**Authors' Responses**

**Editor:**

We sincerely thank the editor for taking time to carefully review our work. We wish to note that the comments and questions from the editor are extremely insightful. We have carefully addressed all the questions/comments by performing new measurements and by improving the clarity of the presentation. In the following, we outline a point by point reply.

1) Specify the targeted application in the Title, Abstract, Introduction, and Conclusions so that the manuscript is consistent with the theme of the journal.  What specific application is particularly well-suited for these materials?

**Authors:**

We would like to thank the eidtor for this comment. We have added discussions of the specificapplication in the above mentioned parts. The 2D anisotropic materials have shown great promise in photonic applications such as polarization-sensitive photodetectors, optical components, polarized light-emitting diodes, polarized lasers.

2) For clarity and to be consistent with the theme of the journal, a revised title is needed; I suggest: Anisotropic Properties of Tellurium Nanoflakes Probed by Polarized Raman and Transient Absorption Microscopy: Implications for  **Authors:**

We thank the editor for the suggestion. We have revised it as the following:

Anisotropic Properties of Tellurium Nanoflakes Probed by Polarized Raman and Transient Absorption Microscopy: Implications for Polarization-sensitive Applications.

3) When the title is revised, please be sure to update the electronic submission form as well as the manuscript and Supporting Information files.   
**Authors:**

We appreciate the editor's reminder.

4) Please add a scheme near the beginning to illustrate the system (structure/chemical composition) and the synthesis/fabrication from a chemistry perspective.

**Authors:**

We completely agree with the editor’s comment. We have added Fig. 1(a) to illustrate hydrothermal growth of Te nanosheets.

5) The tellurium nanoflakes need more detailed characterization, including SEM and TEM with elemental mapping.

**Authors:**

We thank the editor for such a great suggestion. We completely agree with the editor. Following the editor’s comment, we have performed SEM, TEM and EDS characterization of Te nanosheets, as shown in Fig. 1(b-g).

6) The TOC graphic is missing.  In the TOC graphic, all components (including text) must be easily seen when reduced in size to the required dimensions (8.25 cm wide by 4.45 cm tall).  Notably, the width must be greater than the height. For guidance on TOC graphics, see https://pubs.acs.org/doi/10.1021/nn203713e.   
**Authors:**

We appreciate the editor's suggestion. We have added the TOC graphic.

7) Revise Figure 3. Font size too small   
**Authors:**

We completely agree with the editor’s comment. We have now enlarged the font size.

8) Funding Sources: Authors are required to report ALL funding sources and grant/award numbers relevant to this manuscript. Enter all sources of funding for ALL authors relevant to this manuscript in BOTH the Open Funder Registry tool in ACS Paragon Plus and in the manuscript to meet this requirement. See http://pubs.acs.org/page/4authors/funder\_options.html for complete instructions.

**Authors:**

We have revised the funding Sources.

9) ORCID: Authors submitting manuscript revisions are required to provide their own validated ORCID iDs before completing the submission, if an ORCID iD is not already associated with their ACS Paragon Plus user profiles. This iD may be provided during original manuscript submission or when submitting the manuscript revision. You can provide only your own ORCID iD, a unique researcher identifier. If your ORCID iD is not already validated and associated with your ACS Paragon Plus user profile, you may do so by following the ORCID-related links in the Email/Name section of your ACS Paragon Plus account. All authors are encouraged to register for and associate their own ORCID iDs with their ACS Paragon Plus profiles. The ORCID iD will be displayed in the published article for any author on a manuscript who has a validated ORCID iD associated with ACS Paragon Plus when the manuscript is accepted. Learn more at <http://www.orcid.org>.

**Authors:**

We have provided author’s own validated ORCID iDs.

