Response Letter from the Corresponding Author

Dear Dr. Eimy Zhao,

Thank you very much for accepting our revised manuscript and your supervision of the reviewing process of this manuscript with Manuscript ID: nanomaterials-1045035. We also appreciate deeply the reviewers' conscientiousness, and the broad knowledge on the relevant research fields, since he/she has given a number of beneficial suggestions. According to the reviewers' instructions, we have made the following responses and revisions:

Reviewer's comments:

Reviewer #1:

In the current work, the authors fabricate and study four types of split-disk metamaterial absorbers. By changing the geometry, the nanostructures exhibit different absorption resonances in the range from 1.5 to 5.0 μm wavelengths. The manuscript is clearly written; the conclusions are in general supported by the data shown, although additional measurements are required. The current work contributes to the field that is interesting for many readers of the journal. However, several issues listed below should be amended prior to publication.

1. The key major concern that I have is related to the novelty of the current work. Whereas the authors present a new geometry of plasmonic nanostructures that to my knowledge has not been shown yet, I have not found any argument why is it better than many other nanostructures that have been studied before. Although the authors site many of the relevant papers, there are many more recent articles where similar nanostructures where studied. In my view, the authors should clearly answer the question why do they think this geometry is better/easier to produce, etc.

Response:

Thanks for reviewer's kindly instruction. We proposed tunable SDM absorber is a new design. There has not reported such kind of nanostructures of SDM until now. The proposed SDM absorbers are suspended and connected to the MEMS actuators for changing the gaps between top SDMs and bottom Au layers. The resonances of absorption spectra can be tuned in the range of 300 nm and the absorption intensities become lower by increasing gap values. The electromagnetic fields could be effectively coupled into the split-disk nanostructures and the confined between top SDMs and bottom Au layers. The proposed MEMS-based SDM absorber could be realized by using MEMS technique. Fig. R1(a) shows the schematic drawings of proposed MEMS-based SDM array. The top SDM is suspended and supported by using a membrane composed of Si/Si_3N_4 bilayer. Owing to Si and Si_3N_4 bilayer with a very low absorption coefficient at the IR wavelength range, that can be ignored the influence of electromagnetic wave propagation of SDM. Fig. R1(b) shows the unit cell of SDM. The top SDM is suspended and supported by Si/Si_3N_4 bilayer using residual stress induced electrothermal actuators (ETAs). The thermal expansion coefficients of Si and Si_3N_4 layers are mismatched. As the structures are released, the Si layer is under tensile residual stress due to its larger thermal expansion coefficients while the Si_3N_4 layer is under compressive residual stress due to its smaller thermal expansion coefficients. Such initial stress between Si and Si_3N_4 layers makes the cantilever deformed out-of-plane and bending upward. By applying a DC bias voltage on these residual stress induced ETAs, the electrothermal attraction force is induced between the released cantilevers. Meanwhile, the out-of-plane bilayer cantilever will be bended downward to the substrate owing to such electrothermal attraction force. Here, the top SDM is connected to four ETAs, which are ETA_1, ETA_2, ETA_3, and ETA_4, respectively. Therefore, SDM can be tuned vertically by actuating the ETA_1 to ETA_4 to change *z* value between top SDM and bottom mirror layer. The 3D illustration view is shown in Fig. R1(c). The corresponding fabrication process of SDM is illustrated in Fig. R1(d). First, an Au thin-film with 100 nm in thickness is deposited on Si substrate by using sputtering process as bottom mirror layer shown in Fig. R1(d-i). Second, SiO_2 and Si/Si_3N_4 layers with 1 μm in thickness are deposited by using plasma enhanced chemical vapor deposition (PECVD) process on sample surface (Fig. R1(d-ii)). Third, SiO_2 and Si/Si_3N_4 layers are patterned using reactive ion etching (RIE) process to define the supporting frame and ETAs structures of SDM (Fig. R1(d-iii)). Forth, an Au thin-film with 100 nm in thickness is deposited and patterned on sample surface using lift-off process as top SDM (Fig. R1(d-iv)). Fifth, an Al thin-film is deposited and patterned using lift-off process on sample surface as electrode pads of ETAs (Fig. R1(d-v)). Finally, sample is released the supporting frame and ETAs structures using vapor HF (VHF) to perform the vertically tuning mechanism of SDM (Fig. R1(d-v)). When the driving voltage is applied on the Al electrodes pads of ETA_1 to ETA_4, SDM can be tuned to change *z* value vertically.

In the future, we will design and demonstrate the SDM device by using MEMS technique and following up this fabrication process. Such SDM device possess flexibility and applicability which could be used in widespread applications with active tunability, such as switch, filter, absorber, sensor, and so on.

