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Peer Review File

Strain-tuned topological phase transition and unconventional Zeeman effect in ZrTe5 microcrystals

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28th Jun 22

Dear Dr Du,

Thank you for submitting your manuscript, "Strain-tuned topological phase transition and unconventional Zeeman effect in ZrTe5 microcrystals", to Communications Materials. It has now been seen by 3 referees, whose comments are appended below. You will see that while they find your work of interest, some serious points are raised. We are interested in the possibility of publishing your study in Communications Materials, but would like to consider your response to these major concerns in the form of a revised manuscript before we can make a decision on publication. In particular, you will see that Reviewer #2 has severe concerns about the reliability of your results, due to serious issues in the fitting procedure (points 1-3 of the report) and the unverified derivation of Eq. 5 (point 4 of the report). We would naturally expect all other comments of the referees to be appropriately responded to, including their requests for further information and discussion.

We therefore invite you to revise and resubmit your manuscript, taking into account the points raised.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

When submitting your revised manuscript, please include the following:

-A response letter with a point-by-point reply to each of the referee comments and a description of changes made. Please include the complete referee report in the response letter. Please note that the response letter must be separate to the cover letter to the editors.

-A marked-up version of the manuscript with all changes to the text in a different colored font. Please do not include tracked changes or comments. Please select the file type 'Revised Manuscript - Marked Up' when uploading the manuscript file to our online system.

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We hope to receive your revised paper within three months; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we will close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Materials or published elsewhere in the meantime.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Toru Hirahara, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0002-2574-1708

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

Gaikwad et al. report magneto-transport measurements on $ZrTe_5$ with tunable uniaxial strain. This is a famous system reported to be in proximity to a topological phase transition, while direct charge transport evidence has been lacking. The authors present convincing evidence for the closing and reopening of the Dirac band gap depending on the strain, consistent with the predicted topological phase transition. Based on the analysis of the amplitude of the quantum oscillations, they also nicely demonstrate the strain-tunability of the spin-dependent Berry phase. The results are of interest to the researchers in the field, and the paper is well written. Therefore, I consider the subject matter suitable for Communications Materials. However, I have several questions and suggestions that I would like the authors to consider.

1. In the abstract, the authors mention that "we directly map a weak- to strong- topological phase transition through a gapless Dirac semimetal phase via quantum oscillations." However, while the closing of the band gap can be found in Fig. 3b that corresponds to the theoretically predicted topological phase transition, it is not experimentally clear whether the STI phase is realized after the reopening of the band gap. Can the authors strengthen this claim, for example, through comparison between the experimentally obtained spin-dependent Berry phase and theoretical calculations?

2. The critical pressure for the WTI-STI transition has been estimated to be around -0.2%, as shown in Fig. 3b. However, since all the mass gap sizes are shown to exceed five meV, it does not look clear whether the transition happens at around -0.2% or -0.3%. Can the authors give a more detailed explanation of how they determine the pressure where the mass gap closes?

3. Related to the previous question, the spin-dependent Berry phase in Fig. 4c shows a jump-like behavior at around -0.2%, while the jump for the strain-dependent effective g-factor is found at around -0.3% as shown in Fig. 3d. Can the authors give a qualitative explanation of the slightly different trend in the Berry phase and the g-factor?

4. It has been widely predicted that strain can induce topological phase transitions. In fact, theoretical and experimental efforts have been paid to investigate the strain effects on various materials such as Bi2Se3 and TaSe3 [Young et al., Phys. Rev. B 84, 085106 (2011), Liu et al., Phys. Rev. B 84, 245105 (2011), Liu et al., Nature Phys 10, 294 (2014), Nie et al., Phys. Rev. B 98, 125143 (2018) and Lin et al., Nat. Mater. 20, 1093 (2021)]. I understand that it is not realistic to cite all the relevant papers, but several pioneering works should be properly cited in addition to refs [16-21] in the manuscript.

5. Based on Fig. 1d, the authors mention that the strain gauge factor ranges from 10^2 to 10^3. What is the unit for this value? Also, is this value large or small compared to other materials? Such comparison will give readers a better understanding of the strain tunability of ZrTe5.

Reviewer #2 (Remarks to the Author):

Please see the attached PDF file.

Reviewer #3 (Remarks to the Author):

In this work, Gaikwad et al. have studied strain-tuned topological properties of ZrTe5. They studied the SdH oscillations under the uniaxial strain and confirmed the weak and strong topological transition. They find the impact of the spin-dependent Berry phase is critical in quantum oscillations in Dirac semimetals. This work is done carefully and the result is a good complement to the existing research of the well-known ZrTe5. Therefore, I recommend it to publish in Communications Materials.