10) On resubmission, please provide 2 copies of the final manuscript file:   
  
a) The final revised manuscript file that does not contain any highlighting or editing marks. This file should be uploaded as the primary manuscript document file.   
b) A marked copy of the revised manuscript that shows changes made on revision clearly highlighted. This file should be uploaded SEPARATELY FROM THE FINAL MANUSCRIPT FILE as Supporting Information for Review.

**Authors:**

We have uploaded (a) and (b) copies of the final manuscript file.

**Reviewer: 1**

Recommendation: Major revisions needed as noted.

Comments:

The manuscript titled Highly Anisotropic Properties of Tellurium Nanoflake Probed by Polarized Raman and Transient Absorption Microscopy by authors demonstrated that 2D Te nanoflakes are prepared by hydrothermal method. Highly anisotropic properties are illustrated by polarized raman spectrums and transient absorption spectroscopic measurements as well as density functional theory (DFT) calculation. Photo-carrier dynamics are probed by temporally resolved transient measurements with an exciton lifetime of 25 ps. These results support 2D Te as a promising layered material for developing 2D anisotropic devices and applications. In general, this work seems to be quite interesting and I would like to recommend the acceptance of this work provided that authors can well address the following questions before its final acceptance.

**Authors:**

We would like to thank the reviewer for evaluating our manuscript and we appreciate the positive comments on the overall quality of our work. Below, we will provide point-to-point responses to all technical comments and describe the revisions made.

1) Please comment on how to take advantage of the Anisotropic Properties of Tellurium Nanoflake. Any potential device application?

**Authors:**

Anisotropic properties of tellurium nanoflake is beneficial to control variability and uniformity of device performance by manipulating crystal orientation. The 2D anisotropic materials have shown great promise in photonic applications such as polarization-sensitive photodetectors, optical components, polarized light-emitting diodes, polarized lasers.

2) Please try to improve the language this work. There are many typos. For example, illustated.

**Authors:**

Thanks for mentioning this problem. Typos have already been correct in manuscript.

3) Did authors investigate the case of different layers of Tellurium?

**Authors:**

Thanks very much for your question. The flakes were prepared by synthesized by a hydrothermal method, and the thickness of those flakes are quite similar. We did not study the thickness dependence of Tellurium in this work.

4) Besides Tellurium, whether other Xene can show similar Anisotropic properties as Tellurium?

**Authors:**

Yes, phosphorene also shows similar anisotropic properties. However, there is no other Xene with these properties except Tellurium as far as we know.

5) In terms of Transient Absorption Microscopy, did authors investigate whether different polarized light will lead to different light exciton lifetime?

**Authors:**

Thanks very much for your question. We did not investigate this materials particularly with different polarized light. However, the similar measurements were conducted in our previous work (phosphorene). From the results, there is no obvious difference when different polarized light were applied. Exciton lifetimes should be independent with different combination of polarized light .

6) Related work on photonics of Tellurium, such as, Two-Dimensional Tellurium: Progress, Challenges, and Prospects, NANO-MICRO LETTERS 12, DOI10.1007/s40820-020-00427-z; Enhanced Photodetection Properties of Tellurium@Selenium Roll-to-Roll Nanotube Heterojunctions , SMALL 15, 1900902 DOI10.1002/smll.201900902; Two-dimensional tellurium-polymer membrane for ultrafast photonics , NANOSCALE 11, 6235-6242 DOI10.1039/c9nr00736a; 2D Tellurium Based High-Performance All-Optical Nonlinear Photonic Devices, ADVANCED FUNCTIONAL MATERIALS 29, 1806346 DOI10.1002/adfm.201806346; authors should have a discussion on them in the introduction part of the revision.

**Authors:**

We thank the reviewer for the suggestion. We have added a discussion in the introduction part as the following:

Strong nonlinear optical responses of Te illustrates its potential applications for nonlinear photonics and optoelectronic devices. Besides, combining Te with other materials, such as Te/PVP membranes and Te@Se roll-to-roll nanotubes, further improves the performance of practical applications.

