Dear Editor,

We would like to thank the referees for their time and effort to review our manuscript. We are happy to learn that both referees gave positive evaluations on the overall quality of our work and its suitability to Nanoscale.

We have revised our manuscript according to all referees' comments, and we believe that by doing so, the manuscript is much improved in terms of clarify and accuracy. We thank the referees for that. Below, we provide our point-to-point response to all comments and describe the revisions.

Thank you very much for your consideration!

Dawei He, on behalf of all co-authors

Referee: 1

Please find my technical comments and questions:

1. In Fig.2d, the surfaces of both the sample and the substrate are tilted, which may lead to misreading the actual thickness of the sample. Please correct this.

Authors:

We thank the referee for the comment. The tilt is an artifact due to instrument calibration. We have reprocessed the calibration and modified the image, as shown in the new Fig. (page 3).

Referee: 1

2. Did the authors try to investigate the case of different layers of bulk $PdSe_2$, even monolayer or few layers?

Authors:

We thank the referee for the valuable suggestion. We agree that carrier diffusion in thin or monolayer $PdSe_2$ would be interesting to study. However, $PdSe_2$ is rather challenging to exfoliate because flakes break easily. Few layers we could obtain have rather small lateral size. Since our spatial scans require a relatively large sample area, it is not feasible to perform such measurements. We thank the referee for the valuable suggestion.

Referee: 1

3. Since the authors have calculated the diffusion length, is this term widely used in optoelectronic devices to demonstrate their high-performance characteristics? If so, please give some comments.

Authors:

We thank the referee for the comment. In the design and manufacturing of semiconductor optoelectronic devices, the carrier diffusion length is a crucial parameter that directly impacts the device's efficiency and sensitivity. When the diffusion length is too small, it limits the distance that electrons or holes can move in the semiconductor, thereby affecting the response speed and efficiency of the device.

In the revised manuscript, we have added the following discussion (page 2, left column):

The diffusion length of the photocarriers has a significant impact on the performance of semiconductor optoelectronic devices, such as photodiodes and solar cells. In these devices, the diffusion length of the photocarriers determines the distance the photocarriers can reach during their lifetime, and thus impacts the detectivity and efficiency of these devices.

Referee: 1

4. On the third page, there is an error in reference 32. Please correct this.

Authors:

We thank the referee for the comment, and we apologize for this mistake, which we have corrected. (page 3).

Referee: 1

5. The formatting in the text requires careful review, as there are inconsistencies in the use of spaces after commas. Some instances include commas followed by a space, while others lack a space. It is crucial to maintain consistency in formatting as it enhances the readability of the paper and ensures that the message is conveyed effectively. Therefore, it is recommended to thoroughly proofread the text and make necessary revisions to ensure that the format is consistent throughout the paper.

Authors:

We thank the referee for the comment, and we apologize for the mistake. We have carefully proofread the format of the entire text and made corresponding modifications to any mistakes, including spaces, punctuation, and references.

Referee: 2

Comments to the Author

The manuscript "Ultrafast Transient Absorption Measurements of Photocarrier Dynamics in $PdSe_2$" mainly discussed the time-resolved carrier relaxation and photo-induced carrier diffusion. The authors demonstrated that $PdSe_2$ can act as a more suitable photo-sensitive 2D material operating at wide wavelength range. Overall, I think this manuscript need some modification before publication on Nanoscale. Here are some comments:

Authors:

We would like to thank the referee for the time and effort to review our manuscript. We appreciate the positive comments on the overall quality of our study. Below, we will response to all comments and describe revisions to address these comments.

