# More effective 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, 17] and plasmonic [18, 19] devices, study on thermal conductivities of insulating material [20], solar cells[21, 22], and so on.

The problem of scattering from a multilayered cylinder and a sphere was investigated in great detail with Fan et al. [Fan-PRL, Fan-APL]. 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 [TODO] a theoretical limit of a maximum absorption value for a single channel. As a result the absorption coefficents $\tilde{a}_n = \mathrm{Re}\{a_n\} - |a_n|^2$ 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 [TODO Bohren Huffman]). 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 [TODO] illuminated with a plane wave (Fig. TODO ). To optimize width of each layer we implemented [link to GitHub] adaptive differential evolution [23] algorithm named JADE [24]. All the details on the optimization procedure can be found elsewhere [17]. Mie calculations were performed with the Scattnlay [10] software, whose results were verified against a number of other Mie-type codes and commercially available programs Comsol Multiphisics[TODO] and CST Microwave studio[TODO].

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 $Q_{\mathrm{abs}} = C_{\mathrm{abs}}/2\pi R^2_{\mathrm{total}}$, where $R_{\mathrm{total}}$ is the outer radius of the particle and $C_{\mathrm{abs}}$ 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 dependance 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 quadupole ($n = 2$) resonces derived with Tribelsky[TODO] as

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

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 contri-

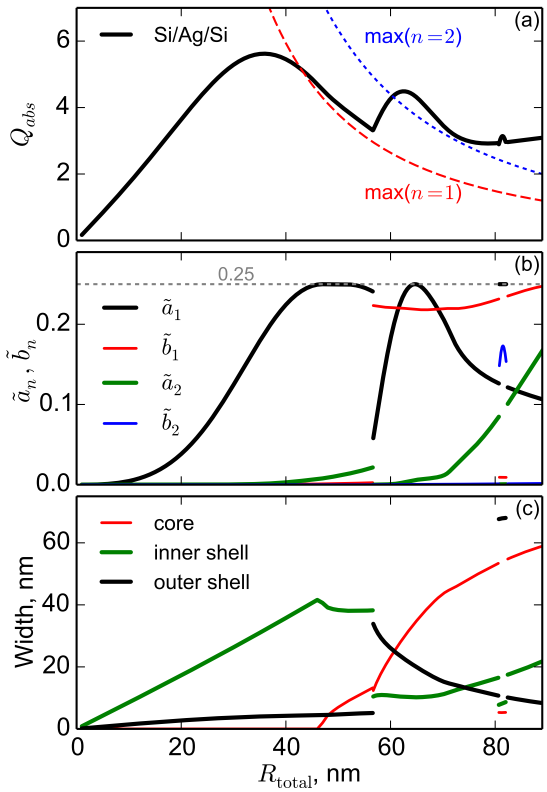


Figure 1. Optimized designs overview at working wavelength $\lambda = 500$ nm. (a) Scattering coefficients (b) Absorption efficiency with best value at total R=36 nm and Ag/Si design (zero sized core) and "super" designs at R=63 nm and R=81 nm. (c) Used layers width, for total $R < 46$ nm the core width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

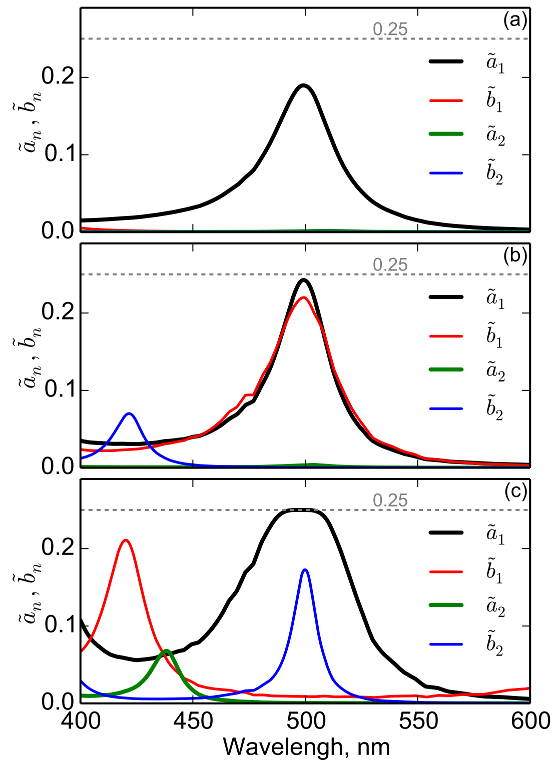


Figure 2. Expansion coefficients spectra of (a) efficient and (b-c) "super" design.

bution of higher multipoles with $n > 2$ in negligible 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. [Fan-PRL, Fan-APL] 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 opimizer switches to the branch of designs, that combine the usage of electric and magnetic 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 $R_{\text{total}}$ from 80.7 nm to 82.1 nm.

Fig. 1c unveils the fact of dipole design branch has two parts. For $R_{\text{total}} < 46$ nm the optimizer nullifies the innermost layers 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 undistiguishable from the theoretical limit (it becomes $\tilde{a}_1 > 0.249$). It looks like the optimizer introduced the inner $Si$ layer in order to keep $\tilde{a}_1$ near the theoretical limit as the $R_{\text{total}}$ increases futher 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 effeciency there is no need to tune several multipole resonances using complex multilayer structure. From prctical 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 obtianed during optimization corresponds to a multipole resonance we plotted in Fig. 2 specta of absorption coefficents for all designs that have a local maxima of $Q_{\text{abs}}$ 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. (TODO add some explanation, why it flat? Any ideas?)

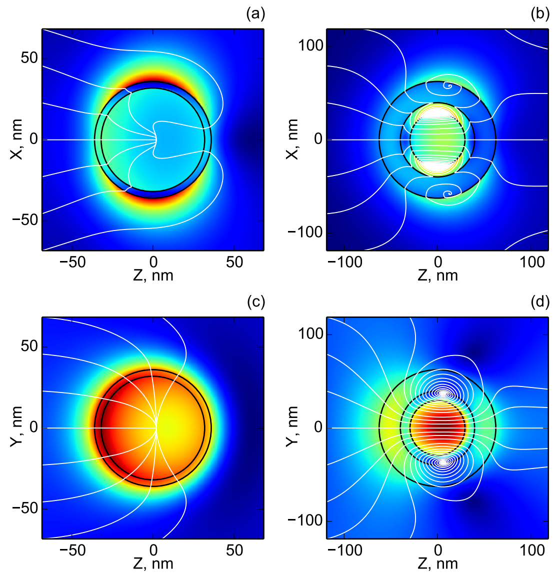


Figure 3. Amplitude of electric field for efficient (a,c) and "super" (b,d) designs in E-k (a-b) and H-k (c-d) planes.

Finally, we present distribution for amplitude of electric field (Fig. 3) for two designs: with the best efficency and a "super" absorber. We set different scale for this two desings so that black cirlces 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 effective 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 resonces. The explanation for the phenomena seems to be quite intuitive. Due to spatial stucture of higher multipoles, namely presence of nodal point, they are not using effectively 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 cleary not the point for the case under consideration, the most effective desing has simply the smallest radius.

It is intersting, that similar conclustion was made by Miller et al [TODO Fundamental Limits to Extinction by Metallic Nanoparticles Phys. Rev. Lett. 112, 123903 Published 26 March 2014 O.D. Miller, C.W. Hsu, M.T.H. Reid, W. Qiu, B.G. DeLacy, J.D. Joannopoulos, M. Soljai, and S.G. Johnson] for extinction of arbitrary particles.

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