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Peer Review File

Title

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The authors study the transport and magnetic properties of a Dirac semimetal $ZrTe_5$. The temperature dependence of susceptibility is found to be correlated with the temperature dependence of resistivity. They further compare the results with simulation and conclude that the magnetic behavior can be well explained by the Berry paramagnetism of Dirac electrons. The authors claim that "This work not only proposes a new experimental way to determine the topological character of the band but also provides a new platform to apply the concept to π Berry phase-related band topology to Dirac semimetals." However, Ref. 6 has already demonstrated such an experiment for the purpose and Ref. 8 applied the method to $ZrTe_5$. The authors carried out similar experiments on the same material and followed the same theoretical analysis. Thus, this work is incremental to previous studies and may be suitable for a more specialized journal.

Some other comments that the authors may consider:

How is the magnetic susceptibility calculated? It is odd that the amplitudes of the susceptibility of three samples are close in Fig. 5A, however, the amplitudes of magnetization differ substantially in Fig. 4A. The confusion continues, as the susceptibilities become almost equal in Fig. 5C.

The abbreviation of EPMA needs to be explained.

In the SI, Ref. 21, 22 are cited, but those are irrelevant in the context.

The page number of Ref. 19 is wrong.

Reviewer #2 (Remarks to the Author):

In this manuscript, the authors grow the single crystals of $Ti_xZr_{1-x}Te_5$ (x = 0, 0.1, and 0.2). In the first part, the authors preformed detailed electrical transport measurements and revealed that the isovalent substitution induces the band shift. Further quantum oscillation analyses show that Ti doped $ZrTe_5$ crystals are 3D Dirac semimetals. In the second part, the authors studied the magnetic properties of $Ti_xZr_{1-x}Te_5$ and found that the temperature T_m of magnetic susceptibility minimum is close to the temperature T_p of resistivity peak. The authors interpreted those observations in terms of the Berry paramagnetism.

The article is timely and paves a new way to determine the band topology by magnetism. My evaluation of this manuscript is positive for a publication in Communication Physics.
I have some comments for the authors to improve the manuscript in order to benefit the readers.

1)Pervious works show that the $ZrTe_5$ is a semi-3D Dirac system with quadratic dispersion in the b-direction and linear in the others [1,2]. Whether such scenario can explain the minimum of magnetic susceptibility at T_m is most remarkable when B//b-axis, but less pronounced in other field directions?

2)A recent work by Nair et al. also reveals a thermodynamic signature of Dirac electrons at low temperatures in $ZrTe_5$ [3]. The model of Berry paramagnetism in this study is quite similar to the model presented in Nair's work. However, I noticed that the experimental data in these two works are inconsistent [See Fig. 4a in this work and Fig.1a in Ref [3]. The author should clarify this point.

3)For CVT samples, two types of electrons are found at low temperatures [4]. One hosts the Dirac nature as extensively discussed in this work. The other one comes from the trivial band. How about their contribution to magnetization and susceptibility? Are they negligible? Why？

4)The abstract does not capture the key point of the paper and needs to reorganize.

References

[1] NPG Asia Mater. 8, e325(2016).
[2] Phys. Rev.lett 118 206601 (2017).
[3] Phys. Rev. B 97, 041111(R) (2018)
[4] Nat. Commun. 7,12516 (2016).

Reviewer #3 (Remarks to the Author):

This manuscript report on Ti doped $ZrTe_5$ material. Recently $ZrTe_5$ has attracted considerable attention both theoretically and experimentally due to three-dimensional quantum Hall effect and metal–insulator transition.

The authors have synthesized the single crystals of $Ti_xZr_{1-x}Te_5$ (x=0, 0.1 and 0.2) and investigated using the transport and magnetic measurements. Their results of transport on x=0 are in agreement with previous published work. Further, they have shown that the band topology of Dirac semimetals can be verified by the magnetic properties. Their magnetization and resistivity results of $ZrTe_5$ show correlation and scaling into a single curve. Further their simulation of the magnetization data show agreement with the experiment and supports their interpretation in terms of the Berry paramagnetism. The work presented on $ZrTe_5$ also agrees with that reported by Nair et al, Phys. Rev. B 97, 041111 (2018).

