More efficient than a "super" absorption in spherical nanoparticles

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There is a theoretical limit for absorption by a sub-wavelength bulk spherical particle. To overcome this limit we applied a widely used "super" design pattern which superpose several electric and magnetic multipole resonances of a multilayered particle. We used a straightforward approach to evaluate a number of designs from realistic materials. However, we found that due to dimension effect it can be preferable to use a properly designed smaller particle with only a dipole response in order to reach the best absorption efficiency.

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Mie theory [1] describes interaction of an electromagnetic wave with a spherical particle. In spite of its long history lasting over a century it is still of great interest our days [2–8]. Development of Mie theory [9, 10] made it possible to explore properties of multilayered spherical particles [11, 12]. Such particles has various applications in cancer treatment [13, 14] and medical diagnostics [15], cloaking [16–18] and plasmonic [19, 20] devices, study on thermal properties of insulating material [21], solar cells[22, 23], and so on.

The problem of scattering from a multilayered cylinder and a sphere was investigated in great detail with Fan et al. [24, 25]. In his work he defined a "super" scatterer as a sub-wavelength object having a scattering cross section that far exceeds the single-channel limit of the maximal total angular momentum involved. From spectral point of view this means the superposition of several electric and/or magnetic resonances.

There is a similar problem to design highly absorbing sub-wavelength particles. Tribelsky has derived [26] a theoretical limit of a maximum absorption value for a single channel. As a result the absorption coefficients $\tilde{a}_n = \mathrm{Re}\{a_n\} - |a_n|^2$ and $\tilde{b}_n = \mathrm{Re}\{b_n\} - |b_n|^2$ become limited with 1/4 in case of largest possible absorption (where a_n and b_n are scattering coefficient as defined in Mie theory [27]). To overcome a single-channel limit we tried to use similar approach and to tune together several absorption resonances.

We used a triple layered $Si/Ag/Si$ spherical particle with experimental material parameters from Palik [28] illuminated with a plane wave (Fig. TODO). To optimize width of each layer we implemented [29] adaptive differential evolution [30] algorithm named JADE [31]. All the details on the optimization procedure can be found elsewhere [18]. Mie calculations were performed with the Scattnlay [10, 32] software, whose results were verified against a number of other Mie-type codes and commercially available programs Comsol Multiphysics [33] and CST Microwave studio [34].

Initially we tried to maximize contribution of several multipole resonances at a given wavelength $\lambda = 500$ nm. However, best results were obtained with the optimizer set to find maximum of the absorption efficiency factor $Q_{\mathrm{abs}} = C_{\mathrm{abs}}/2\pi R_{\mathrm{total}}^2$, where R_{total} is the outer radius of the particle and C_{abs} denotes its absorption cross-section. This way, the efficiency is defined as absorption cross-section normalized to the geometrical cross-section of the particle.

In order to understand the phenomena we run a series of optimizations; we were steadily increasing the outer size of the particle starting from zero. The dependence of highest absorption efficiency obtained with the optimization procedure on the spherical particle outer radius is depicted in Fig. 1a. With dashed lines we marked the absorption limit of dipole ($n = 1$) and quadrupole ($n = 2$) resonances derived with Tribelsky [26] as

$$Q_{\mathrm{abs\ max}}^{(n)} = \frac{2n+1}{2q^2},$$

where size parameter $q = 2\pi R_{\mathrm{total}}/\lambda$. This limits are obviously beaten for $R_{\mathrm{total}} > 60$ nm, as far as the contribution of higher multipoles with $n > 2$ in negligible

