

Response Letter

Dear Editor,

I am writing to respond to the reviewers' critical comments on our manuscript entitled, "Superabsorption of light by nanoparticles" NR-COM-08-2015-005468, which we aim for publication in *Nanoscale*.

I would like to thank the editor and two anonymous reviewers for their constructive comments, which helped us to improve the manuscript. Below, we address all comments point-by-point, discussing the subsequent modifications.

Reviewer #1 comments

1. The authors claimed Combined effect of these resonances is presented to produce the flat and relative broadband electric resonance response. Nevertheless, the broadened absorption band is not very broad. Is there any further way to predict a broadband light absorption. For instance, a broadband absorption in the whole visible spectral range. Maybe, a possible way of using the dispersed size scale nanoparticles should be added for improving the study.

This comment touches two aspects of broadband performance, namely, properties of a standalone particle and in interaction with other particles. The first case, as it was correctly mentioned with the reviewer, has some limitations. We can design a band, which will be broader of a typical band for a given multipole, however, its width is not extremely large. To illustrate this we have run an optimization with a goal to provide a predefined separation between multipole resonances (Fig. 1). Our observation is the following: while the separation is small enough compared to a multiple resonance width the optimizer was successful to find designs with electric and magnetic dipole. If the separation is large the absorption band splits into two. For large separation is also possible to obtain two resonances of the electric dipole response, located at predefined position (Fig. 1(e,f)). Still there are not so many possibilities here to design a good broadband. To control the bandwidth at a given spectral position we need to involve relatively large particles. This leads to appearance of resonances (and absorption) out of the designed band which are hard to control or suppress in discussed triple-layer structure.

To answer the second part of the comment, related to possibility of usage of the particle array with dispersed sizes, we designed two particles with best absorption efficiency at 475 nm and 525 nm (outer radius 34 and 38 nm) and simulated them both with FDTD method using Lumerical FDTD Solutions. Sketch of the simulated system and final results are in Fig. 2. To verify our FDTD we first simulated absorption of standalone spheres, the obtained positions and amplitudes of resonances correspond well with Mie calculations. Next step was to find out the possible interactions between two spheres, so we run a simulation in dimer configuration with zero separation between spheres and with 10 and 30 nm separation. The resulting spectra has a strong contribution from standalone resonances, while coupling effects seems to be minor. This can be easily explained from field distribution in Fig. 4(c) of the manuscript. Positioning spheres in H-k plane we are exploiting the feature of field being highly localized inside the sphere. For sure arranging spheres in other plane, increasing the number of spheres can change the amount of coupling between the spheres. Still, here we show that in principle it is possible to construct an absorption band of an array of dispersed particles simply optimizing the properties of every single sphere.

We changed the manuscript from "As a result, one can design spectrally-selective absorbers or broadband absorbers with almost arbitrary prescribed properties." to "As a result, one can design absorbers with broadened spectra or spectrally-selective absorbers with almost arbitrary prescribed properties. Due to high localization of field inside the particle (especially in H-k plane) it is also possible to design arrays of dispersed size particles by defining the properties of individual particles; we obtained rather small coupling between particles during additional simulation using FDTD method."