The results presented in this work provide interesting changes in the properties of $ZrTe_5$ with Ti doping. I recommend the manuscript to be published in Communications Physics after the authors have considered following comments.

My main comment on the work is on Berry paramagnetism they are discussing. Can the authors give direct evidence of the presence of paramagnetic signal in their material and a detail explanation on the Berry paramagnetism and how it is different than conduction electron diamagnetism? In their experimental data of the magnetization (both temperature and field dependence), I do not see any sign of paramagnetic signal.

Other comments are:

(1)The authors have mentioned that "Though a similar scheme has been applied in $ZrTe_5$ [Ref. 8], our crystals show qualitatively different properties."

Comment: What are these different properties? What are differences between your crystal and the crystal presented in Ref. [8]?

(2) At line-56, "we observed the resistivity peak at Tp = 122 K, " Why not use Tρ instead of Tp?

(3) At line 64, it is mentioned that "…. and an additional phase shift of 1/8".
Comment: Shift in what and what is origin of this additional phase shift? Please clarify this.

(4) Figs.4 A, B. Please write the name of the samples.

(5) At line 113 it says "… converted into the conductivity, σxy and σxy (Fig. SI). It should be "..σxx and σxy…"

(6) In SM Fig.S3 caption it is mention that "Low lifetime for x = 0.1 sample might be from that the crystallinity of the sample can be not great as the other samples"

Comment: What do the authors mean by the crystallinity of the sample? How it was measured? Can authors compare measured crystallinity of all the samples?

(7) τtr, what is physical meaning of this parameter? Please give clear explanation for this in the revised manuscript.

(8) At line 268 they have mentioned, " tune the band structure due to the decrease in the lattice constant"

Comment: Has this changed in the band structure calculated directly from DFT calculations? It would be interesting to see this result.

(9) They have mentioned, "This result suggests that the carriers contributing to the transport properties play an important role in the magnetic properties"

Comment: What is new in this statement? If there are no localized moments in the material then both transport and magnetic properties will be dominated by the carriers contributing.

(10) In SM Fig.S4 &55, Why the magnetization (M) unit is arb. unit/? This simulation is for which composition of Ti? Why simulated M is not plotted with negative sign as given in Fig. S2?

Reviwers' Comments and Replies:

Reviewer #1 (Remarks to the Author):

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→ As the reviewer commented, Ref. 6 reported similar experiments in NbAs and Ref. 8 applied the method to $ZrTe_5$. For that reason, we cited these papers in our manuscript, but the differences do not seem to be clearly stated. Therefore, we would emphasize the distinguished points here and revise the manuscript accordingly.

In Ref. 6, they studied a Weyl semimetal of NbAs to which Berry paramagnetism can be applied. In our study, we tried to extend the concept of Berry paramagnetism to Dirac semimetals. We chose $ZrTe_5$ because it is promising to apply the Berry paramagnetism due to the temperature-induced band shift, i.e., Lifshitz transition. Although similar analytical method was used to $ZrTe_5$ in Ref. 8, as the reviewer pointed out, we would emphasize the significant difference between their and our samples. The most distinguished points are summarized in Fig. R1. $ZrTe_5$ used in Ref. 8 displays the resistivity peak at T_p = 34 K, interpreted as a topological phase transition, while $ZrTe_5$ in our study exhibits the typical behavior of Lifshitz transition at T_p = 122 K. Note that the magnetization shown in Ref. 8 monotonically decreases with increasing temperature, compared to our result showing the magnetic susceptibility minimum at T_m (= T_p). Importantly, in our study we demonstrated the correlation between the electrical resistivity and magnetic susceptibility, compared to the work in Ref. 8. Moreover, we applied the consistent interpretation to the experimental results of $Ti_xZr_{1-x}Te_5$, where the Ti doping induces the band shift. In addition, we observed prominent quantum oscillations of both SdH and dHvA effects, which were not shown in Ref. 8, and confirmed the nature of 3D Dirac fermions with anisotropic Fermi surface. We have revised the manuscript and added this figure (as Fig. S1) in the supplementary information to explain the differences from the previous works.