I have a small suggestion. There have been some works on the experimental study of ZrTe5 under strain, which have been cited in this work. But can the authors make some comparisons between the results of this work and the previous ones? For example, under what strain the topological phase transition occurs? And whether the size of the bandgap is consistent with each other?

In this paper, the authors report on the variation of SdH oscillation for 3D Dirac semimetal $ZrTe_5$ upon external strain. They used a microcrystal on a flexible polyimide substrate and applied strain by bending the substrate. They analyzed the SdH oscillation based on the modified LK formula, which takes into consideration the Zeeman effects on the Fermi surface and Berry phase. The obtained mass gap and Fermi energy exhibit anomaly at the strain of -0.2%, which the authors identify as a topological transition.

The authors present the systematic experimental data on strain and the results of their analysis. However, as detailed below, there are several serious problems about the analyzing methods and hence the main claim of this paper is not scientifically reliable. Moreover, the presentation of this manuscript is poor; neither the fundamental properties for $ZrTe_5$ nor the experimental and theoretical details are not well explained. As a result, readers unfamiliar with $ZrTe_5$ will not be able to understand either the validity of the analysis or the importance of this study. Therefore, I don't recommend the publication of this manuscript in Communications Materials. After addressing the following points, it may be worth publishing in a more specialized journal.

1. How did the authors determine the values of α, μ, and Δ by fitting? These parameters appear only in the inside of cosine term of R_S and hence they should not be uniquely determined. Therefore, the results shown in Figs. 3 and 4 are not reliable.
2. For each strain, the fitting method is not consistent. In Figs. 2d and 2e, a few discrete ΔR values are plotted, while ΔR values in Fig. 2f are almost continuous.
3. 2nd paragraph on page 5: "Under large compressive and large tensile strains, we observed approximately single SdH oscillation frequencies and a monotonic decay of SdH oscillation amplitude over increasing $1/B$, suggesting that SdH oscillations are dominated by single bands there."

   However, in Fig. 3a, two frequencies are plotted over the entire range of strain. If the SdH oscillation is dominated by a single band, the fitted results assuming two frequencies are totally unreliable.
4. Derivation of Eq. 5 (shown in Appendices B and C) is highly nontrivial and appears to be unpublished. In addition, the physical meaning of coefficient α is unclear. Consequently, the validity of the results obtained from Eq. 5, such as Figs. 3f and 4d, is not ensured.
5. Please explain in the main text the fundamental properties of crystal structure and Fermi surface for $ZrTe_5$. In particular, in inset to Fig. 2c, it is not clear which Fermi surfaces correspond to Dirac-like bands and which to trivial parabolic bands.
6. Please explain the detail of the strain-induced topological transition in $ZrTe_5$. In which direction and at what degree of strain should the transition from weak to strong TI occur? The authors also should compare the present results with those obtained in the previous studies, such as Refs. [34]

and [35], and discuss them. Is the magnitude of strain at the topological transition consistent with that reported in Refs. [34] and [35]?

7. The authors mention that the sample was encapsulated with PMMA. Is this sufficient for the sample to adhere firmly to the substrate? How do the authors verify that the substrate strain is actually applied to the sample? In addition, detailed information on the uniformity of strain is needed.
8. 1st paragraph on Page 3: "… which gives rise to the nonzero g-factor terms in the two-band Dirac model for the low energy subspace, g_p and g_s."

   Please explain why there are two types of g factors for two-band Dirac model. What do the authors mean by g_p and g_s?
9. Neither Fig. 3e nor Fig. 4b is explained in the main text at all. They could be removed.
10. 1st paragraph on page 6 appears to explain Fig. 2f, but this is not clearly stated.
11. In the main text and figures, there are two kinds of notations for Fermi energy: μ and E_F. Is there any difference between them?
12. In Fig. 1b, the microcrystal and electrodes cannot be distinguished in the photograph.

Reply to Reviewer #1

Gaikwad et al. report magneto-transport measurements on ZrTe5 with tunable uniaxial strain. This is a famous system reported to be in proximity to a topological phase transition, while direct charge transport evidence has been lacking. The authors present convincing evidence for the closing and reopening of the Dirac band gap depending on the strain, consistent with the predicted topological phase transition. Based on the analysis of the amplitude of the quantum oscillations, they also nicely demonstrate the strain-tunability of the spin-dependent Berry phase. The results are of interest to the researchers in the field, and the paper is well written. Therefore, I consider the subject matter suitable for Communications Materials. However, I have several questions and suggestions that I would like the authors to consider.

We thank the reviewer for the supportive response and constructive comments. Below is our point-by-point reply to the reviewer's questions/comments.

1. In the abstract, the authors mention that "we directly map a weak- to strong- topological phase transition through a gapless Dirac semimetal phase via quantum oscillations." However, while the closing of the band gap can be found in Fig. 3b that corresponds to the theoretically predicted topological phase transition, it is not experimentally clear whether the STI phase is realized after the reopening of the band gap. Can the authors strengthen this claim, for example, through comparison between the experimentally obtained spin-dependent Berry phase and theoretical calculations?