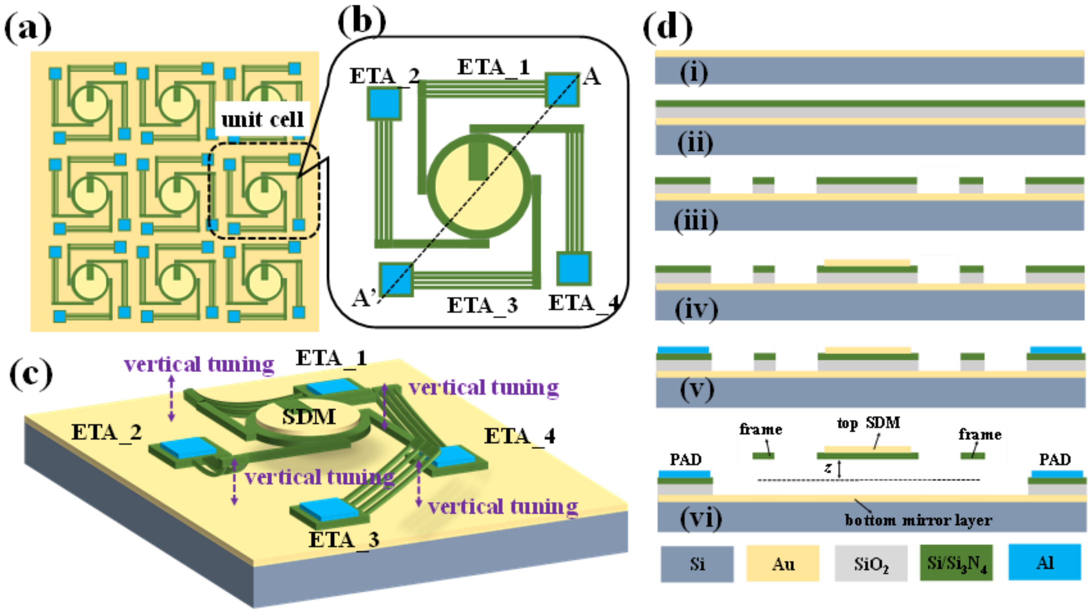

Fig. R1. Schematic drawings of (a) proposed tunable SDM array, (b) unit cell using residual stress induced ETAs, and (c) 3D illustration view of unit cell after released the structures. (d) Fabrication process of proposed tunable SDM using MEMS technique along AA' line in (b). (d-i) First, an Au thin-film with 100 nm in thickness is deposited on Si substrate as bottom mirror layer. (d-ii) Second, SiO_2 and Si/Si_3N_4 layers with 1 μm in thickness are deposited by using PECVD process sequentially on sample surface. (d-iii) Third, SiO_2 and Si/Si_3N_4 layers are patterned using RIE process to define the supporting frame and ETAs structures of SDM. (d-iv) Forth, an Au thin-film with 100 nm in thickness is deposited and patterned on sample surface using lift-off process as top SDM. (d-v) Fifth, an Al thin-film is deposited and patterned using lift-off process on sample surface as electrode pads of ETAs. (d-vi) Finally, sample is released the supporting frame and ETAs structures using VHF to perform the vertical tuning mechanism of SDM.

2. Second, I believe that a proper characterization of new types of metamaterials is hardly possible without their direct visualization using SEM imaging. The authors could show a few examples of images of the nanostructures studied.

Response:

Thanks for reviewer's kindly instruction. As the response of comment 1, we will design and demonstrate the SDM device by using MEMS technique and following up the proposed fabrication process. Such SDM device possess flexibility and applicability which could be used in widespread applications with active tunability, such as switch, filter, absorber, sensor, and so on.

3. Minor points:

RIU should be spelled out.

Figures 2-5: absorbance is a logarithm and hence, is a dimensionless value; therefore no a.u. is needed for the y-axes.

Response:

Thanks for reviewer's kindly instruction. We apologize that we didn't describe clearly. The absorption spectra used in this study are absorption intensity not logarithm. Thus, the unit of absorption spectra is used arbitrary unit (a.u.).

Reviewer #2:

The topic is appropriate for publication in the journal. The question is original but not well defined. First, it is not clear in the presentation that the paper deals with a numerical study. Then, the scientific background is not well presented: it is not clear if the proposed metamaterials have already been studied in the literature (theoretically or experimentally) or they are originally proposed by the authors. It is not clear the experimental feasibility of the theoretical design. In section 2 and 3 authors declare: “The gaps could be actively tuned by using electrostatic or electrothermal forces”. It is not clear the feasibility of the integration of such technologies with realization of the proposed metamaterial. Is there some reference with experimental results supporting this guess?

In section 3 the authors declare: “SDM-1 exhibits perfect absorption”. I think this observation deserve a specific comment: it seems an important aspect to be underlined and discussed better.

Always in section 3, the authors say that the refractive index range of investigation is between 1 and 1.4. It is a wide range not really specific for gas sensing or biosensing. Comparison of sensitivity parameters with a different optical sensor working in this range for the specific application could be useful to understand the sensing performance of the proposed transducer.

Figures 7 and 8 attest that “The electromagnetic energies are confined within the top SDM nanostructures and bottom Au mirror layers”. This aspect is not geometrically favourable for gas or biosensing purposes owing to the experimental difficulty in immobilizing the sensing surface with the specific receptor. Have the authors taken in consideration this drawback?