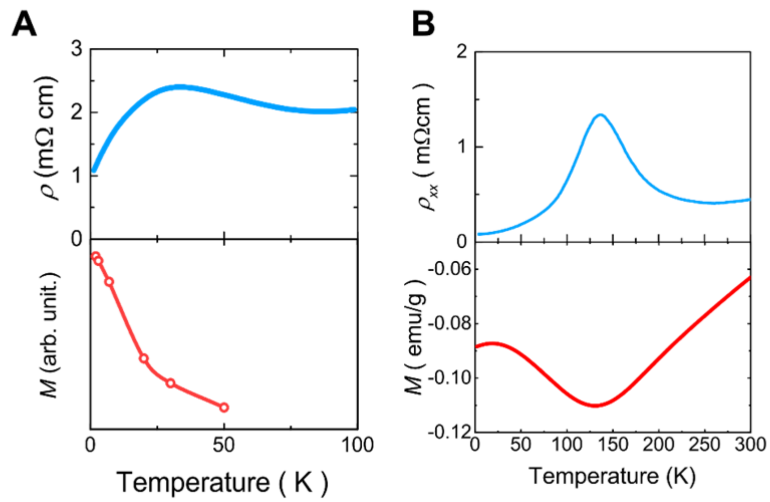

Fig. R1. Comparison between the previous work and the present work. Temperature-dependent resistivity and magnetization, $\rho(T)$ and $M(T)$, for $ZrTe_5$ taken from **(A)** Ref. 8 and **(B)** this study.

Some other comments that the authors may consider:

How is the magnetic susceptibility calculated? It is odd that the amplitudes of the susceptibility of three samples are close in Fig. 5A, however, the amplitudes of magnetization differ substantially in Fig. 4A. The confusion continues, as the susceptibilities become almost equal in Fig. 5C.

→ Thanks for your careful reading and for pointing out our mistake. The reason that the data differ in the two Figs. 4A and 5A is because different sample holders were used. Since the straw holder used for Fig. 4A has a background signal depending on the sample space and the quartz holder used for Fig. 5A has negligible background signal, we have removed the background contributions and corrected the magnetization data in Figs. 4A and 5F.

As for Fig. 5C, we just shifted the data vertically, but we did not mention it in the figure caption, which caused confusion. Thus, we have corrected the magnetic susceptibilities normalized with the minimum values to $\chi/|\chi(T_m)|$ vs. T-T_m plot for clarity, as we did in the resistivity plot of Fig. 5D.

The abbreviation of EPMA needs to be explained.

→ We have explained the abbreviation of EPMA in the revised version.

In the SI, Ref. 21, 22 are cited, but those are irrelevant in the context.

→ We have rearranged the references to relevant ones. Thanks again for reading carefully.

The page number of Ref. 19 is wrong.

→ We have corrected the wrong page numbers.

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In this manuscript, the authors grow the single crystals of TixZr1-xTe5 (x = 0, 0.1, and 0.2). In the first part, the authors preformed detailed electrical transport measurements and revealed that the isovalent substitution induces the band shift. Further quantum oscillation analyses show that Ti doped ZrTe5 crystals are 3D Dirac semimetals. In the second part, the authors studied the magnetic properties of TixZr1-xTe5 and found that the temperature Tm of magnetic susceptibility minimum is close to the temperature Tp of resistivity peak. The authors interpreted those observations in terms of the Berry paramagnetism.

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→ Thank you for valuable comment. It is correct that the band dispersion is related to the anisotropic magnitude of magnetic susceptibility, which is obviously observed in our experimental data shown in Fig. 5A. The magnetic signal along the b direction is much larger than the others because of smaller effective mass in the ac plane. On the other hand, the magnetic susceptibility shows a minimum because the Berry paramagnetism is changed by the Lifshitz transition. This implies that the minimum feature of magnetic susceptibility does not necessarily depend on the band dispersion. Note that the magnetic susceptibility data in Fig. 5A show the minimum at T_m but the different magnitude depending on the field direction.

2) A recent work by Nair et al. also reveals a thermodynamic signature of Dirac electrons at low temperatures in ZrTe5 [3]. The model of Berry paramagnetism in this study is quite similar to the model presented in Nair's work. However, I noticed that the experimental data in these two works are inconsistent [See Fig. 4a in this work and Fig.1a in Ref [3]. The author should clarify this point.

