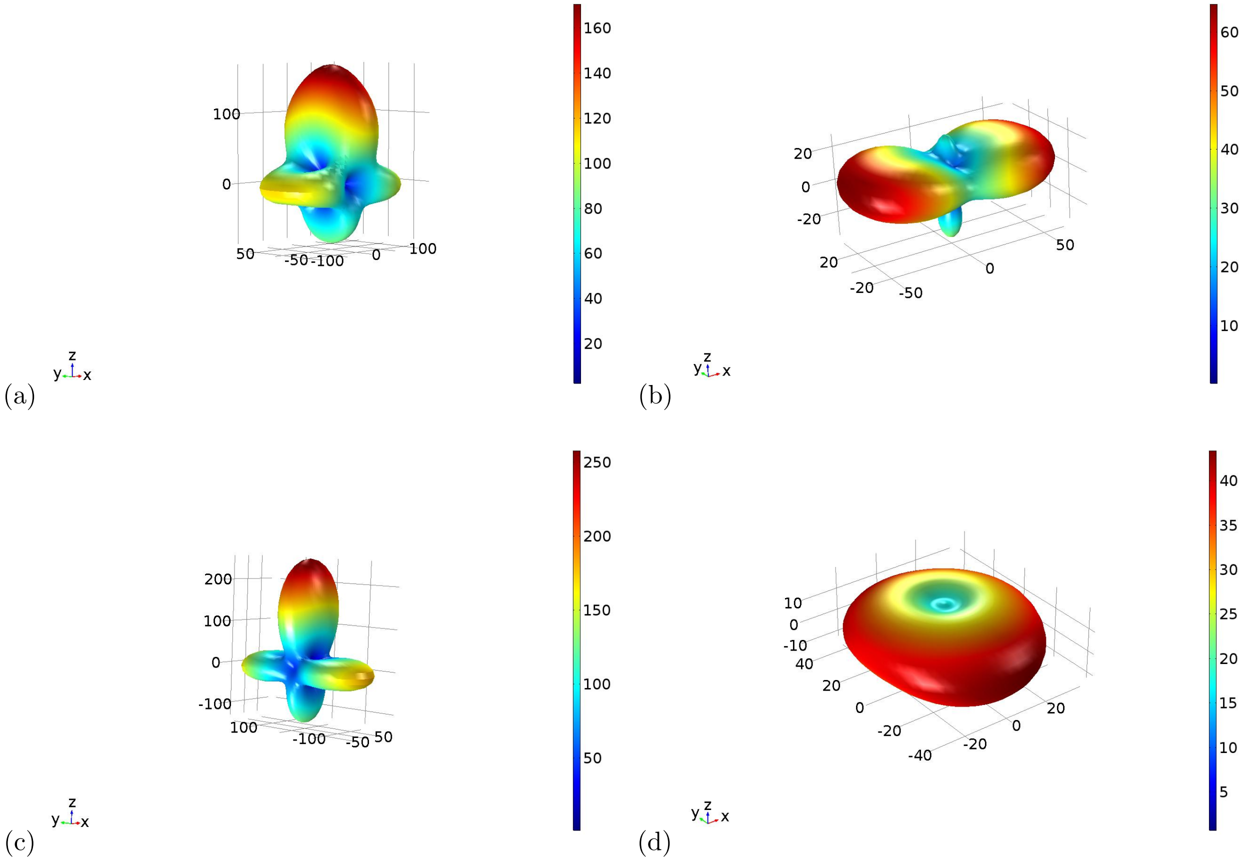


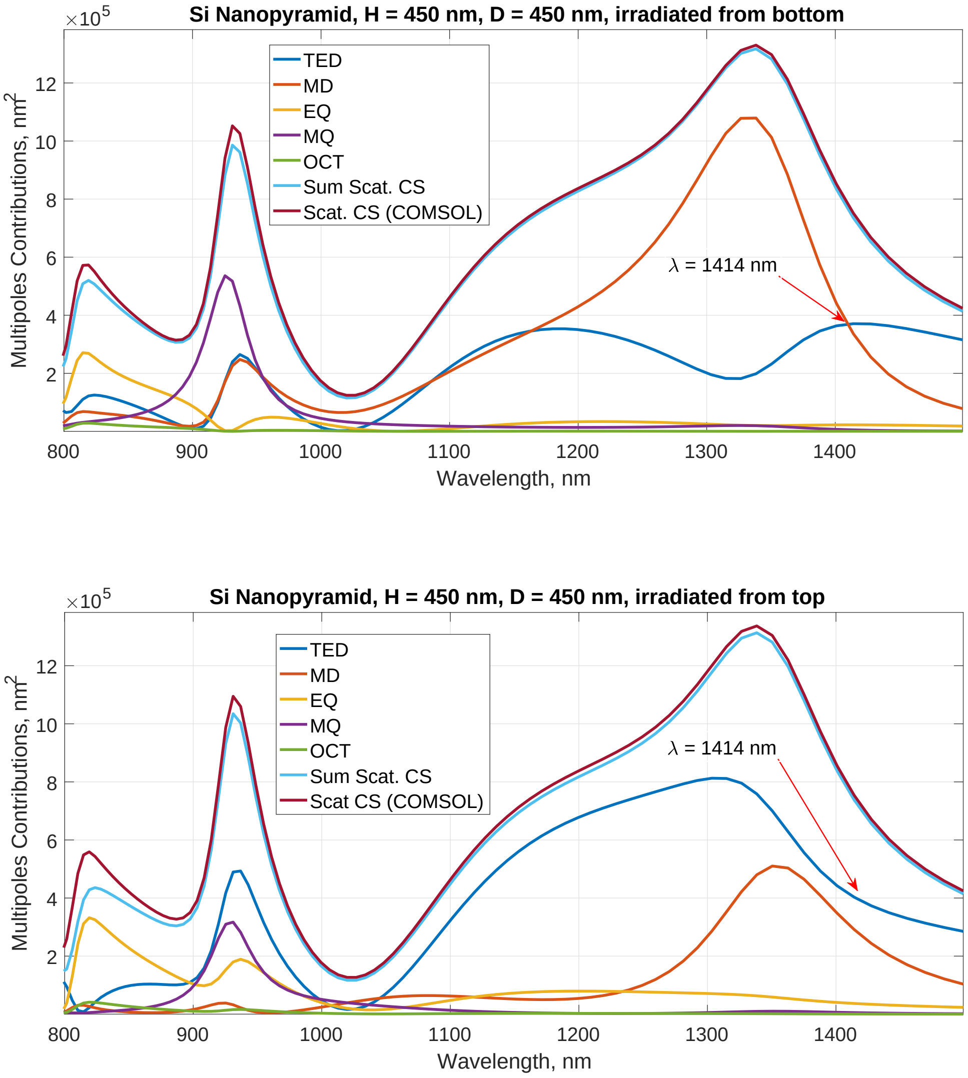
Figure 2. Radiation patterns of the cubic nanoparticle with edge H = 320 nm calculated at the wavelengths indicated by the black arrows in Fig. 1. (a) $\lambda = 957$ nm (b) $\lambda = 1020$ nm (c) $\lambda = 1059$ nm (d) $\lambda = 1114$ nm. k-vector of incident wave is oriented along z, electric field polarization is oriented along x.

simultaneous suppression of forward and backward scattering. At the same time, side lobes of magnetic and electric quadrupole moments result in homogeneous side scattering in the plane, perpendicular to the incident wave direction.

### 3.2 Pyramidal nanoparticles

In this section we expand our study to non-symmentrical pyramidal nanoparticles. We consider silicon nanopyramid with height H = 450 nm and base edge D = 450 nm. Fig. 3a presents the multipole decomposition of such nanopyramid, illuminated by light plane wave from the base edge side. One can see that for pyramidal shape there is no separated MQ resonance. Small resonant contributions of TED and MD also appear in same wavelength range around $\lambda = 925$ nm. There is also the scattering peak in wavelength $\lambda = 810$ nm, explained by joint contribution of EQ, MD and TED moments. The big scatering resonance in the range of $\lambda$ from 1100 to 1500 nm can be explained by joint contribution of TED and MD. In the wavelength $\lambda = 1414$ nm one can see the usual Kerker effect of backward scattering suppression (Fig. 4a). In Fig 3b we present the scattering cross-section of the same particle irradiated from the top of pyramid. It is worth noting that despite total scattering cross-section remains changeless, multipole contributions change sufficiently. Moreover, one can see that magnetic moments contributions decrease, but electric multipole moments contributions increase in the same time.

