

## Response Letter

Dear Editor,

Many thanks for sending us referees reports on our manuscript entitled Superabsorption of light by nanoparticles by K. Ladutenko et al. (editorial code NR-COM-08-2015-005468). We are pleased with overall very positive tone of these reports, as well as with referees constructive comments.

We have addressed all the comments of the referees, and we present our response and the summary of the changes made to the manuscript below.

#### 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.

We thank the referee for suggestion. First, we would like to note that the term "broadband" is quite loose, when not defined explicitly. For example, if we compare our absorption band with the whole C-band of the optical fibres, which is used to carry Terabits of data, and which is just 35 nm wide, then our absorbers are extremely broadband, since they have the bandwidth approaching 100 nm. Compared to the whole visible spectrum, however, indeed our absorption band is not very broad. So, to address the referee comment, we decided to study the possible ways to enhance the width of the absorption band.

We can think of a two possible approaches to achieve the broadband performance. First approach utilizes the mix of different particles, while the second approach uses several resonances within one particle. The later approach is nontrivial and requires large additional study, that would deserve a separate publication. The first approach, however, mentioned by the Referee, is more straightforward, and we can use a mixture of different particles, so that various sized particles absorb different wavelengths. To illustrate this we have run an optimization procedure to find two designs: one absorbs at 475 nm, while another is centered at 525 nm (outer radii 34 and 38 nm, respectively). We then combine such particles to form a dimer, and calculate its properties using Lumerical FDTD Solutions. Sketch of the simulated system and final results are shown in Fig. 1. As a reference, we also perform FDTD simulations of standalone spheres, and the obtained positions and amplitudes of resonances match well results of the Mie calculations.

When we mix the different sized spheres, there will be an effect of their mutual interaction. To take this into account, we simulate a dimer configurations with 0 nm, 10 nm and 30 nm separation between spheres. The resulting spectra have a strong contribution from standalone resonances, while coupling effects seems to be relatively minor. This can be explained from modal field distribution in Fig. 4(c) of the manuscript. Positioning spheres in H-k plane we are exploiting the features of fields being highly localized inside the spheres. Arranging spheres in other planes with respect to incident polarization, or increasing the number of interacting spheres can change the effect of coupling. As an example, we tested arrangement of spheres in E-k plane Fig. 1(c); for zero separation the interaction between spheres is strong, however, due to near-field nature of this coupling, it rapidly decays with the separation width Fig. 1(d), for separation of 30 nm responses of individual particles dominate in overall spectra for both planes of polarization. This way we show that in principle it is possible to construct an absorption band of an array of dispersed particles simply optimizing the properties of individual particles.

In order to reflect these findings in the manuscript, we have rewritten the text 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 broad spectra or spectrally-selective absorbers with almost arbitrary prescribed properties. One can also achieve broadband performance by mixing particles whose absorption is optimised for different wavelengths. We have verified that due to strong localization of electric dipole field it is possible to desing dimers whose absorbing properties are dominated by the properties of individual particles, and not by their mutual interaction. As a result, by appropriately spacing the resonances of individual particles, their mixture will exhibit a combined broadband response. Providing such an additional control, this approach complements the case of self-organized particles of various size and shape, which was experimentally proved to have a wide absorption band [ACS-AMI-Liu-2015]. Further control of the absorption spectrum can be achieved by arranging our spherical structures in a periodic and non-periodic arrays."