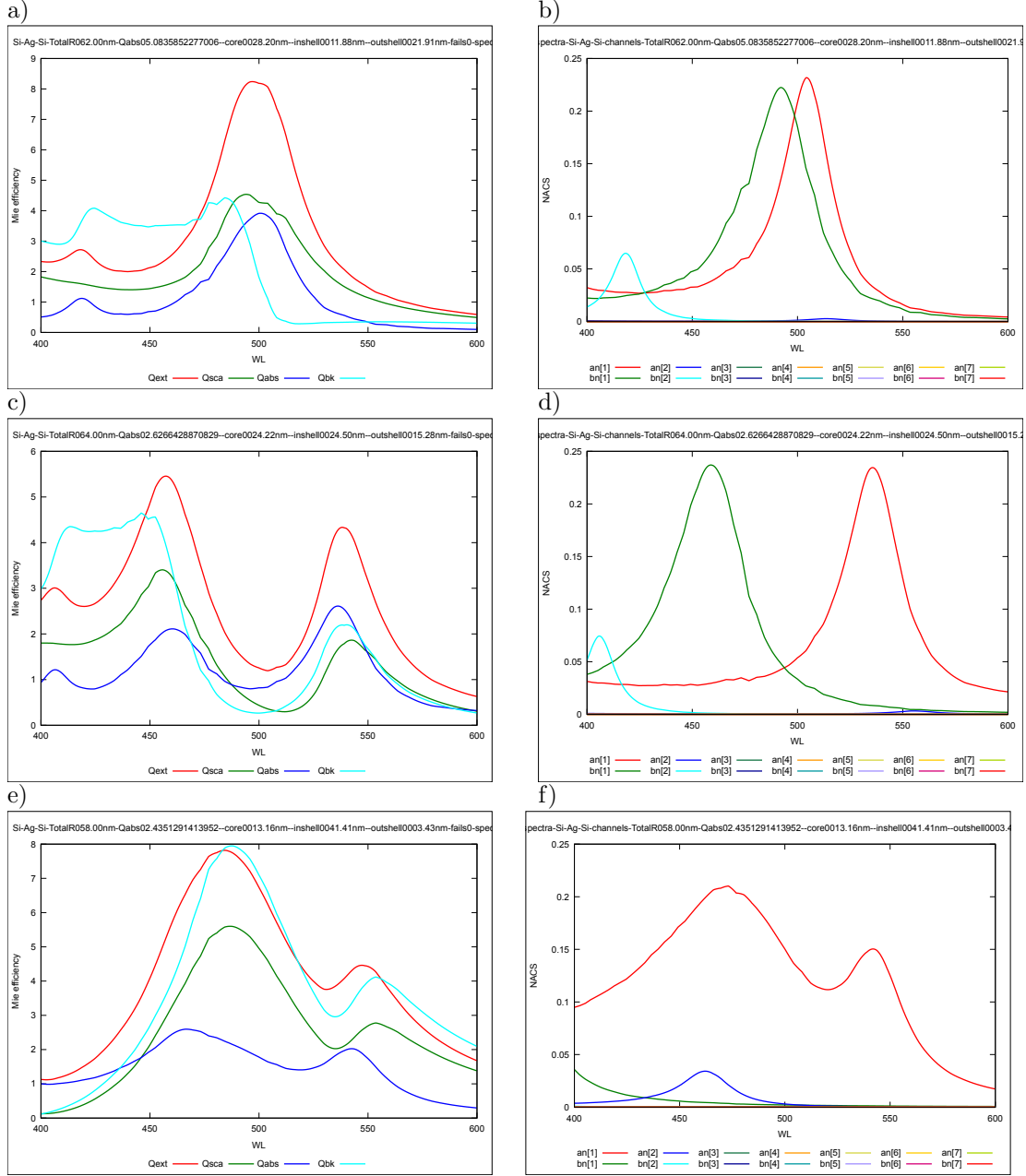


Figure 1: Single particle designs for separation between dipole resonances of 20 nm (a-b) and 80 nm (c-f). Left column with figures (a,c,e) contains spectra of Mie efficiency for extinction (Q_{ext}), scattering (Q_{sca}), absorption (Q_{abs} , blue curve) and backscattering (Q_{bk}). Right column (b,d,f) presents contribution of multipoles to the spectra (red and green stands for electric and magnetic dipoles, blue and cyan corresponds to quadrupole electric and magnetic modes, respectively). It is interesting to note that for 80 nm separation optimizer was able to find a design with two resonances of a dipole mode (f). We suppose that the latter most likely has the same nature as the response presented in Fig. 3(c,d) of the manuscript: there are several resonances of an electric dipole response binded to different layers of the particle.