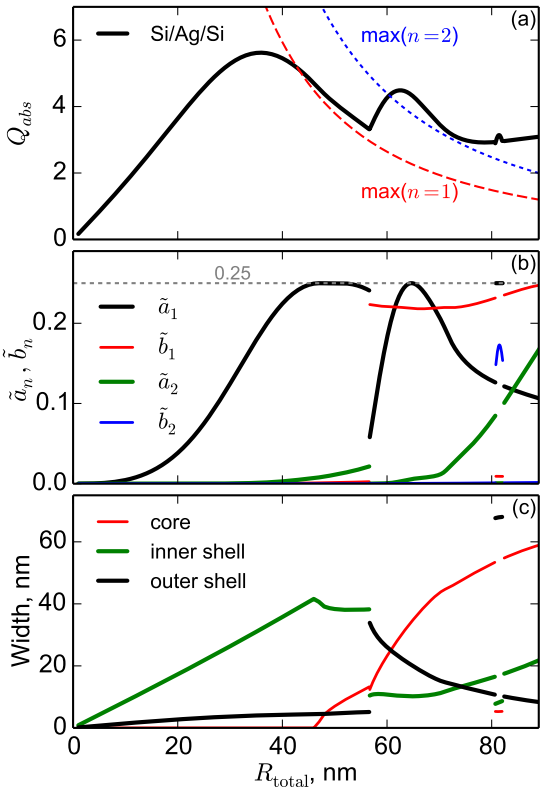

Figure 1. Optimized designs overview at working wavelength $\lambda = 500$ nm. (a) Absorption efficiency with best value at total R=36 nm and Ag/Si design (zero sized core) and "super" designs maxima at R=63 nm and R=81 nm. (b) Absorption coefficients for electric and magnetic dipole ($\tilde{a}_1$ and $\tilde{b}_1$) and quadupole ($\tilde{a}_2$ and $\tilde{b}_2$) modes (c) Used layers width, for total $R < 46$ nm the core width was optimized to be zero, the design become bi-layer Ag/Si particle.

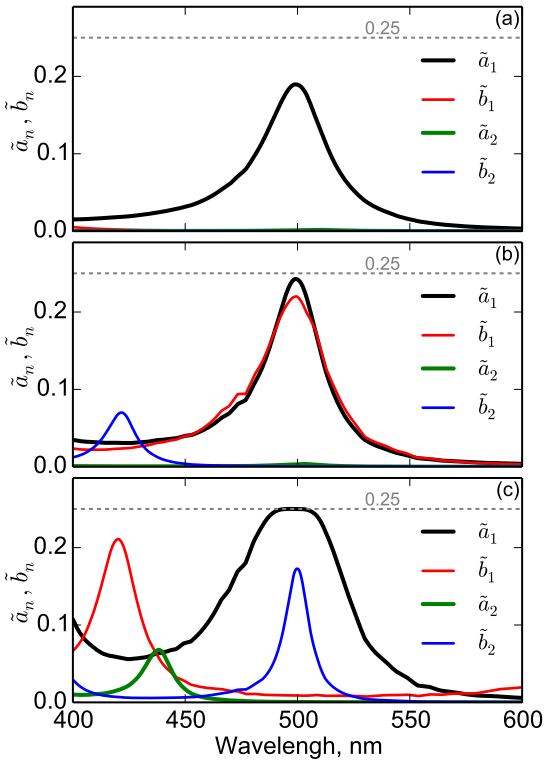

Figure 2. Expansion coefficients spectra of (a) efficient and (b-c) "super" design.

for the plotted range of outer size of the particle. All this designs should be classified as "super" absorbers as it follows from the Fan et al. [24, 25] definition.

In Fig. 1b we present values of absorption coefficients; horizontal dashed line denotes their theoretical limit. For small particles the absorption is dominated with electric dipole $\tilde{a}_1$. ~~At~~ $R_{\text{total}} = 56.6$ nm the optimizer switches to the branch of designs, that combine the usage of electric $\tilde{a}_1$ and magnetic $\tilde{b}_1$ dipoles, ~~as far as~~ they start to outperform the single electric dipole designs branch. There is one more branch of designs using electric dipole $\tilde{a}_1$ and magnetic quadrupole $\tilde{b}_2$; however, it turns to be the best in a very ~~limited~~ range of R_{total} from 80.7 nm to 82.1 nm.

Fig. 1c unveils the fact that dipole design branch has two parts. For $R_{\text{total}} < 46$ nm the optimizer nullifies the innermost layer width (marked as a "core" layer); the particle design was reduces to Ag/Si bi-layer. At $R_{\text{total}} = 46$ dipole channel becomes practically indistinguishable from the theoretical limit (it becomes $\tilde{a}_1 > 0.249$). It looks like the optimizer introduces the inner Si layer in order to keep $\tilde{a}_1$ near the theoretical limit as the R_{total} increases further on. As a side effect quadrupole $\tilde{a}_2$ appears, however, it do not help to reach "super" absorption limit $n = 2$.

The most important feature of Fig. 1 is that the best absorption efficiency was reached in non "super" absorption mode; even more, it was reached before $\tilde{a}_1$ hit the single channel limit. *This way to achieve the best absorption efficiency for a spherical particle there is no need to tune several multipole resonances using complex multilayer structure.* From practical point of view it is even more important that the maxima can be reached in bi-layer structure, instead of triple-layer; it should be easier (and cheaper) to produce such an absorber.