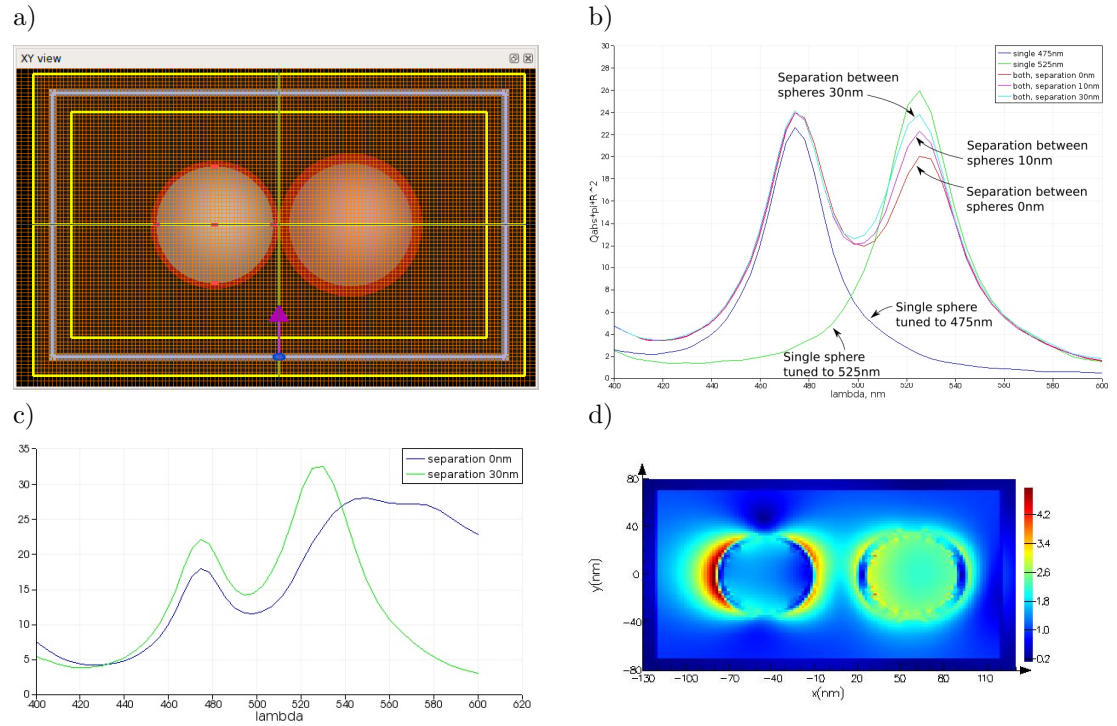


Figure 1: (a) Sketch of the simulated dimer in full wave simulations by Lumerical FDTD. (b) Absorption spectra of stand-alone spheres and in dimer configuration with separation between spheres of 0, 10, and 30 nm, particles are in H-k plane; (c) Separation 0 and 30 nm for particles in E-k plane; (d) Field distribution for the incident wavelength of 500 nm 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 would like to thank the reviewer for the references. We have included them into our manuscript with relevant discussions, with one exception. Reference Applied Physics Letters, 104, 081116 (2014) with the title " $\lambda^3/20000$  plasmonic nanocavities with multispectral ultra-narrowband absorption for high-quality sensing" is not related to broadband light coupling and confinement" and was probably listed by an accident.

We agree with the referee that closely packed particles can lead to the increase of the bandwidth. This broadening originates from the appearance of a collective mode (or, in other words, the hybridization of plasmon responses of several particles) with additional impact from retardation effects. While for some resonances, as we have discussed in the response to the previous comment, the interaction between our nanoparticles can be weak, it should still be possible to optimize simultaneously the geometry of individual particles and the array parameters to achieve enhanced absorption. As a proof of concept, we performed FDTD simulations of the  $3 \times 3$  square array of spheres (to provide a good coupling both in E and H directions) with the separation of 4 nm, see Fig 2(a). Several collective modes can be recognized from field distribution while changing the incident wavelength Fig 2(c-e). We see that for the used materials, nanoparticle and array designs, spectral range, and plane of polarization, we observed a broadened resonance, with the broadening mostly occurring in the blue shift direction Fig 2(b).

We believe that the detailed study of the effect of arranging the elements in array is a whole new project, and it lies outside the scope of our current manuscript. We have mentioned, however, in the revised manuscript that further control of the absorption spectrum can be achieved by arranging our spherical structures in a periodic and non-periodic arrays.

