Dear Dr Wang,  
  
Re: "Energy Transfer in a Type-I van der Waals Heterostructure of WSe2/PtSe2"  
  
Manuscript reference: 2DM-107572  
  
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REVIEWER REPORT(S):  
  
Referee: 1  
  
COMMENTS TO THE AUTHOR(S)  
The authors have performed transient reflectance contrast (TRC) measurements on a singe WSe2/PtSe2/(sapphire substrate) sample, which suggest that the transition-metal dichalcogenide heterostructure WSe2/PtSe2 is a type-I semiconductor, with conduction- and valence-band edges confined to the PtSe2 layer.  
  
The authors base their conclusions on three main results:  
  
\* The reduced photoluminescence signal from the WSe2 A exciton in the heterostructure, as compared with a WSe2 monolayer (presumably, on the same sapphire substrate, although the paper never states that explicitly)  
  
\* A reduced TRC signal in the heterostructure, as compared with the WSe2 monolayer, when pumping e-h pairs at an energy much larger than the WSe2 band gap and probing at the WSe2 A exciton energy  
  
\* A vanishing TRC signal in the heterostructure and both monolayers when pumping e-h pairs at an energy lower than the WSe2 band gap and probing at the WSe2 A exciton energy  
  
While I do have some comments and questions regarding the authors' interpretation of their measurements, I believe that their conclusions are correct, and that their findings are novel enough to be published in 2D Materials. For that reason, I will recommend publication of this paper, once the following comments & questions are properly addressed by the authors:  
  
1) As mentioned above, the authors mention that a control WSe2 monolayer was transferred "onto a bare substrate". Since the WSe2 A exciton PL signals in WSe2 and WSe2/PtSe2 have similar broadenings and peak energies, with only a slight red shift in the latter (understandable, from the additional screening introduced by the PtSe2 layer), it seems logical that the "bare substrate" is the same sapphire substrate. However, I recommend that this be stated explicitly, to avoid any ambiguities.

Thanks for mentioning this error. It has already been corrected in manuscript.

2) The TRC signal from the heterostructure reported in Fig. 4b is quite similar to that from the WSe2 monolayer, with the exception of the signal amplitudes. The short lifetimes of 0.50 and 0.56 ps, respectively, are quite comparable. Yet, the authors assign different meanings to the two: in the former case, the authors suggest that the 0.50 ps lifetime has to do with electron transfer into the PtSe2 layer, whereas in the latter case, the 0.56 ps lifetime is assigned to exciton formation. Could the authors comment on what motivated these different assignments? Could it be that exciton formation also explains the 0.50 ps lifetime in the heterostructure?

As you mentioned, the signal amplitudes are different, so well as the proportional of the fast decay process (for monolayer WSe2, it is 74% and for heterostructure, it is 92% ). It is quite obvious that the different processes occur from different sample. According to our previous work, electron transfer is highly efficient which contributes 92% change of the signal amplitude. Exciton formation in the heterostructure also occurs at similar time range. However, it is not the main process in heterostructure, and it is not easy to distinguish them from each other. Hence, we would like to focus on charge transfer process here in heterostructure sample.

3) The authors also exclude the possibility of type-II band alignment citing that, in that case, long-lived interlayer excitons would form in the heterostructure, whereas no long-lifetime (~100 ps) appears in their TRC measurements. However, they are probing at the WSe2 A exciton energy, whereas an interlayer exciton would appear at a much lower energy. Could the authors comment on this?

First, since the bandgap edge of WSe2 and PtSe2 are quite close (form DFT thoery), hence interlayer exciton should appear similar energy. Second, it is quite difficult to probe the interlayer exciton directly. Our techniques is based on transient absorption theory, and absorption is not sensitive to wavelength at this range. Finally, we did different pump/probe combination to confirm the type I band alignment, which suggest that interlayer exciton is of absence in heterostructures.

Referee: 2  
  
COMMENTS TO THE AUTHOR(S)  
The paper concerns the study of the interface between 2D materials WSe2 and PtSe2.  By experiments of PL and absorption spectroscopy this vdW heterostructure is characterized, especially the band alignment, with evidences in the direction  of being   type-I kind. Additionally,   it provides also the  description of exchange of charge and energy in the systems. I think that the paper is short and straight, with robust and important conclusions. I only have some points to be discussed.  
  
(i) In 2D heterostructure, the characterization of interfaces is sometimes not so simple than in 3D well established cases (typeI, type II, etc.)   Stronger interlayer interactions, leading to larger shifts in band alignments compared with natural cases , or even significant orbital overlap is found, which create systems with undefined band offset as ZrS2/SnS2 (PRB B 97, 165402 (2018) or MoS2/WS2 heterobilayers (Phys. Rev. B 101, 121404 (2020)). Is it possible to confirm based on these experiments that this simple theoretical interface model can fully describe this system?  
(ii) Is it possible define beyond the interface type, the absolute values of band offset our even speculate about its magnitude?  
(iii) Is it possible, to define the geometrical configuration of interfaces (AA, AB, etc) ?  
(iv) The interface properties should change depending on the kind of interface configuration?  
(v) There are previous DFT calculations concerning this interface?  
  
Referee: 3  
  
COMMENTS TO THE AUTHOR(S)  
The authors studied energy transfer of a van der Waals heterostructure of WSe2/PtSe2 by steady-state photoluminescence and time-resolved transient absorption spectroscopy. A main finding is that heterostructure forms a type-I band alignment with both the conduction band minimum and the valence band maximum located at the PtSe 2 layer. The work is interesting in the community of vdW materials. I think it is worth to be published in 2D materials, however there are few questions that the authors needed to be clarified.  
1. To exclude the type-II band alignment of the heterostructure, the authors pumped a light of 1.51eV and probe at 1.67eV. This is insufficient to exclude the type-II band alignment, because as long as the pumping light is less than 1.67eV, it is impossible to probe the resonance at 1.67eV. This is the case regardless of interlayer charge transfer. In fact, if the heterostructure is a type-II band alignment, the charge transfer would lead to the formation of interlayer exciton. Then, the authors should exclude the possibility to form intelayer excitons, so as to claim that the heterostructure is in a type-I band alignment?

2. A theoretical first-principles calculation would give some information on the band alignment of the heterostructures.

band alignment of the heterostructures  
3. Monolayer Wse2 has a valley contrast circular polarization behavior. Can the author study how the circular polarization changes when a WSe2/PtSe2 heterostructure forms?  
  
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