Room temperature Planar Hall effect in nanostructures of trigonal-$PtBi_2$
Response to referees

REPORT #2

We thank the reviewer for their extremely in-depth report. Below, we provide a point-by-point response to their comments. We have implemented multiple modifications to the manuscript in response, mainly by clarifying the link between this study and Ref. 19, more details on the analysis, more clarity on the different Weyl nodes in the system, a dedicated Methods section, and additional details about the samples. Modifications to the main text are highlighted in blue.

This manuscript reports transport measurements on the new topological material t-PtBi2. The main finding is the observation of the planar Hall effect that witnesses the influence of Weyl nodal lines on the electrical transport. This work certainly deserves publication as a very careful experimental study of an interesting new material. In my opinion, the manuscript would potentially meet the acceptance criteria of SciPost Physics with regard to advancing the characterization and understanding of a very non-trivial quantum material, the Weyl semimetal that combines nodal lines with the exotic effect of surface superconductivity. That said, the connection to these phenomena is somewhat indirect, as the present manuscript (in contrast to several other pre-prints and publications from the same group) lacks any explicit theory / ab initio component and analyzes the data on a rather empirical level. The main outcome, beyond the mere observation of the planar Hall effect, appears to be its unusual $B^{1.24}$ field dependence, but no further conclusions are derived from this result, and no explicit comparisons to the field dependence in other materials are made.

Several major issues have to be addressed by the authors:

1. The manuscript appears in parallel with arXiv:2410.02353 where at least part of the same data are used to reveal the so-called anomalous planar Hall effect (APHE). For example, Fig. 3a of the present manuscript seems to show almost the same data as Fig. S2A of the APHE preprint (sample D1 at 14 T and 50 vs. 100 K, respectively). However, the fits are different, especially in the case of Rxy where the APHE preprint shows a better fit. This difference can be traced back to the different fitting functions. The manuscript under consideration relies on an assumption that the Rxy signal may be contaminated by Rxx [Eq. (4)], but no such assumption is made in the APHE manuscript [Eq. (S5) in arXiv:2410.02353]. It feels very confusing. It also raises questions on the consistency of the analysis across the two concurrent manuscripts from the same group. A related question is the reasoning behind Eq. (4) vs. the discussion of the discrepancies in page 6. It is argued that a misalignment between the ab-plane of the sample and the rotation plane of the instrument would lead to the 2pi-periodic contribution to Rxy and the pi-periodic contribution to Rxx, but Eq. (4) is written under an assumption that the same kind of misalignment results in the 2pi-contribution to both Rxx and Rxy. Why the difference?

The reviewer raises several important points here, and we answer them below: the link between this manuscript and arXiv:2410.02353 (aka Ref. 19) and the similar data between the two manuscripts, the potential contamination of the Ryx signal with Rxx signal, the difference between the fits in the APHE and PHE manuscripts, and the periodicity of the contribution from out-of-plane fields due to sample misalignment.

1. First, this study is indeed mainly based on the same raw data as Ref. 19. The two manuscripts are however entirely distinct as to the effects they are studying: While this manuscript focuses on the PHE, which is a signature of Weyl physics, Ref. 19 focuses on an anomalous PHE (APHE) which is two orders of magnitude smaller, and originates from topological nodal lines. As a matter of fact, the first analysis done in Ref. 19 (e.g. the fits shown on Fig. S2) is to remove the PHE from the data, as it "hides" the smaller APHE. Therefore, the analysis of both papers is distinct: In this manuscript, the APHE is a vanishingly small perturbation other the PHE signal, and in Ref. 19, the PHE signal is treated as a background to the APHE signal. We have modified the introduction of the manuscript to clarify this: We note that, in a complementary study based on the same raw data[1], we evidence and thourougly analysed another effect known as the anomalous PHE (APHE). This effect, which is two orders of magnitude weaker PHE than the PHE, does not originate from Weyl nodes already existing without external magnetic field, but rather from a conversion of topological nodal lines (which we also

predict in that separate study) into very distant Weyl nodes (in k-space). Because of their different origin and different analysis required for both signals, as well as the fact the APHE constitutes but a small perturbation compared to the PHE signal, the two effects are studied separately in each study and the latter is simply ignored in the present study.