→ As we described in the manuscript, $ZrTe_5$ is located at the phase boundary between weak and strong topological insulators, providing that its physical properties change very sensitively to external parameters, even the growth methods and growth conditions, so that there have been many debating results. The reason why the experimental data in the two works are inconsistent is because the two samples are different. The most distinguished points are summarized in Fig. R1. $ZrTe_5$ used in Nair's work displays the resistivity peak at T_p = 34 K, interpreted as a topological phase transition, while

$ZrTe_5$ in our study exhibits the typical behavior of Lifshitz transition at T_p = 122 K. Note that the magnetization shown in Nair's work monotonically decreases with increasing temperature, compared to our result showing the magnetic susceptibility minimum at T_m (= T_p). Importantly, in our study we demonstrated the correlation between the electrical resistivity and magnetic susceptibility, compared to the work in Nair's et al. Moreover, we applied the consistent interpretation to the experimental results of $Ti_xZr_{1-x}Te_5$, where the Ti doping induces the band shift. In addition, we observed prominent quantum oscillations of both SdH and dHvA effects, which were not shown in Nair's work, and confirmed the nature of 3D Dirac fermions with anisotropic Fermi surface. As the reviewer commented, we have revised the manuscript and added this figure (as Fig. S1) in the supplementary information to clarify this difference.

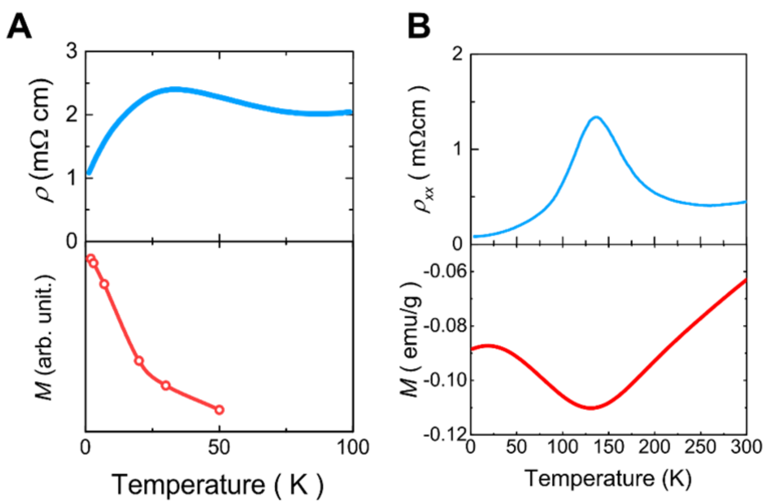

Fig. R1. Comparison between the previous work and the present work. Temperature-dependent resistivity and magnetization, $\rho(T)$ and $M(T)$, for $ZrTe_5$ taken from **(A)** Ref. 8 and **(B)** this study.

3) For CVT samples, two types of electrons are found at low temperatures [4]. One hosts the Dirac nature as extensively discussed in this work. The other one comes from the trivial band. How about their contribution to magnetization and susceptibility? Are they negligible? Why ?

→ The trivial band contributes Pauli paramagnetism and Landau diamagnetism, which are normally temperature independent. Moreover, these contributions are small when the carrier density is low (n_2 < 10^{17} cm^{-3}). These results suggest that the trivial band of carrier 2 serves as a small magnetization that is almost independent of temperature.

4) The abstract does not capture the key point of the paper and needs to reorganize.

→ We appreciate the reviewer's comment in helping us improve the quality of our manuscript. The abstract has been reorganized to capture the key point of our paper, as the reviewer suggested.

References

[1] NPG Asia Mater. 8, e325(2016).

[2] Phys. Rev. Lett. 118 206601 (2017).

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The results presented in this work provide interesting changes in the properties of ZrTe5 with Ti doping. I recommend the manuscript to be published in Communications Physics after the authors have considered following comments.

My main comment on the work is on Berry paramagnetism they are discussing. Can the authors give direct evidence of the presence of paramagnetic signal in their material and a detail explanation on the Berry paramagnetism and how it is different than conduction electron diamagnetism? In their experimental data of the magnetization (both temperature and field dependence), I do not see any sign of paramagnetic signal.

