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 stanalone particle and in interaction with other particles. The first case, as it was correctly mentioned with the reviewer, has some limiations. We can design a band, which will be broader of a typical band for a given multipole, however, width is not extremly large. To illustrate this we have run an optimization with a goal to provide a prediefined separation between multipole resonanses (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 desings with electric and magnetic dipole. If the separation is large the absorption band splits into two. For large separation is is also possible to obtain two resonanses 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 involeve relatively large particles. This leads to appereance of resonances (and absorbtion) out of the designed band which are hard to control or suppress in disscussed triple-layer structure.

To answer the second part of the comment, related to possiblity of usage of the pariticle array with dispered sizes, we designed two particles with best absorption effeciency at 475 nm and 525 nm (outer radius 34 and 38 nm) and simulatied 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 fist simulated absorption of standalone spheres, the obtained postions and amplitudes of resonances corresponde well with Mie caclulations. 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 exploting the feature of field being highly localized inside the the sphere. For sure arranging spheres in other plane, increasing the number of spheres can change the amount of coupling between the spheres. As an example we tested arragement of sphers in E-k plane Fig. 2(c); for zero separation the intraction between spheres is strong, however, due to near-field nature of this coupling it rapidly decays with the separation width Fig. 2(d), for separation of 30 nm responses of individual particle dominates in overall spectra for both planes of polarization. This way we show that in principle it is possible to construct an absoption band of an array of dispersed particles simply optimizing the properties of every single sphere carefully tuning the distance between particles (which still is much smaller than the wavelength).

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 strong localization of electric dipole field (espatially in H-k plane)