To verify large absorption coefficient obtained during optimization corresponds to a multipole resonance we plotted in Fig. 2 spectra of absorption coefficients for all designs that have a local maxima of Q_{abs} on Fig. 1a. As expected design with maxima at R=36 nm has a single electric dipole resonance with the center at optimized wavelength $\lambda = 500$ nm. Spectra of designs with maximum at R=63 nm and R=81 nm has a "super" structure; there ~~is~~ a superposition of electric and magnetic resonances. These spectra also has some other resonances, however, they are located rather far from the ~~used~~ wavelength.

A noticeable feature of Fig. 2c is an almost flat top of the electric dipole resonance. ~~We do not have any good~~

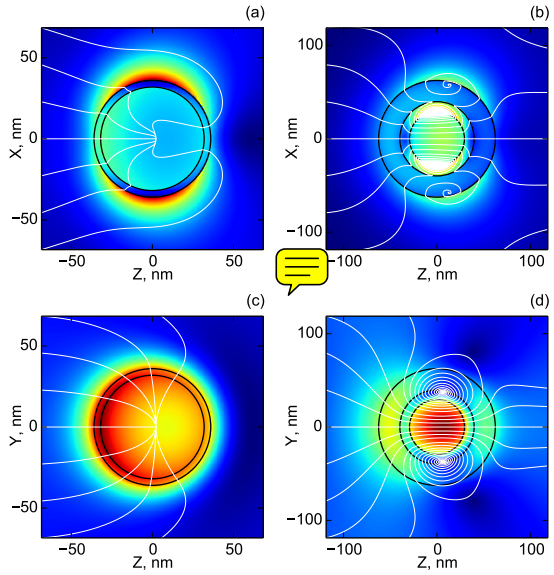

Figure 3. Amplitude of electric field for efficient (a,c) and "super" (b,d) designs in E-k (a-b) and H-k (c-d) planes.

explanation for this phenomena, however, we can suggest that it is due to coupling of the electric dipole $\tilde{a}_1$ with the magnetic quadrupole $\tilde{b}_2$. Whereas resonant responses of other desings originate from coupling of incident plane wave with the corresponding multipole, it is not the case for the design with R=81 nm. Here, the particle is mostly composed from the Si outer shell, that give enough volume for $\tilde{b}_2$. Dipole response comes from the small inner part of the sphere and can not be directly exited with the incident wave. It takes the power from the surrounding $\tilde{b}_2$ mode and very soon it reaches the fundamental limit for absorption. This way we can observe a flat top for the $\tilde{a}_1$ response accompanied with a sigificat value of $\tilde{b}_2$. Our suggestion is indirectly proved by the reduced width of the $\tilde{b}_2$ resonance compared to other quadrupole responses.

Finally, we present distribution for amplitude of electric field (Fig. 3) for two designs: with the best efficiency and a "super" absorber. We set different scale for this two designs so that black circles that denote outer boundary are plotted to be the same size. We also plot streamlines for a Poynting vector, the last one is tangent at each point to the white curve. For the efficient design the power flow goes into the particle. In case of "super" absorption presence of the magnetic response leads to the existence of power flow vortexes, there is some power leaking throw the particle.

This way we conclude, that to design a good absorber it is not necessary to superpose several resonances. The explanation for the phenomena seems to be quite intuitive. Due to spatial structure of higher multipoles, namely presence of nodal points, they are not using efficiently the whole volume of the particle for the absorption. At the same time in 3D the increased absorption of higher multipoles should compete against quadratic growth of the geometrical cross-section. It is clearly not the point for the case under consideration, the most efficient design has simply the smallest radius.

It is interesting, that similar conclusion was made by Miller et al. [35] for extinction of arbitrary particles: small size with only dipole responce is preferable for geometric volume normalized efficiency.

As a final word we would like to notice that the use of stochastic optimizer to design absorbing particles seems to be superior to pure theoretical approach. Grigoriev et al. [36] derived expressions for ideal absorber; however, they considered only a dipole approximation, the final value for our range of particle sizes is very close to the dipole limit predicted with Tribelsky [26]. This way, "super" designs are out of scope from their ideal case; it is easy to check that "super" designs from Fig. 1c gives larger absorption cross-sections. Grigoriev et al. [36] also provide an equation to desing a core-shell structure from predefined materials. However, in case of material parameters that we used for Si and Ag and size parameter from our best design, Grigoriev's equation gives a complex value for relative share of two layers, which is not suitable for most of the simulation software or experiment.

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