→ As in the manuscript, there are two types of electrons in $ZrTe_5$. One hosts the Dirac nature which was extensively discussed in the paper, and the other comes from the trivial band. The conduction electrons in the trivial band contributes Pauli paramagnetism and Landau diamagnetism, which are normally temperature independent. Moreover, these contributions are small when the carrier density is low (n_2 < 10^{17} cm^{-3}). The results suggest that the trivial band of carrier 2 serves as a small magnetization that is almost independent of temperature. On the other hand, the nontrivial conduction band above the Dirac point contributes the Berry paramagnetism, while the valence band below the Dirac point is diamagnetic. In the manuscript, we displayed the total magnetization, that is the sum of Berry paramagnetic and diamagnetic signals. In Figs. S4 and S5 of our previous supplementary information, we showed the simulated Berry paramagnetism only. For clarity, we plot each of these two (Berry para- and dia-) signals separately, which is shown in Fig. R1. Please note that the magnitude of paramagnetic signal is smaller than that of diamagnetic signal, resulting that the total magnetization looks to be diamagnetic. In order to avoid confusion, we have added this figure (as Fig. S7) in the revised supplementary information.

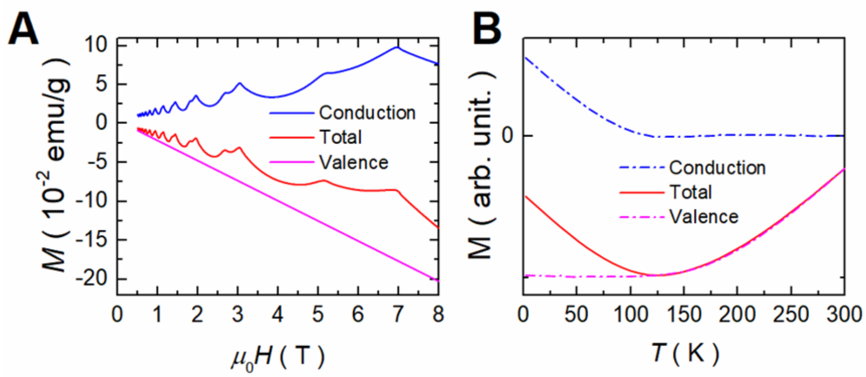

Fig. R1. (A) Simulated field-dependent and (B) schematic temperature-dependent magnetization curves (red), which is the sum of the Berry paramagnetism (blue) and diamagnetism (magenta). These simulated curves are consistent with the experimental data shown in Figs. 5A and 5F.

The Berry paramagnetism comes from the field-independent 0^{th} Landau level (LL). Unlike the parabolic case, this LL stays intact regardless of magnetic field strength and the Landau level degeneracy grows with the field; hence the 0^{th} LL acts like a reservoir. Therefore, thermodynamic potential can be reduced unless only 0^{th} LL is occupied. This reduction of thermodynamic potential with respect to the magnetic field gives paramagnetic contribution, which is referred to as Berry paramagnetism. The conventional conduction electron with parabolic dispersion does not have this contribution. In the revised version, we have added the detail explanation on the Berry paramagnetism.

Other comments are:

(1) The authors have mentioned that "Though a similar scheme has been applied in ZrTe5 [Ref. 8], our crystals show qualitatively different properties."

Comment: What are these different properties? What are differences between your crystal and the crystal presented in Ref. [8]?

→ As plotted in Fig. R2, the most different physical property between their and our crystals is that $ZrTe_5$ in Ref. [8] displays the resistivity peak at T_p = 34 K, interpreted as a topological phase transition, while ZrTe5 in our study exhibits the typical behavior of Lifshitz transition at T_p = 122 K. In addition, the magnetization shown in Ref. [8] monotonically decreases with increasing temperature, compared to our result showing the magnetic susceptibility minimum at T_m (= T_p). This temperature-dependent susceptibility is one of the crucial evidences of band shift in $ZrTe_5$, which was extensively discussed in our manuscript. We have revised the manuscript and added this figure (as Fig. S1) in the supplementary information to explain the differences from the previous work.