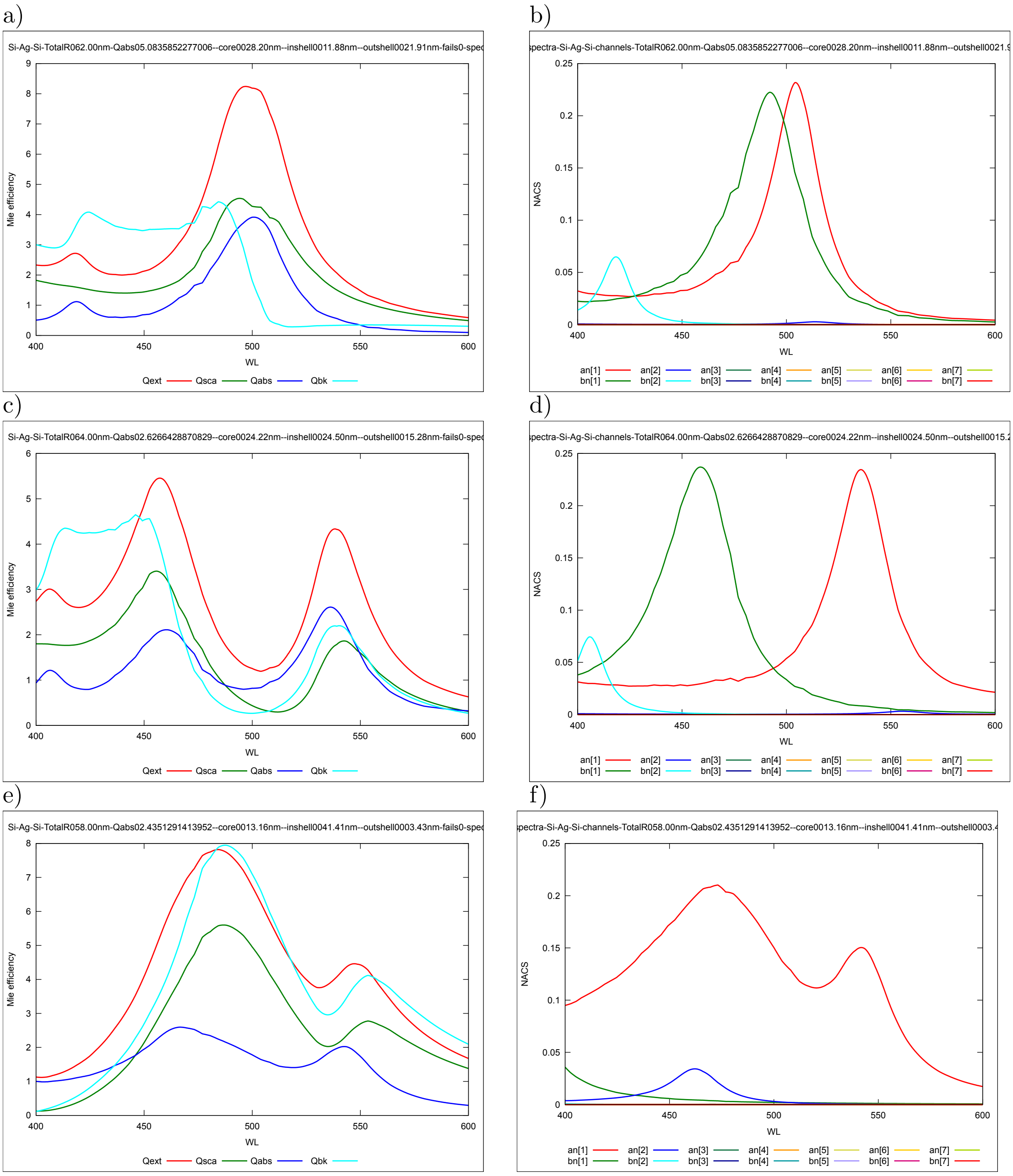

Figure 1: Single particle desings for separation between dipole resonances of 20 nm (a-b) and 80 nm (c-f). Left column with figures (a,c,e) contains spectra of Mie efficiency for extinction (Qext), scattering (Qsca), absorption (Qabs, blue curve) and backscattering (Qbk). Right column (b,d,f) presents contibution 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 desing 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.