We fully agree with the reviewer that the closing and reopening of a gap does not necessarily guarantee a topological phase transition. A topological phase transition would require that the gap changes sign which, based on recent theoretical studies, does not necessarily induce an abrupt change in the quantum oscillations (Phys. Rev. X, 8, 011027,2018). Here our claim of a topological phase transition is based on a more quantitative comparison between the experimental observations and theoretical modeling. In particular, a sign-change in band gap is revealed by the strain-dependent SdH amplitude factor Rs (which reflects the spin-dependent Berry phase), shown in Figure 3e. The sign of the band gap Δ affects the value of Rs and its strain dependence. Our experimental observations are consistent with the band gap having sign-change over the transition.

We have added a discussion to strengthen our claim, as suggested by the reviewer.

2. The critical pressure for the WTI-STI transition has been estimated to be around -0.2%, as shown in Fig. 3b. However, since all the mass gap sizes are shown to exceed five meV, it does not look clear whether the transition happens at around -0.2% or -0.3%. Can the authors give a more detailed explanation of how they determine the pressure where the mass gap closes?

We agree with the reviewer that the exact value of strain is not very accurately determined here. This is mainly due to two reasons. First of all, we have limited data points near the critical strain of the WTI-STI transition. Secondly, the calculation of the gap value is dependent on the choice of Dirac band velocity. We adopted the reported value of ~5e6 m/s and fine-tuned it within ~10% to optimize the overall fitting quality for our data. But not knowing the actual velocity gives uncertainty to the exact transition point. The transition strain is a very rough estimation based on Fig.3b.

We would also like to note, as stated in the paper, that all the values of strain are relative to when the substrate is not bent. But there is always certain unintentional strain from the fabrication process. So, the absolute value of strain is unknown. Instead we focus on the (more experimentally practical) tuning of band structure and emergence topological phase transition over ***changing*** strain.

3. *Related to the previous question, the spin-dependent Berry phase in Fig. 4c shows a jump-like behavior at around -0.2%, while the jump for the strain-dependent effective g-factor is found at around -0.3% as shown in Fig. 3d. Can the authors give a qualitative explanation of the slightly different trend in the Berry phase and the g-factor?*

We thank the reviewer for the keen observation! Tracing back on our analysis and the figures, we found a typo in the data column which generates the plot in Fig.4d, on the data point with the strain of -0.24% (we compute the effective g-factor by calculating α using Eq.5, using Matlab; then we type the results into OriginLab to generate the plot and the .eps vector image). After correcting the data point, Fig.4c and Fig.4d show the same trend in the strain dependence. We have carefully double-checked all the other data points and they are identical in Matlab and in OriginLab.

4. It has been widely predicted that strain can induce topological phase transitions. In fact, theoretical and experimental efforts have been paid to investigate the strain effects on various materials such as Bi2Se3 and TaSe3 [Young et al., Phys. Rev. B 84, 085106 (2011), Liu et al., Phys. Rev. B 84, 245105 (2011), Liu et al., Nature Phys 10, 294 (2014), Nie et al., Phys. Rev. B 98, 125143 (2018) and Lin et al., Nat. Mater. 20, 1093 (2021)]. I understand that it is not realistic to cite all the relevant papers, but several pioneering works should be properly cited in addition to refs [16-21] in the manuscript.

We have added the references suggested by the reviewer in the manuscript.

5. Based on Fig. 1d, the authors mention that the strain gauge factor ranges from 10^2 to 10^3. What is the unit for this value? Also, is this value large or small compared to other materials? Such comparison will give readers a better understanding of the strain tunability of ZrTe5.

Gauge factor is defined as $(\Delta R/R)/(\Delta L/L)$ and is unitless. The gauge factor observed in ZrTe5 is very large compared to that of the metal thin films (~2) , and is also a bit larger than typical single crustal semiconductors such as Si and Ge (~100). The large gauge factor manifests strain-sensitive band structure and is also facilitated by the small Fermi surface. We have added a sentence to highlight this point.

Reply to Reviewer #2

In this paper, the authors report on the variation of SdH oscillation for 3D Dirac semimetal ZrTe5 upon external strain. They used a microcrystal on a flexible polyimide substrate and applied strain by bending the substrate. They analyzed the SdH oscillation based on the modified LK formula, which takes into consideration the Zeeman effects on the Fermi surface and Berry phase. The obtained mass gap and Fermi energy exhibit anomaly at the strain of -0.2%, which the authors identify as a topological transition. The authors present the systematic experimental data on strain and the results of their analysis. However, as detailed below, there are several serious problems about the analyzing methods and hence the main claim of this paper is not scientifically reliable. Moreover, the presentation of this manuscript is poor; neither the fundamental properties for ZrTe5 nor the experimental and theoretical details are not well explained. As a result, readers unfamiliar with ZrTe5 will not be able to understand either the validity of the analysis or the importance of this study. Therefore, I don't recommend the publication of this manuscript in Communications Materials. After addressing the following points, it may be worth publishing in a more specialized journal.