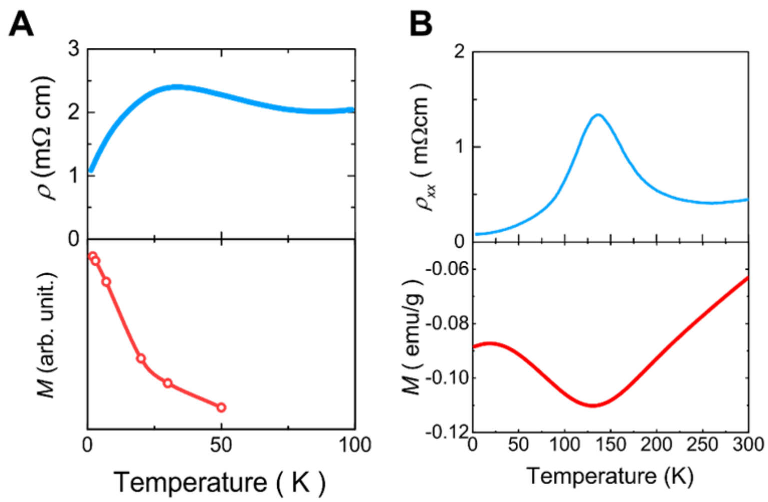

Fig. R2. Comparison between the previous work and the present work. Temperature-dependent resistivity and magnetization, $\rho(T)$ and $M(T)$, for $ZrTe_5$ taken from **(A)** Ref. 8 and **(B)** this study.

(2) At line-56, "we observed the resistivity peak at Tp = 122 K, " Why not use Tr instead of Tp?

→ In the manuscript, we used the subscript of "p" for the meaning of resistivity peak, and the subscript of "m" for the meaning of susceptibility minimum.

(3) At line 64, it is mentioned that "…. and an additional phase shift of 1/8".
Comment: Shift in what and what is origin of this additional phase shift? Please clarify this.

→ The SdH and dHvA oscillations can be described by the Lifshitz-Kosevich formula, $\cos[2\pi(F/B + \gamma)]$, where B is the magnitude of magnetic field, and F is the oscillation frequency, and γ is the phase shift. It is widely believed that an energy band with linear dispersion carries an extra π Berry phase, leading to phase shifts of $\gamma = 0$ and ±1/8 in 2D and 3D Fermi surfaces, respectively [1-3], compared to 1/2 and 1/2±1/8 for parabolic energy bands without the Berry phase. The sign of the phase shift depends on the curvature of the Fermi surface along the applied field direction. The explanation for the additional phase shift is written on page 7 in the present manuscript.

1. Liu, Y. *et al.* Nat. Commun. **7**, 12516 (2016).
2. Wang, J. *et al.* PNAS **115**, 9145–9150 (2018).
3. Hu, J., Xu, *et al.* Annu. Rev. Mater. Res. **49**, 207–252 (2019).

(4) Figs.4 A, B. Please write the name of the samples.

→ We have written the name of the samples in all the figures.

(5) At line 113 it says "… converted into the conductivity, σxy and σxy (Fig. SI). It should be "..σxx and σxy…"

→ We have corrected the typos. Thank you for the careful reading.

(6) In SM Fig.S3 caption it is mention that "Low lifetime for x = 0.1 sample might be from that the crystallinity of the sample can be not great as the other samples"

Comment: What do the authors mean by the crystallinity of the sample? How it was measured? Can authors compare measured crystallinity of all the samples?

→ Thank you for your valuable comment. Initially, we intended to mean the crystallinity as a kind of structural disorder, which could be induced by Ti doping. However, after reading your comment and thinking again, we felt that the word "crystallinity" is not appropriate. Besides, we do not have any experimental evidence for the comparison of crystallinity, and thus we have removed such ambiguous word from the manuscript.

(7) ttr, what is physical meaning of this parameter? Please give clear explanation for this in the revised manuscript.

→ The transport lifetime, τ_{tr}, is evaluated from the Hall coefficient (= $1/ne$) and the electrical conductivity (= $ne^2\tau_{tr}/m^*$) measurements, compared to the quantum lifetime, τ_q, that is calculated from the analysis of quantum oscillations. τ_{tr} is a measure of the motion of charged particles along the electric field gradiant, while τ_q is the mean time a carrier remains in a particular state before being scattered to a different state. Thus, τ_{tr} mainly concerns the backscattering, so that it can overweight large scattering angle over small angle. As the reviewer commented, we have added more explanation for the physical meaning of τ_{tr} in the revised manuscript.