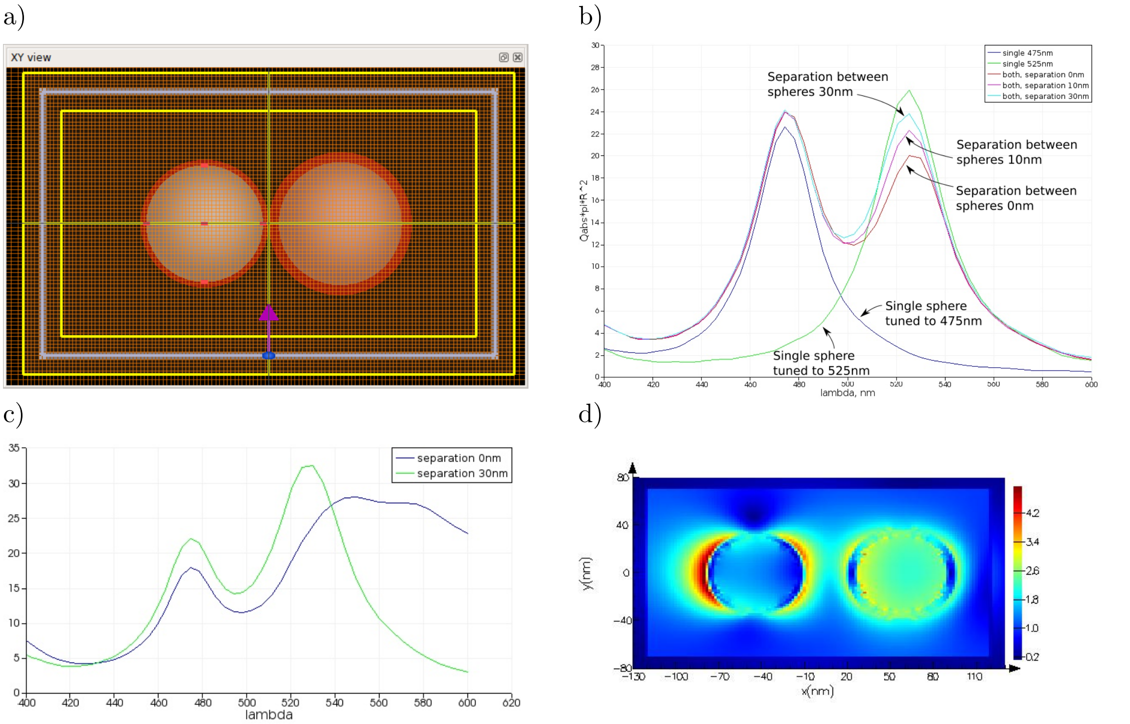

Figure 2: (a) Fullwave simulation prepared in Lumerical FDTD. (b) Absorption spectra of stand-alone spheres and in dimer configuration with separation between spheres of 0, 10, and 30 nm, particles are in H-k plane (c) Separation 0 and 30 nm for particles in E-k plane (d) Field distribution at 500nm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

it is also possible to desing dispersed size arrays particles by defining their individual properties for composite absorption spectra; we obtained rather small coupling between particles during additional simulation using FDTD method."

> 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.

In order to reply this comment first of all we checked all the citations provided with the reviewer.

ACS Applied Materials & Interfaces, 7, 49624968 (2015);

Materials Letters 158, 262265 (2015);

Applied Physics Letters, 104, 081116 (2014);

Nanotechnology, 24, 155203 (2013) titled "Near-unity transparency of a continuous metal film via cooperative effects of double plasmonic arrays" describes light interaction with a gold film, which has an array of gold spheres positioned on top, bottom, or both sides of the film. Provided simulations show a strong field enhancement between the spheres. Absorption related part of the paper references [18-21], where [20] is Le F, Brandl D W, Urzhumov Y A, Wang H, Kundu J, Halas N J, Aizpurua J and Nordlander P 2008 ACS Nano 2 707 titled "Metallic Nanoparticle Arrays: A Common Substrate for Both Surface-Enhanced Raman Scattering and Surface-Enhanced Infrared Absorption". It this paper absorption spectra in Fig. 5(b) calculated with electrostatic plasmon hybridization method showes, as it was stated with a reviewer, a significat increase of the band when packing particles to an array. This broadering originates from the appearance of collective mode (or, in other words, the hybridization of plasmon responce of several particles) with additional impact from retardation effects. It can be of great intereset (including applied examples) to apply the same approch from our manuscript to this system. It should be possible to optimize simultniously geometry of individual particles and their positioning in array to degenerate several absorption resonances of such collective modes in order to achieve superabsorption. However, we do not have any working code for plasmon hybridization method to check it with our optimizer. Using brute force approach with FDTD simulation looks to be rather computationaly expensive without any prior analytical estimations.

For a fast check of this idea we simulated the 3x3 rectangular array of spheres (to provide a good coupling both in E and H direcitons) with the separation of 4 nm (so it is only 2 mesh cells to resolve the separation in the used coarse grid, selected to get a resonable timing of a 3D full-wave simulation) Fig 3(a). Several collective modes can be recognized from field distribution while changing the incident wavelength Fig 3(c-e). However, for used materials, nanoparicle and array designs, and spectral range we observed a blue shift of absoption responce (it looks to be dominated with silver plasmon resonance) Fig 3(b), which is opposite to results of Le et al. in Fig. 5(b) of their paper.

This way it looks that using particle array adds a whole banch of new physical features which deserve a separate investigation. No changes to the manuscript were done.

" 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?