We thank the reviewer for the critical comments which help us improve the manuscript. The main criticisms from the reviewer include 1). Data analysis is not clearly described, which leads to the reviewer's doubt on the main claim of this work; 2) Some introductory and experimental details are lacking, for the general audience. Below we provide a point-by-point answer to the reviewer's questions/comments, which we hope will clarify all the issues raised by the reviewer.

1. How did the authors determine the values of a, μ, and Δ by fitting? These parameters appear only in the inside of cosine term of RS and hence they should not be uniquely determined. Therefore, the results shown in Figs. 3 and 4 are not reliable.

We would like to clarify that the values of the band parameters (including *μ and Δ*) are not obtained from fitting R_S. Instead they are determined by fitting the 1) period; 2) magnetic field dependence and 3) temperature dependence of the SdH oscillations, using the LK formula (Eq.4). These are associated with the R_T, R_D, and the prefactor of 1/B in the cosine term in the LK formula.

The term R_S is not used for determining the band parameters. Instead, R_S comes out of each fitting as a constant factor, and we obtain the strain-dependence of the values of R_S which reflects the strain-tuning of band parameters. We then try to understand such dependence by comparing it with the theoretic model. There we found out that in order to understand the strain- (hence band parameter-) dependence of R_S, it is necessary to take into account a spin-dependence Berry phase. This leads to Eq. 2 and Eq.5, which compute α and gives a relation between R_S and band parameters (*μ, and Δ*) that is consistent with our experimental observation, as shown by Fig.3e and 3f.

So, the process of analysis here is: SdH oscillations → band parameters (Δ, μ) → R_s ←→ α, ϕ_s and modeling.

We also note that in fitting the LK formula (which also contains a cosine term), all the fitting parameters for the SdH oscillations affect the period and amplitude of the oscillations. Therefore, there is no ambiguity in their values and the values of all these parameters are uniquely determined with small uncertainties.
Finally, we would also like to point out that the general fitting scheme for SdH oscillations with LK formula used here is a standard approach for obtaining band parameters. For a single band-associated SdH oscillations, the key band parameters are obtained by fitting the SdH oscillation period (in 1/B), and the magnetic field and temperature dependence of the oscillation ***amplitude***. While for a multiple-band associated SdH oscillations, the band parameters can be obtained by directly fitting the magnetic dependence of the oscillatory resistance lineshape. (See our reply to the next question regarding fittings in Fig.2d-f.)

2. For each strain, the fitting method is not consistent. In Figs. 2d and 2e, a few discrete DR values are plotted, while DR values in Fig. 2f are almost continuous.

We thank the reviewer for point out the issue of mixed labels in the Fig.2d-f (y-axis), which is indeed confusing. In Figs.2d and 2e, we fit the ***amplitude*** of SdH oscillations (explained below) versus 1/B, while in Fig.2f we directly fit the lineshape of the ***oscillatory part of the R vs. 1/B***. Both fittings are consistent with each other.

To differentiate the two types of fitting, we have changed the y-axis label in Fig2d and Fig.2e to A_{osc}, while keeping y-axis label in Fig.2f (ΔR).

Under strains where SdH oscillations can be modeled with a single 1/B period, directly fitting the temperature and magnetic field dependences of the SdH amplitude, i.e., the prefactor of the cosine term allows more accurate determination of the band parameters (as in Fig.2d and 2e), and such fittings are very commonly used in the SdH oscillations analysis. Here we first read out the oscillation amplitudes and their corresponding 1/B values at every resistance peaks and valleys from the measured SdH oscillations. These correspond to the discrete points in Fig.2d and 2e. We then fit their 1/B dependence at every temperature, following the LK formula. Finally, the band parameters can be calculated based on the fittings and the (single) period of the SdH oscillations (in 1/B).

For two-component SdH oscillations, the above fitting method becomes impractical because there are two cosine terms and one cannot directly read out the 1/B-dependent amplitudes the two sets of oscillations. Instead, one needs to directly fit the lineshape of the ΔR vs. 1/B curves with the two-component LK formula.

In our fittings, we assume that band parameters (including Fermi energy and band gap) evolve smoothly with changing strain. Under single-band dominated strains, band parameters obtained in Fig.2d and2e serve as “reference points”: smoothly modifying these parameters under gradually changing strain greatly helps the fitting process and allow identification of band parameters for the more general 2-band system with values which are physical (i.e., no abrupt ”jump” over gradually changing strain).