(8) At line 268 they have mentioned, " tune the band structure due to the decrease in the lattice constant"

Comment: Has this changed in the band structure calculated directly from DFT calculations? It would be interesting to see this result.

→ In the discussion part, we just wanted to mention the possibility of changing the band structure by Ti doping because the topological properties (related to the band structure) of $ZrTe_5$ changes sensitively depending on the crystal volume change (related to Ti doping). We inferred this from the experimental results of the shift of T_p and T_m shown in electrical resistivity and magnetic susceptibility, respectively. Unfortunately, there are no DFT calculations for the Ti-substituted $ZrTe_5$ system.

(9) They have mentioned, “This result suggests that the carriers contributing to the transport properties play an important role in the magnetic properties”

Comment: What is new in this statement? If there are no localized moments in the material then both transport and magnetic properties will be dominated by the carriers contributing.

→ It is generally accepted that the transport properties are governed by carrier motion around the Fermi energy, whereas the magnetization is a bulk property related to the total magnetic moment below the Fermi energy. In that sense, we thought it surprising that the carriers contributing to the transport properties contribute to the magnetic properties, especially in such low carrier density system. However, we recognized that the English phrase might be confusing to deliver our point, so that we have corrected the above sentence.

(10) In SM Fig.S4 &55, Why the magnetization (M) unit is arb. unit/? This simulation is for which composition of Ti? Why simulated M is not plotted with negative sign as given in Fig. S2?

→ As frequently reported in previous simulation data, we just used arbitrary units. In the revised version, we redisplayed the simulation data by using emu/g. The simulation was performed for $x = 0$, which has been added in the figure caption.

As we answered in the reviewer's main comment, we first simulated the data with the contribution of Berry paramagnetism as shown in Figs. S4 and S5 of our previous supplementary information. Then, we added the diamagnetic contribution from the valence band electrons, which was not displayed. In Fig. R1, we plot each of these two (Berry para- and dia-) signals separately. For clarity, we have added this figure (as Fig. S7) in the revised supplementary information.

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The manuscript has been improved in some aspects. As for my first concern, however, I am not convinced by the authors that this manuscript represents a significant advance in understanding of the Berry paramagnetism, given the previous works. I agree that there are differences between ZrTe5 samples in Ref. 8 and this work. The carrier density in Ref. 8 is lower. This is in fact advantageous as it enables reaching the quantum limit, so that a transition of a Berry paramagnetism to diamagnetism can be observed, a clear evidence of the Berry paramagnetism. In contrast, this work reports only a suppression of a diamagnetism, a relatively indirect evidence. In Ref. 8, the Berry paramagnetism dominates over the diamagnetism of the valence band at low fields. In sharp contrast, the diamagnetism is much stronger in this work. This needs to be explained, otherwise it adds confusion to the understanding of this effect. By the way, the sample in Ref. 8 is of high quality, as they DID observe SdH oscillations.

The submitted work is a more comprehensive study, as it presents the temperature dependence of the magnetization and investigation on doped materials.

Reviewer #2 (Remarks to the Author):

The authors have made clear all of my concerns, I have no more questions.

Reviewer #3 (Remarks to the Author):

I appreciate the authors for replying the comments of all three referees and revising the manuscript based on the referees’ comments. The authors’ replies to the referees’ comments are satisfactory and revised manuscript and supplementary material read very well. Recommend the manuscript to be published in Communications Physics.

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The submitted work is a more comprehensive study, as it presents the temperature dependence of the magnetization and investigation on doped materials.

→ We thank the reviewer for the valuable comment, and we totally agree with the reviewer's concern. We hope that this time we clearly answer your concern.

As reported earlier, the Dirac nature on the border of topological phase transition is often controversial. In particular, $ZrTe_5$ is located at the phase boundary between weak and strong topological insulators, which has provided more controversial results. As one representative example, the resistivity peak has been observed to be strongly sample-dependent and range from T_P = 10 K to 150 K. This peak position is the most fundamental difference between Ref. 8 (T_P = 35 K) and ours (T_P = 150 K), although the samples in the two works are of high quality, as manifested in the observation of quantum oscillations. It is also true that a low carrier density is advantageous to reach the quantum limit, which does not guarantee the observation of a Berry paramagnetic to diamagnetic transition.