Finally the conclusion that the proposed metamaterial “tuned spanned the perfect zone of “molecule fingerprint” should be scaled back as there is not enough experimental evidences of this statement.

Minor comments: English should be improved in lines: 125, 152-153, 204-205.

1. First, it is not clear in the presentation that the paper deals with a numerical study.

Response:

Thanks for reviewer's kindly instruction. We have revied the presentation in the last paragraph of session 2.

“To numerically calculate the optical properties of SDM devices, the Lumerical solution’s software finite difference time-domain (FDTD) solutions are employed in the full-field plane electromagnetic wave. The permittivity value of Au material at the IR wavelength range is calculated according to the Drude-Lorentz model [47,48]. The propagation direction of incident IR wave is set to be perpendicular to the x-y plane.

The periodic boundary conditions are applied in the x- and y-axis direction, while the perfectly matched layer (PML) boundary conditions are assumed in the z-axis direction. The reflection (R) and the transmission (T) of incident IR wave are calculated by spectra monitors set below and above the devices, respectively. The corresponding absorption (A) is defined as $A = 1 – T – R$. Since the transmission of incident electromagnetic light is blocked by the bottom Au mirror layer, the absorption can be simply obtained by $A = 1 – R$. It apparently means that high absorption is earned from low reflection."

2. The scientific background is not well presented: it is not clear if the proposed metamaterials have already been studied in the literature (theoretically or experimentally) or they are originally proposed by the authors.

   Response:

   Thanks for reviewer's kindly instruction. We proposed tunable SDM absorber is a new design. There has not reported such kind of nanostructures of SDM until now. The proposed SDM absorbers are suspended and connected to the MEMS actuators for changing the gaps between top SDMs and bottom Au layers.

3. It is not clear the experimental feasibility of the theoretical design. In section 2 and 3 authors declare: "The gaps could be actively tuned by using electrostatic or electrothermal forces". It is not clear the feasibility of the integration of such technologies with realization of the proposed metamaterial. Is there some reference with experimental results supporting this guess?

   Response:

   Thanks for reviewer's kindly instruction. Please refer to the response of comment 1 by reviewer #1. The proposed MEMS-based SDM absorber could be realized by using MEMS technique. Fig. R1 shows the schematic drawings of proposed MEMS-based SDM array and relative fabrication process.

4. In section 3 the authors declare: "SDM-1 exhibits perfect absorption". I think this observation deserve a specific comment: it seems an important aspect to be underlined and discussed better.

   Response:

   Thanks for reviewer's kindly instruction. We have revised in the revised manuscript.

5. Always in section 3, the authors say that the refractive index range of investigation is between 1 and 1.4. It is a wide range not really specific for gas sensing or biosensing. Comparison of sensitivity parameters with a different optical sensor working in this range for the specific application could be useful to understand the sensing performance of the proposed transducer.

   Response:

   Thanks for reviewer's kindly instruction. We consider the sensitive refraction index of proposed SDM absorber is in the range of 1.0 to 1.4 and the refraction index of most gas and bio-liquids are in this range. It is considering the reviewer's suggestion we have compared it with some literatures for optical metamaterial sensors working in this range.

6. Figures 7 and 8 attest that “The electromagnetic energies are confined within the top SDM nanostructures and bottom Au mirror layers”. This aspect is not geometrically favourable for gas or biosensing purposes owing to the experimental difficulty in immobilizing the sensing surface with the specific receptor. Have the authors taken in consideration this drawback?

Response:

Thanks for reviewer's kindly instruction. The proposed SDM absorbers are suspended and connected to the MEMS actuators for changing the gaps between top SDMs and bottom Au layers. The electromagnetic fields could be effectively coupled into the split-disk nanostructures and the confined between top SDMs and bottom Au layers. The proposed MEMS-based SDM absorber could be realized by using MEMS technique. The fabrication process could be referred to the response of comment 1 by reviewer #1. In brief, the top SDM is suspended and supported by Si/Si_3N_4 bilayer using residual stress induced electrothermal actuators (ETAs). These tunable nanostructures could be detected the specific receptor absorbed on the SDM surface or within the top SDM layer and bottom Au mirror layer for the use of absorbers in sensing application.

7. Finally the conclusion that the proposed metamaterial “tuned spanned the perfect zone of “molecule fingerprint” should be scaled back as there is not enough experimental evidences of this statement.

Response:

Thanks for reviewer's kindly instruction. We have scaled back this conclusion as “tuned within the “molecule fingerprint” wavelength range.” Because by tailoring the geometrical configurations and changing the gaps of air insulator layer, four SDMs exhibit different tunable absorption resonances spanned from 1.5 μm to 5.0 μm wavelength range, which is in the “molecule fingerprint” wavelength range from 2.5 μm to 25 μm.

8. Minor comments: English should be improved in lines: 125, 152-153, 204-205.

Response:

Thanks for reviewer's kindly instruction. We have revised as follows reviewer’s suggestion.