3. 2nd paragraph on page 5: "Under large compressive and large tensile strains, we observed approximately single SdH oscillation frequencies and a monotonic decay of SdH oscillation amplitude over increasing 1/B, suggesting that SdH oscillations are dominated by single bands there." However, in Fig. 3a, two frequencies are plotted over the entire range of strain. If the SdH oscillation is dominated by a single band, the fitted results assuming two frequencies are totally unreliable.

First we would like to clarify that for most of the strains, the SdH oscillations are of 2-band nature, with comparable contributions from the Dirac and the parabolic bands. In addition, the topological phase transition happens over strains where the Dirac band plays a dominating role (with a larger weight than the parabolic band) in the SdH oscillations. So, for the main claims of this work (which focuses on the strain-evolution of the Dirac band), the results are positively solid and reliable. In our SdH analysis, we choose band parameters for each band and their relative weight to optimize the fittings, and the results are shown in Fig.3a.

However, it is true that under large tensile strains, SdH oscillations are dominated by a single parabolic band. There, changing the fitting parameters of the minority band does not very significantly affect the lineshape of the fitting curves. In our revised manuscript we use semitransparent color to present these points and add a description in the figure caption.

4. Derivation of Eq. 5 (shown in Appendices B and C) is highly nontrivial and appears to be unpublished. In addition, the physical meaning of coefficient a is unclear. Consequently, the validity of the results obtained from Eq. 5, such as Figs. 3f and 4d, is not ensured.

The equation 5 is derived from the standard semiclassical theory of the Bloch electrons with the low energy effective model being the anisotropic Dirac models. The detail derivation process has been fully presented in the appendices. Although our result has not been published before, its isotropic limit actually agrees well with the analytical results presented in reference: Phys. Rev. X, 8, 011027, 2018 (from Glanzmann's group).
The physical meaning of α is the splitting of the k-space area enclosed by two otherwise degenerate Fermi surface under magnetic field, as stated in our manuscript.

5. Please explain in the main text the fundamental properties of crystal structure and Fermi surface for ZrTe5. In particular, in inset to Fig. 2c, it is not clear which Fermi surfaces correspond to Dirac-like bands and which to trivial parabolic bands.

We have added a discussion in the main text on the fundamental properties of $ZrTe_5$. Regarding Fig.2c, we did have a description in the figure caption: "The inset show the pockets of Fermi surface, with the Dirac band at the Γ point and a parabolic side band between the R and E points dominating the SdH oscillations under strong compressive and tensile strains, respectively. "
We have also clarified this in the revised main text, following the reviewer's suggestion.

6. Please explain the detail of the strain-induced topological transition in ZrTe5. In which direction and at what degree of strain should the transition from weak to strong TI occur? The authors also should compare the present results with those obtained in the previous studies, such as Refs. [34] and [35], and discuss them. Is the magnitude of strain at the topological transition consistent with that reported in Refs. [34] and [35]?

The strong-to-weak TI transition happens as the strain is tuned from compressive to tensile, as shown in Fig.3b and Fig.4d., and is consistent with the previous reports. The magnitude of absolute strain is complicated by the presence of a random built-in strain (typically tensile, due to the hot transfer process using PDMS) during the sample fabrication. Nevertheless, the applied strain range of several ~0.1% is comparable to the previous experimental works. In addition, we also found that the transition happens near the strain of minimal zero-field resistance, which is consistent with Sci. Adv. 2019;5: eaav9771. The bandgap observed through our magnetotransport study is strain tunable. Within the range of strain of a few 0.1% it varies by a few 10meV. This is in good agreement with the previous report on ARPES study of ZrTe5 under strain (Nat. Comm (2021) 12:406).

We have added a discussion comparing our results with the previous reports.

7. The authors mention that the sample was encapsulated with PMMA. Is this sufficient for the sample to adhere firmly to the substrate? How do the authors verify that the substrate strain is actually applied to the sample? In addition, detailed information on the uniformity of strain is needed.

The application of uniaxial strain through substrate bending has been extensively studied as a reliable and effective approach previously. In our work reported here, the application is strain is manifested by the **elastic** tunability of the charge transport characteristics. All results discussed in our work are reproducible upon repeated application of substrate bending. (i.e., same resistance is measured under the same strain, independent on the history of strain application). The gauge factor observed in ZrTe5 is very large compared to that of the metal thin films (~2) , and is also a bit larger than typical single crystal semiconductors such as Si and Ge (~100). The large gauge factor manifests strain-sensitive band structure and is also facilitated by the small Fermi surface.

We did observe that with very severe substrate bending, the resistance of the sample starts drifting with time and becomes non-reproducible, suggesting possible sliding between PMMA and ZrTe5. We avoid the data after such observations.