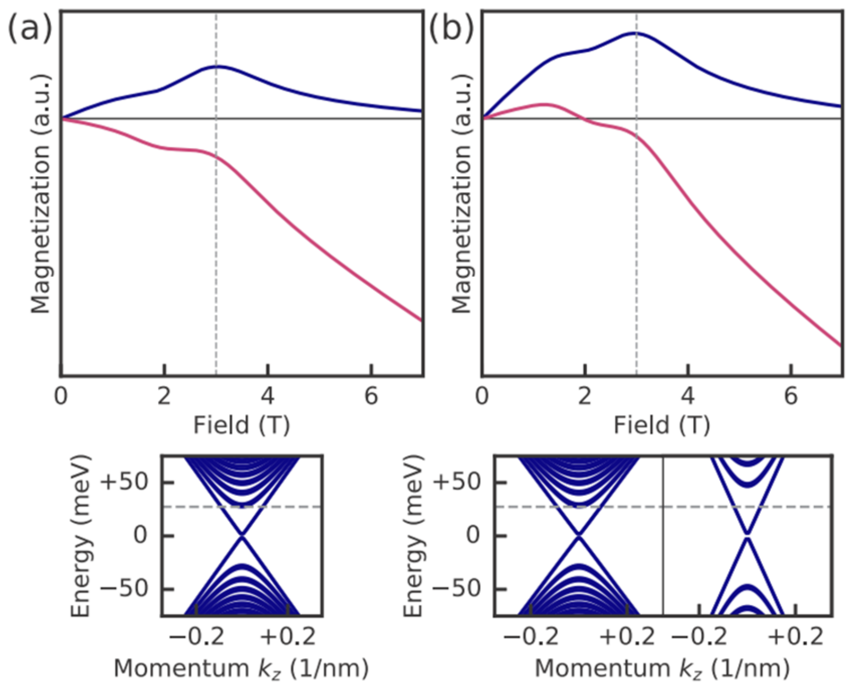

Fig. R1. Simulated magnetization of **(a)** a single Dirac band and **(b)** two Dirac bands [ref. 8], with (red) and without (blue) a linear diamagnetic background.

In Ref. 8, they observed a magnetization reversal from a paramagnetic response at the low field to a diamagnetic response at the high field. In order to explain the low-field paramagnetic signal, they employed a nonlinear diamagnetic background arising from the presence of a second Dirac band. As shown in Fig. 3(b) of Ref. 8 [see Fig. **R1(b)**], the simulated result based on this scenario where there are two Dirac bands reproduced the low-field paramagnetic signal, although they did not observe the quantum oscillations caused by the second Dirac band. They described that the second Dirac pocket enters its quantum limit at low fields and enhances the low-field paramagnetic signal. On the contrary, in our work, we observed a strong diamagnetic signal without any magnetization reversal. This feature is consistent with the simulated magnetization of a single Dirac band with a linear diamagnetic background, which is shown in Fig. 3(a) of Ref. 8 [see Fig. **R1(a)**]. As aforementioned, $ZrTe_5$ is located at the topological phase boundary and is extremely sensitive to volume change. Thus, the band structure of the two samples in Ref. 8 and ours can be topologically different: We have one Dirac cone with Lifshitz transition but no topological phase transition, while Ref. 8 shows two Dirac cones with topological phase transition but no Lifshitz transition. Therefore, these two works should be treated separately, and consequently, a comprehensive understanding of the Berry paramagnetism can be established when these two different works are well explained. We believe that this work would advance the understanding of the Berry paramagnetism by elucidating the features in the unexplored topological regime of $ZrTe_5$.

To clarify the difference, we have added more descriptions in Fig. S1 of the revised supplementary information.

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The authors have made clear all of my concerns, I have no more questions.

Reviewer #3 (Remarks to the Author):

I appreciate the authors for replying the comments of all three referees and revising the manuscript based on the referees' comments. The authors' replies to the referees' comments are satisfactory and revised manuscript and supplementary material read very well. Recommend the manuscript to be published in Communications Physics.

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

Thank the authors for their reply. I have no further comment and recommend its publication.