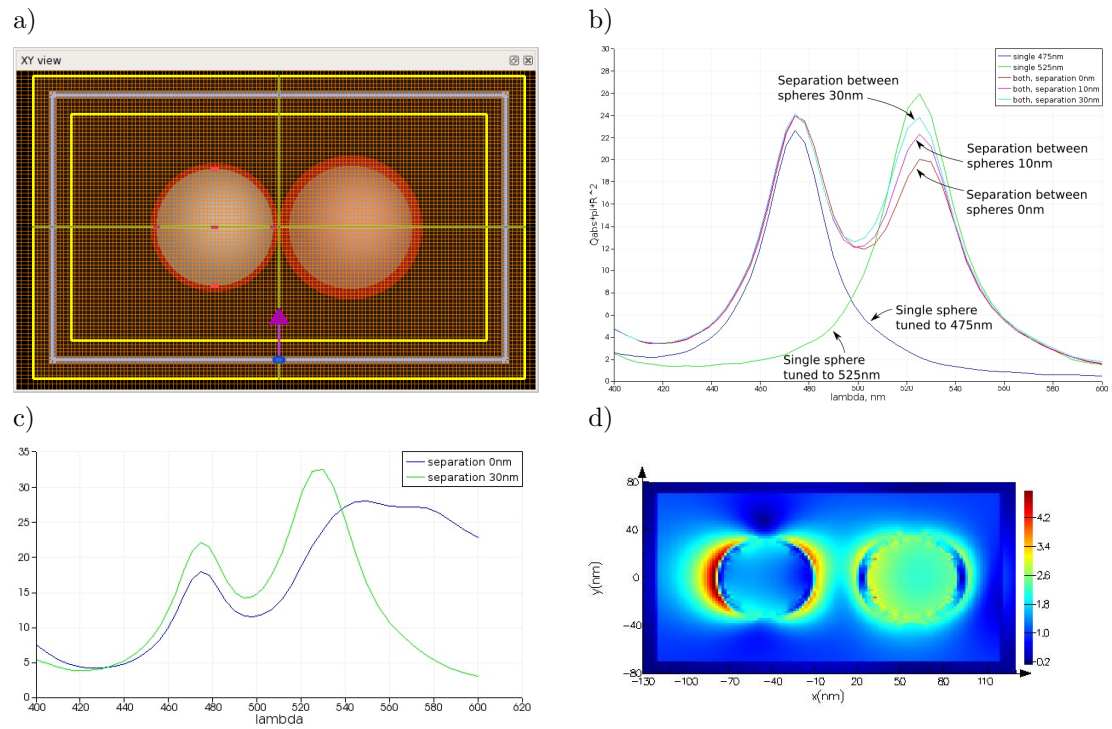


Figure 2: (a) Fullwave simulation prepared in Lumerical FDTD. (b) Absorption spectra of stand-alone spheres and in dimer configuration with separation between spheres of 0, 10, and 30 nm, particles are in H-k plane (c) Separation 0 and 30 nm for particles in E-k plane (d) Field distribution at 500nm of incident wavelength for particles in E-k plane with separation of 30 nm.

2. To achieve super-absorption behavior, it is interesting to know what will happen when the multilayered nanoparticles are closely packed as the plasmonic crystal. As reported in the previous papers [ACS Applied Materials & Interfaces, 7, 49624968 (2015); Materials Letters 158, 262265 (2015); Applied Physics Letters, 104, 081116 (2014); Nanotechnology, 24, 155203 (2013)], the packed plasmonic crystals have been demonstrated to show broadband light coupling and confinement. Thereby, it would be interesting to show improved broadband light absorption based on the plasmonic crystal of this proposed multilayered nanoparticles.

We totally agree with the reviewer’s statement. It seems that our manuscript was not clear enough at this point. ... However, as we compare final optimization results we conclude that To make it clearer we rewrite this part of the manuscript as follows:

“Some text”

Reviewer #2 comments

1. As we can find from the manuscript, the highest absorption efficiency is achieved for a Si/Ag core-shell structure, which is not located in the super absorbing regime. Moreover, the authors also claimed that from practical aspect, the core-shell structure (not in the super absorbing regime) could be easier and cheaper to fabricate than three layered structure (in the super absorbing regime). Therefore, the authors should clearly clarify what are the advantages or significances of the super absorption nanoparticles?
2. In Fig. 2, we noticed a discontinuity at 80 nm. The authors explain it as the design supporting electric dipole and magnetic quadrupole has larger ACS. However, this explanation is not clearly to me since it is lacking physics behind this phenomenon. The authors should clarify why the magnetic quadrupole only plays a significant role in this small wavelength range.
3. In Fig. 3 (c), the authors observed a flat top of electric dipole resonance. They attributed this flat resonance to the excited several electric dipole resonances with close resonance frequencies. Nevertheless, as we can see in Fig. 3 (d), even without considering the resonances located in outer and inner shell, the resonance inside the core is much broader than the other two cases. The authors should explain this broadened resonance clearly.
4. Some sentences are not clear to me. For instance, there is a strong conterplay between the increased absorption for larger particles vs size for smaller particles. In summary, I do not think the manuscript is acceptable at its current stage.

Sincerely Yours,
On behalf of the authors,
Konstantin Ladutenko