In our experimental setup, the strain homogeneity is expected to be small, due to the total emersion of the crystal in PMMA, and more importantly the extremely small ratio of crystal thickness (~190nm) to the total crystal length (~100um, we note that the length of the crystal outside the electrodes also helps creating an uniform strain throughout the thickness of the crystal). A discussion on the importance of this ratio can be found in: Sci. Adv. 2019;5: eaav9771 (Materials and Methods section). Here in our setup the thickness-to-length ratio is over an order of magnitude smaller than what has been reported previously. This guarantees an unprecedented strain uniformity in our measurements.

We have added a brief discussion in Appendix A to address these concerns from the reviewer.

8. 1st paragraph on Page 3: "… which gives rise to the nonzero g-factor terms in the two-band Dirac model for the low energy subspace, gp and gs." Please explain why there are two types of g factors for two-band Dirac model. What do the authors mean by gp and gs?

The two lowest bands near the Fermi level of ZrTe5 are originated from bonding (g_s) and anti-bonding (g_p) Te 5p orbitals. The g-factors for these two different bands are very different according to our first-principle calculations, which are labeled as g_s and g_p respectively. More detail discussion about how the g-factor get renormalized for different bands in crystals with strong SOC can be found in the reference 43 of the revised manuscript (First Principle Calculation of the Effective Zeeman's Couplings in Topological Materials, Chapter 11, pages 263–281. World Scientific, 2021.).

9. Neither Fig. 3e nor Fig. 4b is explained in the main text at all. They could be removed.

We thank the reviewer for pointing this out. These figures are important for the discussion of our work and indeed should be cited in the main text. We have added citations to these two figures in the main text.
"The theory shows good qualitative agreement with the data on the strain dependence of Rs, as illustrated in Fig. 3f with the band parameters tuned along the trajectory in the (Δ,μ) parameter space shown in Fig.3e."
"For systems with nontrivial Berry curvature, the Berry phase splitting ϕ_s along the Fermi surface will be nonzero (Fig.4b)"

10. 1st paragraph on page 6 appears to explain Fig. 2f, but this is not clearly stated.

We are not sure what the reviewer meant by "1st paragraph on page 6" (which is not related to the explanation of Fig.2f). However we hope our explanation to the fitting process (answers to questions 1,2) clarifies the reviewer's question on Fig.2f.

11. In the main text and figures, there are two kinds of notations for Fermi energy: μ and EF. Is there any difference between them?

We thank the reviewer for pointing out this typo. We have corrected the notation of E_F in the main text to μ.

12. In Fig. 1b, the microcrystal and electrodes cannot be distinguished in the photograph.

We have added labels to the electrodes and the microcrystal in the figure.

Reply to Reviewer #3

In this work, Gaikwad et al. have studied strain-tuned topological properties of ZrTe5. They studied the SdH oscillations under the uniaxial strain and confirmed the weak and strong topological transition. They find the impact of the spin-dependent Berry phase is critical in quantum oscillations in Dirac semimetals. This work is done carefully and the result is a good complement to the existing research of the well-known ZrTe5. Therefore, I recommend it to publish in Communications Materials.

I have a small suggestion. There have been some works on the experimental study of ZrTe5 under strain, which have been cited in this work. But can the authors make some comparisons between the results of this work and the previous ones? For example, under what strain the topological phase transition occurs? And whether the size of the bandgap is consistent with each other?

We thank the reviewer for the supportive response and constructive suggestions. We have added a brief discussion compare our results with the previous works.

As stated in our manuscript, the values of strain in this work are "external" strains, in relative to when the substrate is not bent. And the absolute strain can have a significant offset from the random built-in stress (typically tensile), resulting from the hot-transfer fabrication process. Nevertheless the order of magnitude of strain studied here (~few 0.1%) is comparable with the previous experimental works on STI-WTI transitions.

The strong-to-weak TI transition happens as the strain is tuned from compressive to tensile, as shown in Fig.3b and Fig.4d., and is consistent with the previous reports. In addition, we observed the STI-WTI transition to happen near the minimum resistance point (Fig.1e). This observation is consistent with the previous experimental study on chiral magnetic effect in ZrTe5 under strain (Sci. Adv. 2019;5: eaav9771).

The bandgap observed through our magnetotransport study is strain tunable. Within the range of strain of a few 0.1% it varies by a few 10meV. This is in good agreement with the previous report on ARPES study of ZrTe5 under strain (Nat. Comm (2021) 12:406).

7th Sep 22

Dear Dr Du,

Thank you for submitting your manuscript, "Strain-tuned topological phase transition and unconventional Zeeman effect in ZrTe5 microcrystals", to Communications Materials. It has now been seen by 3 referees, whose comments are appended below. You will see that while they find your work of interest, some important points are raised. Especially, Reviewer #2 is still raising some important points which we think should be clarified. We are interested in the possibility of publishing your study in Communications Materials, but would like to consider your response to these concerns in the form of a revised manuscript before we make a decision on publication.