In Fig. 4 we present the comparison of 2D scattering diagrams in the same wavelength $\lambda = 1414$ nm. In Fig. 4a one can see precise fulfillment of Kerker conditions, which leads to total suppression of backward scattering. However, for the case in Fig. 4b the magnitudes of TED and MD contributions to scattering cross-section are



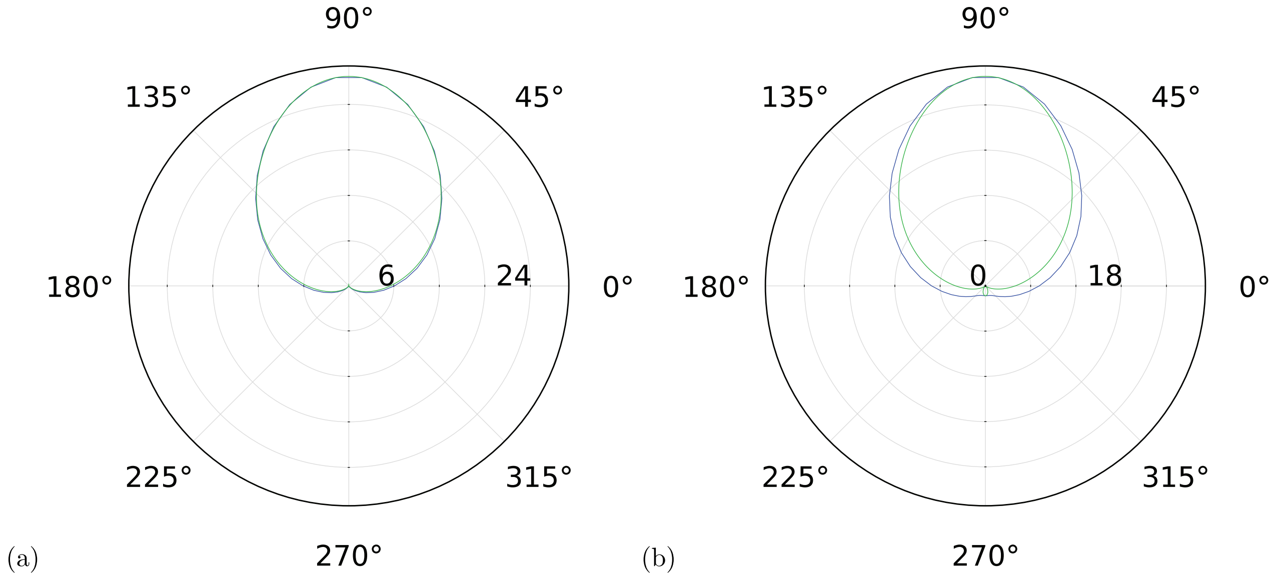
(a)

(b)

Figure 3. Spectra of the scattering cross sections and corresponding multipoles calculated for the silicon nanopyramid (height of H = 450 nm; base edge of D = 450 nm). Sum Scat is the sum of multipoles contributions, Total Scat (COMSOL) is a scattering cross section obtained via direct numerical simulation; other abbreviations can be found in the text. (a) The pyramid is illuminated by light plane waves from the pyramid base edge side.(b)It is illuminated from the pyramid top. The red arrows correspond to the spectrum point for which the 2D scattering patterns are presented in Fig. 4.

slightly different in this point, and Kerker conditions cannot be fulfilled completely. The difference between these two scattering diagrams clearly show that the scattering properties of pyramidal nanoparticles are unsymmetric with respect to incident plane wave direction. Similar properties can be seen in[10] for visible range.

This asymmetry effect can be used for different applications where the scatterer triggered properties controlled by the external illumination are needed, for example, functional bilateral nanoantenas, emitting light diodes, antireflective structures, and surface enhanced phenomena.



(a) (b)

Figure 4. 2D radiation patterns of the silicon pyramidal nanoparticle with height H = 250 nm and edge D = 450 nm calculated at wavelength $\lambda = 1114$ nm (red arrow in Fig. 3. Here 90° corresponds to forward scattering, 270° correspond to forward scattering.

## 4. CONCLUSION

In conclusion, using the multipole decomposition technique, the multipoles up to the third order that were excited by light in cubical and pyramidal silicon nanoparticles were investigated in near-infrared spectral range. It has been demonstrated that peculiar scattering patterns with certain predominant scattering directions can be obtained by tuning the spectral overlap of several multipoles. In particular, using cubic particles it is possible to get a strong isotropic side scattering with simultaneous suppression of the forward and backward scattering. It has been also shown that the effect of the asymmetrical multipole response in pyramidal particles depends on the illumination direction. Our investigation provides important information about the roles of the high order multipoles in the light scattering by nonspherical nanoparticles. We expect that the changing of the wave vector and the polarization from the chosen direction will also affect the scattering properties of considered particles. In addition, since the optical properties of the metamaterials are determined by the properties of their building blocks which are single resonators, we expect that the reciprocity feature will preserve in periodic arrays based on dielectric pyramidal nanoparticles. The results obtained can be used for the development of the nanoantennas in the visible and near-infrared spectral ranges, creating metasurfaces and metamaterials, providing flexible control over light behavior, and for invention and realization of different coatings for optics and photovoltaics.

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