7) Did authors investigate the light matter effect in Tellurium Nanoflake? Such as, by Z-scan measurement?

**Authors:**

Thanks very much for your question. Light matter effect is always a significant area, our group have conducted some works on other materials with multiple harmonic generation, including graphene and ReS2. However, the main idea of this manuscript is the anisotropic properties, this effect may sound irrelevant here. We may conduct some measurements in next work.

8) How about the long term stability of Tellurium Nanoflake? Did authors find the evidence of oxidation of Tellurium Nanoflake by Raman spectrum?

**Authors:**

Thanks very much for your question. As we mentioned, there is no degradation of tellurium nanoflakes during all measurements, which suggests that this material has great stability. Also, no evidence of oxidation is found when Raman spectrums were taken.

9) In the Raman measurement, did authors investigate the existence of additional new peaks due to the change of thickness?

**Authors:**

Thanks very much for your question. The measurements were taken in limited samples as we mentioned, we were unable to investigate new peaks due to the change of thickness. Also, since the flakes are actually bulk, there is no big difference in raman spectrums. Additional new peaks only occur when the samples are monolayer or few-layer in low wave-number.

**Reviewer: 2**

Recommendation: Publish after minor revisions noted.

Comments:

In this work, the authors have systematically investigated the highly anisotropic properties of tellurium nanoflake probed by polarized raman and transient absorption microscopy. The topic is interesting and they also show some interesting results. This manuscript shows that the authors fabricated 2D Te nanoflakes via hydrothermal method, and their qualities have been confirmed with raman map. The anisotropic properties of this material in optical response and carrier dynamics have been predicted by first-principle calculations, and further been confirmed with optical measurements, including polarized raman spectrums and transient absorption spectroscopic measurements. Besides, dynamics of photocarries are studied with an lifetime of 25 ps.

This work has a deep discussion of the arguments. However, there are some minor issues need to be further improved as well. And a couple of specific comments for the authors are listed below:

**Authors:**

We would like to thank the reviewer for evaluating our manuscript and we appreciate the positive comments. Below, we will provide point-to-point responses to all technical comments and describe the revisions made.

1)The first sentence of introduction “transition metal oxides, hexagonal boron nitride (hBN)” need change to “transition metal oxides, and hexagonal boron nitride (hBN)”.

**Authors:**

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

2)In the results and discussion of page 3, Figure 1d and Figure 1e are not described before the description of Figure 1f.

**Authors:**

Thanks for pointing out this error. The descriptions of Figure 1d and Figure 1e have been added in manuscript.

3) In page 5 of the manuscript, the 2th line from the bottom. The authors claimed that “The low-frequency optical branches E1(bond bending vibration) at 104 cm-1, high-frequency optical branches A1(bond breathing mode) at 82 cm-1 and E2(bond stretching mode) at 130 cm-1 are in agreement with the our raman scattering results where three vibration modes are at 122 cm−1, 94 cm−1 and 142 cm−1(see Fig.2 (a)), respectively”. However the above description is not conform to Figure 2a, please check it.

**Authors:**

We are sorry for the inaccurate description. Our theoretical calculations of the phonon spectrum are intended to explain the vibrational modes of Raman scattering experimentally. The E1, A1 and E2 peaks of the phonon spectrum are slightly lower than the Raman vibration results due to the absence of spin-orbit coupling. Therefore, previous sentences are revised as:

Due to the absence of spin-orbit coupling, the low-frequency optical branches E1 (bond bending vibration) at 104 cm−1, high-frequency optical branches A1 (bond breathing mode) at 82 cm−1 and E2 (bond stretching mode) at 130 cm−1 are slightly lower than our Raman scattering results where three vibration modes are at 122 cm−1, 94 cm−1 and 142 cm−1 (see Fig.2 (a)), respectively.