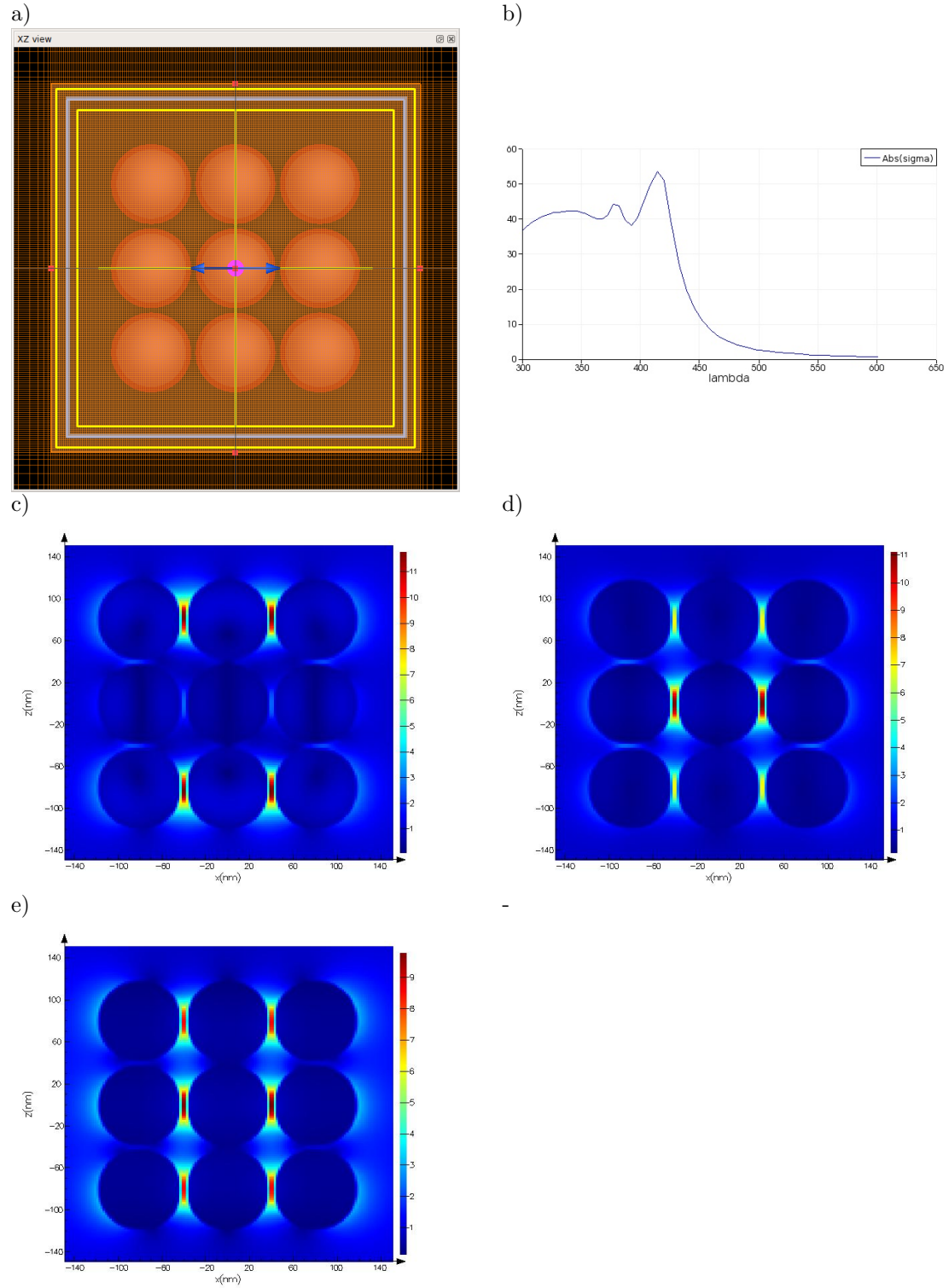


Figure 2: FDTD simulation of a 3x3 array of spheres, which are individually optimized for absorption at 525 nm (a) Fullwave simulation performed in Lumerical FDTD. (b) Cross-section absorption spectra (c,d,e) Field distribution for the wavelengths of 408, 452, and 600 nm.

## 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?

The super-absorption has a physical meaning of a regime, when we create a structure, which has several highly absorbing resonances overlapping in frequency, so that total absorption is larger than what can be created in the structure. The main conclusions of the paper are that a) We can create structures that shows superabsorption b) For small particles (below 60nm), the efficiency of absorption is higher for single mode regime. c) For larger particles the superabsorption regime allows to achieve best efficiency of absorption.

To make this more clear in the manuscript, we have made the following changes to the manuscript:

In the end of the first paragraph of page 3, right column, after "..., since it should be easier and cheaper to fabricate." We add: "At the same time the best absorption efficiency for larger particles (with  $R > 60$  nm for provided materials) can be achieved in superabsorption regime. This may be important when fabrication of smaller multi-layer particles is not available."

2. In Fig. 2, we noticed a discontinuity at  $\sim 80$  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.

The short qualitative answer to this is that it is not possible to excite the magnetic quadrupole resonance to the particle of a smaller size, and, it is hard to keep it efficient for larger sizes, while also maintaining the large contribution of the electric dipole.