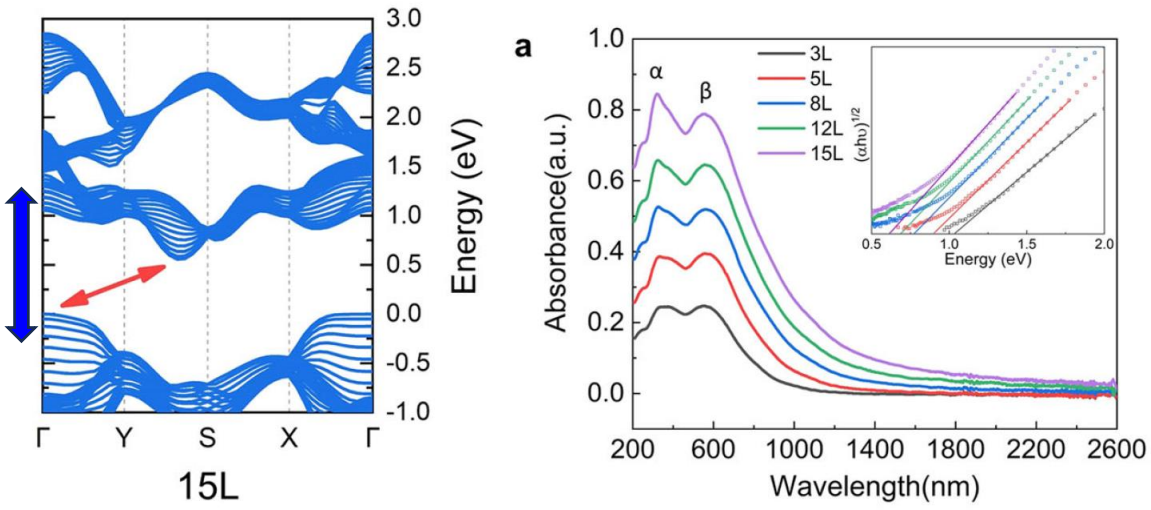
Referee: 2

1. Why the probe light is chosen as 800 nm? In another saying, what exact process is probed when you use a probe laser in 800 nm?

2. How large is the band gap at such a thickness of measured sample?

Authors:

We thank the referee for both comments. We would like to respond to them together since they are closely related. Bulk $PdSe_2$ has an indirect band gap of about 0.6 eV and a direct band gap of about 1 eV. We copy two figures from Wei et al., npj 2D Materials and Applications 6, 8 (2022) as an example: Left figure below shows the calculated band structure of 15-layer $PdSe_2$, which can be viewed as bulk in terms of electronic states. It has an indirect bandgap (red double arrow). The direct band gap at the Γ point is about 1 eV. The calculated band structure is consistent with the absorption spectrum, shown in the right figure, with a strong absorption band below about 1200 nm. Our 800 nm probe (1.55 eV) thus couples the energy states in the Γ valley.



Although our 800-nm probe direct senses carrier population in Γ valley, it effectively monitors the entire carrier population for two reasons. First, most holes populate in the Γ valley. Second, electrons form a thermal distribution in the conduction band, and thus a fraction of the electrons populate the Γ valley. Hence, we can treat the differential reflectance of the 800-nm probe as being proportional to the carrier density.

We have added the following discussion on this issue in the revised manuscript (page 3, left column):

> A combined experimental and computational study shew that bulk-like $PdSe_2$ has an indirect band gap of about 0.6 eV and a direct band gap of about 1.0 eV at its Γ point [1]. Hence, the 400-nm (3.10-eV) pump excites photocarriers with large excess energies. The 800-nm (1.55-eV) probe coupled to the direct transitions in the high energy states in the Γ valley, and thus monitors carrier density in these states. However, since the carrier system

form a thermal distribution, these carriers represents the overall carrier population and its dynamics.

Referee: 2

3. Are the measured data shows a superiority of $PdSe_2$ than well-studied TMDC materials?

Authors:

We thank the referee for the comment. We have revised this part of the discussion to clarify that our measured data for certain indicators shows superiority compared with well-studied TMDC materials. For example, the lifetime of photocarriers is longer, and the diffusion coefficient and diffusion length are larger.

In the revised manuscript, we have added the following discussion:

> The long time constant of approximately 210 ps represents the lifetime of the photocarriers. Notably, this carrier recombination lifetime is longer than that of commonly studied two-dimensional bulk TMDCs, such as WS_2 (110 ps)[2], $MoTe_2$ (80 ps) [3], and MoS_2 (180 ps) [4]. (page 3, right column)

> It is worth noting that the diffusion coefficient and diffusion length measured in our experiment are larger than the previously reported values of typical TMDC crystals. For instance, the diffusion coefficient and length bulk WS_2 is 3.5 cm^2s^{-1} and 196 nm, respectively [2], and 4.2 cm^2s^{-1} and 270 nm, respectively, in bulk MoS_2 [4]. (page 4, right column)

[1] M. Wei, J. Lian, Y. Zhang, C. Wang, Y. Wang and Z. Xu, npj 2D Materials and Applications, 2022, 6, 1.

[2] J. He, D. He, Y. Wang, Q. Cui, F. Ceballos and H. Zhao, Nanoscale, 2015, 7, 9526–9531.

[3] S. Pan, W. Kong, J. Liu, X. Ge, P. Zereshki, S. Hao, D. He,Y. Wang and H. Zhao, ACS Applied Nano Materials, 2018, 2,459–464.

[4] N. Kumar, J. He, D. He, Y. Wang and H. Zhao, Journal of Applied Physics, 2013, 113, 133702.