4)Whether other 2D materials are also have highly anisotropic properties, and how this property affects the devices that made from 2D materials? (such as “Vacancy-Induced Synaptic Behavior in 2D WS2 Nanosheet−Based Memristor for Low-Power Neuromorphic Computing” and “A New Memristor with 2D Ti3C2Tx MXene Flakes as an Artificial Bio-Synapse”) Please explain in the manuscript.

**Authors:**

Yes, black phosphorus, group IV monochalcogenides (SnS, SnSe, GeSe, GeS), and TMDs MTe2 and ReX2 (M = Mo, W, and X = S, Se) also show highly anisotropic properties. For instance, the electronic and phonon properties of strongly anisotropic materials are dependent on the in-plane orientation. These different properties along different crystal orientations are enhanced when transistors are built along a specific direction. In the photoconduction operating mode, anisotropic materials can control the crystal orientation to enhance the separation of photogenerated electron-hole pairs. This characteristic can increase the photoconductive sensitivity and achieve an ultrahigh responsivity and external quantum efficiency (EQE) under precise regulation. In the photocurrent operating mode, the regulated crystal orientations of anisotropic materials can tune the photogenerated current very well by changing the built-in electric field at junctions to both improve the responsivity and control the wavelength range of light detection.

5)Authors need to check the format and writing issues in the manuscript. Such as Equation 5 is not aligned, and “(see Fig.1 (d)” is without closing parenthesis in page 8 of the manuscript the 2th line from the bottom.

**Authors:**

Thanks for helping us check the format and writing issues. In fact, all equations are center aligned, although equation 5 may look differently. The second issues have been corrected in manuscript.

**Reviewer: 3**

Recommendation: Not appropriate for ACS Applied Nano Materials.

Comments:

**Authors:**

We would like to thank the referee for pointing out these issues and we will be happy to edit the text further, based on helpful comments from the referee. Below, we will provide point-to-point responses to all technical comments and describe the revisions made.

1) Please clarify why the author chose 120 nm 2D Te flake to do the related optical measurements. The thickness of 120 nm Te nanoflakes can no longer be considered as “2D materials”. The experimental characterization needs to be performed again on thinner Te nanoflakes that make more sense as 2D materials.

**Authors:**

We completely agree with the reviewer’s comment. We have removed "2D" in our manuscript.

2) Please discuss in more detail the novelty and significance of this work. Overall, the manuscript is poorly constructed and has no novelty in advancing 2D Te anisotropic properties.

**Authors:**

As we mentioned, besides black phosphorene, tellurium is the second elemental materials which shows anisotropic properties. In this manuscript, the anisotropic properties of tellurium have been studied in several methods. First, a theoretical study was carried by DFT method. We calculated the band structure of this material, and obtained the effective mass of electrons and holes along different direction. The difference of effective mass provide a proof of anisotropic properties. Then, the anisotropic properties of tellurium have further confirmed by polarized raman spectrums and transient absorption spectroscopic measurements with 2-fold symmetry. These results provide basic understanding for implementing tellurium in various anisotropic optoelectronic devices, such as polarization-sensitive photodetectors.

3)It is mentioned that the measured exciton lifetime is 25 ps. Is it good or bad? Please discuss.

**Authors:**

Exciton lifetime is the nature of materials. Since our measurements have been verified for many years, the result is convincing. if you mean whether it is good for applications, the number is similar to other 2D materials, including WS2 and MoS2 monolayers. Consider our sample is bulk, exciton lifetime of monolayer or few layer Te could be even shorter. Hence, it is good for applications.

4) The authors mentioned the use of DFT calculation in the abstract, but there is no discussion or mention about the use. Where is the result from the DFT calculation? What do the authors hope to achieve using DFT calculation?

**Authors:**

Figure 1(g), (h) and figure 2(b) are the results from DFT calculation. DFT calculation could provide us electronic structure of tellurium and we could further obtain the effective mass along different direction, which is the origin of anisotropic properties in theory.