To present this idea in more details we run an optimization with changed fitness function - we maximize the absorption efficiency of only the electric dipole, with results presented in Fig. 3. With the increase of the particle radius  $R$  the  $\tilde{a}_1$  rapidly reaches its theoretical maximum. Optimizer was quite successful in maintain this for all larger values of  $R$ . When we increase the  $R$ , other multipole resonances can appear. Such an appearance of  $\tilde{b}_2$  formes the discussed maxima at  $\sim 80$  nm, and it is clearly observed in Fig. 3(b).

Another way to qualitatively explain the effect is the following. We have a multiple resonance structure, and by changing parameters we can overlap several different resonances. Depending on the total particle size, different combination of resonances can give maximum absorption, therefore it is not surprizing that in a certain parameter range, the combination of electric dipole and magnetic quadrupole can give us the best possible absorption.

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.

We thank the referee for pointing this out, we must have not explained this point clearly. The most interesting feature of this resonance that we wanted to bring attention to is that the resonance has an unusual flat top part. The main shape and width of the resonance is explained by one dominating dipole mode, as it is seen in the mode decomposition in Fig. 3(d). Two additional electric dipole resonances are much weaker, but their contribution "flattens" the top of the combined absorption resonance. We have clarified this in the modified manuscript changing the caption of Fig. (3), after "Panel (d) shows the superposition of the squared absolute values of the Mie coefficients for electric dipole contribution inside each layer" we added "of design from panel (c), which is needed to explain an almost flat top of the electric dipole resonance."

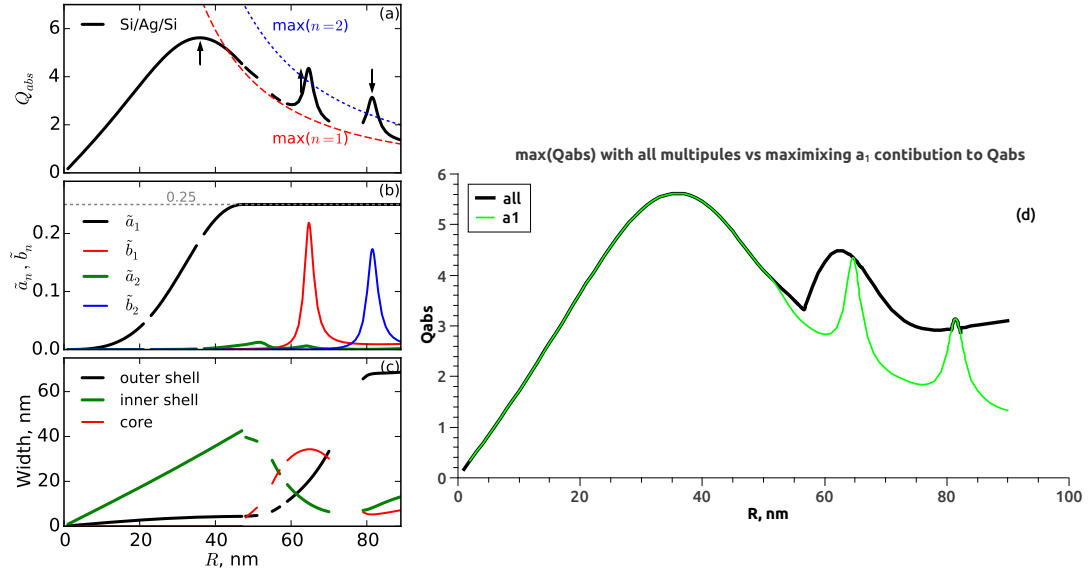


Figure 3: (a-c) Same as Fig.2 of the manuscript with different fitness function: the optimization aims to maximize the electric dipole absorption. (Missing points did not pass the stability test.) (d)  $Q_{abs}$  for maximized  $\tilde{a}_1$  from the first subplot of present figure without the additional stability test (green line) compared with  $Q_{abs}$  optimization regarding all multipoles (black line, same as in Fig 2(a) of the manuscript).

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.

We have worked on improving the readability of the manuscript. Particularly, we changed the instance metioned with the reviewer, full sentence in the updated manuscript is now the following: “Here we show that the increased peak absorption rate of high order multipoles available in larger particles does not compensate the increased volume with low absorption rate compared to volume of smaller particles, and quite remarkably we find that the most *efficient* absorption can be achieved in a single channel limit for small particles.”

Sincerely Yours,  
On behalf of the authors,  
Konstantin Ladutenko