2. Second, regarding the assumptions related to the fits: The goal of the fit from eq. S5 of Ref. 19 (shown in Fig. S2) is to remove the π-periodic and 2π-periodic signals from the data, to evidence the APHE, which has a different symmetry (the APHE is $2\pi/3$-periodic). For this purpose, we simply remove a best fit of the data for both signals. This is akin to performing a stop-band filter. Since the combination of the π-periodic PHE signal in Rxx and Ryx still gives a π-periodic signal, and that the APHE has no component in Rxx, there was no need to consider Rxx contamination in the Ryx signal in the APHE manuscript.

3. Third, as said above the fits in Fig. S2 of the APHE manuscript are simple, unconstrained fits with a π-periodic component and a 2π-periodic component, and cannot be used to obtain quantitative information about the PHE. The fits shown in Fig. 3 of the present manuscript, meanwhile, are heavily constrained fits of the π-periodic component (e.g. the phase of the oscillation, the geometric constants and the amplitude has to stay the same for both Rxx and Ryx and between every magnetic field or temperature) made to ensure that the signal shows the hallmarks of a PHE. Therefore, the fits are not the same between the two papers.
We added a footnote to Fig. 3 to note the difference in the nature of the fits between this study nd Ref. 19: We note that, although the raw data shown is the same as in [1], the fits in the present figure are different from that other study, as the latter were unconstrained π- and 2π-periodic signals, while the presents ones are heavily constrained, as discussed in the main text..

4. Lastly, due to ab-plane misalignment of the sample in the rotation plane, we expect that the out-of-plane field will contribute a π-periodic signal to Rxx and a 2π-periodic signal to Ryx, due to the magnetoresistance and Hall responses being respectively even and odd in out-of-plane fields. We discuss this in page 6, in the deviations to the model, but also in page 4 (paragraph "Finally, as mentioned above..."). In eq. 4, we indeed consider only 2π-periodic background terms for both Rxx and Ryx. As stated in page 4, this is to take into account any possible 2π-periodic terms which may be in our data, so that they do not affect the quality of the fit for the π-periodic terms. Since the contribution of the out-of-plane field to Rxx is π-periodic, it will not be accounted for by the fit from eq. 4, and will therefore appear as a deviation between the fit and the data. Since we focus the analysis on the π-periodic signal, we decided to separate only the 2π-periodic contribution of the out-of-plane field (in Ryx). We have addapted the text on page 4 to make this more clear: In order to account for any possible 2π-periodic background, we consider additional 2π-periodic signals in both resistances ($\mathrm{R}_{xx}^{2\pi}$ and $\mathrm{R}_{yx}^{2\pi}$). Any out-of-plane field component should therefore result in a deviation of the longitudinal resistance from our fit, as discussed in the next section.

2. The data analysis is certainly complicated by the possible contamination of the Rxy signal with Rxx. It may be a naive question, but why can't one use the 5-probe method and eliminate the longitudinal component?

The contamination of longitudinal component in the transverse measurement is indeed a complicated issue. In our measurements, it stems from the imperfect Hall bar shape of the devices due to the invasive contacts. In standard configurations, with out-of-plane magnetic fields, such contamination is easily removed by symmetrizing / antisymmetrizing the data (for R_{xx} and R_{yx}, respectively), however with an in-plane configuration such symmetry-based analysis cannot be performed. We are unfamiliar with the "5-probe" method referenced by the reviewer. If this method is similar to the one from 10.1016/j.measurement.2023.113039, it would be very difficult to implement in our case as it requires taking both edge contacts (which we already do) and "inner" contacts (away from the edges).