We therefore invite you to revise and resubmit your manuscript, taking into account the points raised.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

When submitting your revised manuscript, please include the following:

-A response letter with a point-by-point reply to each of the referee comments and a description of changes made. Please include the complete referee report in the response letter. Please note that the response letter must be separate to the cover letter to the editors.

-A marked-up version of the manuscript with all changes to the text in a different colored font. Please do not include tracked changes or comments. Please select the file type ‘Revised Manuscript - Marked Up’ when uploading the manuscript file to our online system.

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We hope to receive your revised paper within three months; please let us know if you aren’t able to submit it within this time so that we can discuss how best to proceed. If we don’t hear from you, and the revision process takes significantly longer, we will close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Materials or published elsewhere in the meantime.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

Please do not hesitate to contact me if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Toru Hirahara, PhD
Editorial Board Member
Communications Materials
orcid.org/0000-0002-2574-1708

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

I consider that the authors have fully addressed the questions raised and that the article can now be published in Communications Materials.

Reviewer #2 (Remarks to the Author):

The authors responded to most of the referee's comments and partially revised the manuscript. However, in my opinion, the most important issue about the data analyses has yet to be addressed as explained below. Therefore, I cannot recommend the publication of the present manuscript.

1. I am confused by the authors' response 3:
"However, it is true that under large tensile strains, SdH oscillations are dominated by a single parabolic band. There, changing the fitting parameters of the minority band does not very significantly affect the lineshape of the fitting curves."

This statement clearly indicates that the SdH period of the Dirac band has a large error bar under tensile strains. In this region, furthermore, the SdH periods for a parabolic band and Dirac band are comparable (Fig. 3a), which means that the Dirac band is not a minority band in terms of carrier density. From these facts, there should be large error bars in Fermi energy, carrier density and band gap as well. In addition to the color change of the fitted data, the authors should quantitatively show the error bars in Figs. 3 and 4. If the error bars are large for the single-1/B-period region, the authors' claims are not experimentally supported.

2. Related to the above, the fitting method (single or two-component 1/B period) changes at the strain of -0.2%, where the authors insist that the topological transition of the Dirac band occurs. I suspect the abrupt changes in band parameters of the Dirac band at this strain come from the difference in fitting method. The authors should anlayse the data using the fitting method consistent over the entire strain region. Moreover, if the contribution from the parabolic band suddenly disappears below -0.2% strain, some transition may happen to the parabolic band (not the Dirac

one).

3. I would recommend that the authors include the summary of the response 1 in the manuscript, since the fitting procedure is important when there are many fitting parameters.

Reviewer #3 (Remarks to the Author):

The author has responded to my comments, and now I recommend publishing the revised manuscript.

Reply to Reviewer 2

1. I am confused by the authors' response 3:"However, it is true that under large tensile strains, SdH oscillations are dominated by a single parabolic band. There, changing the fitting parameters of the minority band does not very significantly affect the lineshape of the fitting curves."

This statement clearly indicates that the SdH period of the Dirac band has a large error bar under tensile strains. In this region, furthermore, the SdH periods for a parabolic band and Dirac band are comparable (Fig. 3a), which means that the Dirac band is not a minority band in terms of carrier density. From these facts, there should be large error bars in Fermi energy, carrier density and band gap as well. In addition to the color change of the fitted data, the authors should quantitatively show the error bars in Figs. 3 and 4. If the error bars are large for the single-1/B-period region, the authors' claims are not experimentally supported.

We thank the reviewer for the suggestion which motivate us to evaluate the uncertainty of our SdH oscillation simulations, especially under tensile strains.

First, we would like to clarify that the weight contribution to the SdH oscillations from each band is mainly determined by its amplitude factors (here R_S plays a major role, as result of cancellation from the 2 spin bands). Therefore, while the Dirac band is not a minority band in terms of carrier density it can still have a small SdH weight.

Our original analysis focus on identifying a set of strain-dependent parameters which agree well with the magnetic field and temperature dependences of the SdH oscillations, and are also "physical" in that they evolve smoothly with strain. Following the reviewer's suggestion, we checked the uniqueness/sensitivity of the choices of parameters, and estimated the ranges of the parameters which allow reasonable simulations to the SdH line-shapes. The raw fitting parameters for the Dirac band and their corresponding uncertainties are summarized below. Detailed numbers for each strain are added to the revised table SI in the Supplementary Information, and are shown by error bars in the figures in the main text.