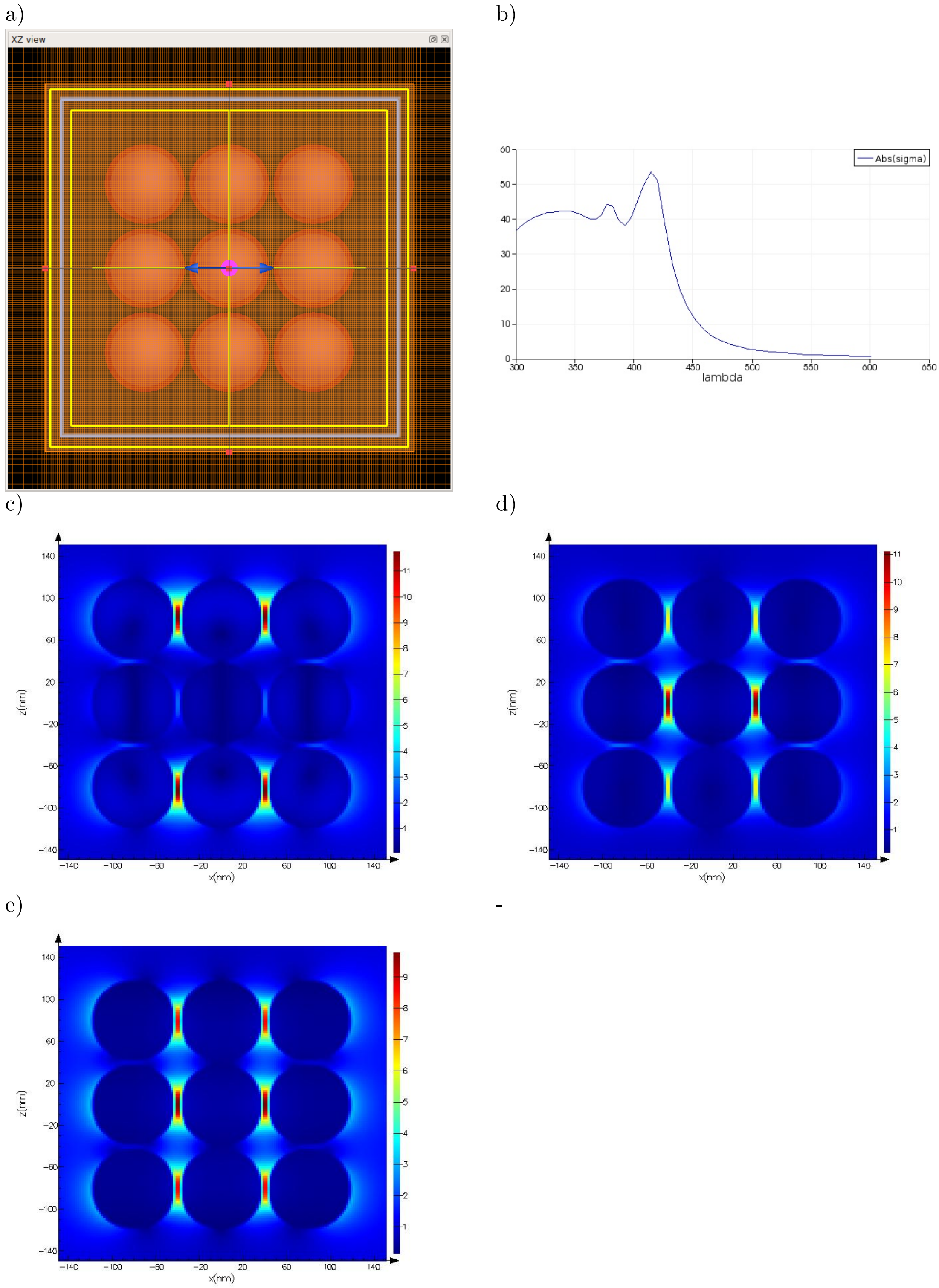

Figure 3: FDTD simulation of 3x3 grid of spheres optimized for absorption at 525 nm(a) Fullwave simulation prepared in Lumerical FDTD. (b) Absorption spectra of stand-alone spheres and in dimer configuration with separation between spheres of 0, 10, and 30 nm, particles are in H-k plane (c) Separation 0 and 30 nm for particles in E-k plane (d) Field distribution at 500nm of incedent wavelength for particles in E-k plane with separation of 30 nm.

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