Dirac Band	Compressive and zero	Tensile 0.1%	Tensile 0.2%	Tensile 0.3%
1/B period (1/Tesla)	+/- 0.002 (~1-2%)	+/- 0.01(~5%)	+/- 0.02 (10%)	+/-0.05 (25%)
cyclotron mass (m_e)	+/- 0.002-0.003(5-10%)	+/- 0.005 (10%)	+/- 0.01 (20%)	+/-0.02 (40%)
SdH weight	+/- 0.05 (5-10%)	+/- 0.1 (15%)	+/- 0.2 (30%)	+/- 0.4 (80%)

We note here that: the 1/B period can be very precisely determined from the peak/valley positions of the SdH oscillations for most strains where the SdH oscillations from the two bands have well-separated periods; cyclotron mass is determined from its effects on the 1/B and temperature decay of the SdH oscillations; and the SdH effects the overall line-shape of the SdH oscillations.

The very large uncertainties at 0.3% tensile strain originate from the accidental overlap of the SdH periods from the Dirac and the parabolic bands (as shown in Figure 3a). (If such overlap is exact, then there will be no way to tell the weights from the two bands). Out of precaution, we have removed the 0.3% strain-associated data points in the main text. (However, we should note that this doesn't imply that the original parameters at this strain were incorrect. They ARE consistent with the SdH line-shape and the conclusions, and they are "physical" in terms of strain-evolution. But mathematically they are not sufficiently unique.)

The main source of error is the uncertainty in the cyclotron mass, whose effect is magnified when calculating the mass gap (through standard error propagation). Nevertheless, the overall strain dependence is well in agreement with a topological phase transition (Figure 3b), and the amplitude of the SdH oscillations agrees well with the theory which takes into account the spin-dependent Berry phase (Figure 3f).

In summary, based on our analysis to the SdH oscillation, all but the 3 tensile strain points (one removed on the revised manuscript) are semi-quantitatively consistent with the theories on topological phase transition in ZrTe5 and on spin-dependent Berry phase; the 3 tensile strain points are at least **not inconsistent** with the conclusions.

From different perspective, since the experimental data are objective, we can also confidently claim that the theory predicts band parameters which lead to SdH oscillations that are consistent with the ALL experimental observations (including all points under tensile strains).

2. Related to the above, the fitting method (single or two-component 1/B period) changes at the strain of -0.2%, where the authors insist that the topological transition of the Dirac band occurs. I suspect the abrupt changes in band parameters of the Dirac band at this strain come from the difference in fitting method. The authors should anlayse the data using the fitting method consistent over the entire strain region. Moreover, if the contribution from the parabolic band suddenly disappears below -0.2% strain, some transition may happen to the parabolic band (not the Dirac one).

We would like to clarify and stress that we did not change the fitting method for different strains. Eventually all the SdH curves were simulated with the two-band LK formula. But for strains (the two shown in Figure 2d,e) where the SdH oscillations are dominated by a single period (which are both far from the topological transition), we were able to perform SdH amplitude fittings and obtain band parameters with error bars(Figure 2d,e). We did so because such analysis bypasses the complication of the residue background and hence allows accurate determination of the band parameters. It is also very commonly performed and widely accepted in single-period SdH analysis. And in the LK formula simulations, these band parameters at these two strains were further confirmed to be consistent with the SdH line-shape. We then used these two special cases as references, and smoothly modified the parameters under changing strains in the more general two-band LK formula simulations for all other strains. Both of these single-period strains are away from the topological transition. The key features in the obtained band parameters which support the conclusions on topological phase transition and the spin-dependent Berry phase are definitively not due to any inconsistency in the analysis.

Regarding the rather sudden decrease of the parabolic band contribution under strong compressive strains, it is possible that the bottom of this (conduction) band moves out of the Fermi sea with compressive strain within the range of the strain we applied. This would not necessarily correspond to a phase transition. Further experimental and computational work focusing on the parabolic band would be needed, which is beyond the focus and scope of this work.

3. I would recommend that the authors include the summary of the response 1 in the manuscript, since the fitting procedure is important when there are many fitting parameters.

We have added error bars to the Dirac band parameters in the related figures and brief discussions on the uncertainty of the analysis in the main text.

18th Oct 22

Dear Dr Du,

Your manuscript titled "Strain-tuned topological phase transition and unconventional Zeeman effect in ZrTe5 microcrystals" has now been seen again by our referees, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Materials under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to revise your paper one last time to comply with our journal policies and formatting style, in order to maximise the accessibility and therefore the impact of your work.

Please note that it may still be possible for your paper to be published before the end of 2022, but in order to do this we will need you to address these points as quickly as possible so that we can move forward with your paper.

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Best regards,

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orcid.org/0000-0002-2574-1708

&

Dr Aldo Isidori
Senior Editor
Communications Materials

REVIEWERS' COMMENTS:

Reviewer #2 (Remarks to the Author):

The authors responded to Referee’s comments and revised the manuscript by adding error bars. In my opinion, this paper is now technically sound and can be published.