3. One more question to the Rxx and Rxy data: Fig. 2k shows a visible asymmetry for the Rxx data but not for Rxy. Fig. 2j and Fig. 2l are the opposite: the Rxy data are asymmetric, whereas the Rxx data are not. I assume that this asymmetry is due to the 2pi-periodic contribution, but why does it show up so unsystematically? The data in Figs. 6-9 of the Supplemental Material are somewhat different and always show the 2pi-component in Rxy but not in Rxx. That looks more systematic.

We heartly thank the reviewer for pointing this out! Due to an analytical error, the Rxx and Ryx of fig. 2.k were mixed up (although fig. 2e and 2f did show the correct data). This has been fixed in the new version of the manuscript. If by asymmetry the reviewer is refering to the difference between the two oscillations, it does indeed reflect the 2π-periodic contribution, probably from an out-of-plane field: the three samples were measured simultaneously, and show similar assymetries. Regardless of its origin, we note that at similar amplitudes, a 2π-periodic component is more likely to be visible in Ryx data than Rxx data, due to the weaker Ryx signal. Regarding Figs 6-9 of the SM: the data comes from the same set of measurements as Figs 3-5 (main), obtained in a VTI, and not from the dilution refrigerator set of measurements as in Fig 2 of the main, which may explain small differences.

4. Most references to the band structure of t-PtBi2 suggest that it features nodal lines in zero field. Magnetic field converts these lines into Weyl nodes. However, at one point in the discussion section, in the very beginning of page 7, one reads "...stronger than the one originating from the zero-field 12 Weyl nodes". What are these 12 nodes, and how are they related to the nodal line? The text in the same paragraph refers to "the additional Weyl nodes" that presumably lie further away from the Fermi level and can be important yet. However, this is also very hard to understand without an graph showing the band structure. I appreciate that the authors are very familiar with the band structure of t-PtBi2, but potential readers are not.

We reported previously in Ref. 8 (10.1021/acs.nanolett.2c04297) that $PtBi_2$ has 12 symmetry-related Weyl nodes close to the Fermi energy (in Ref 19, G3). More recent calculations (cf Ref 19) show the existence of 3 other groups of 12 symmetry-related Weyl nodes further away from E_F (in Ref 19, G4, G7 and G8). These Weyl nodes exist at zero magnetic field, independently from the nodal lines. In addition, we show in Ref 19 that the nodal lines convert into Weyl nodes when a magnetic field is applied and breaks the mirror symmetry. These were the ones refered to as the additional Weyl nodes. We note that the current study does not rely on nor exploits the Weyl nodes originating from the nodal lines, but rather limits itself to the understanding of t-$PtBi_2$ as a Weyl semimetal, which corresponds to the current published state-of-the-art (excluding our complementary study in Ref. 19). We have clarified the discussion section to make this distinction clearer, and have added a new figure (Fig. 1, with the Brillouin zone position of the 12 Weyl nodes closest to E_F and some band structure cuts) to support this. Almost the entire first paragraph of the discussion (starting with "The band structure of t-$PtBi_2$ is quite complexe..." has been modified.

5. The manuscript would benefit from a dedicated Methods section. The information about the samples and the measurement (vector magnet vs. rotator) is scattered around the text and hard to find, especially when one tries to compare it with other publications. On a related note, readers would benefit from a systematic notation of the samples across the different publications. Sample D3 of the present work seems to be different from sample D3 in the APHE preprint, so they should be given different names, especially if the same D1 and D2 were used in both studies.

We appreciate this comment, and have restructured the manuscript to include a dedicated methods section with the relevant information. We named samples in order of importance to the current study (device D1, device D2 etc.), hence why D1 and D2 are shared between the two studies. Sample D3 (41nm) in this manuscript was not used in Ref 19, and inversly sample D3 (320nm) of Ref 19 was not used in this study. In order to avoid any confusion, we have renamed sample D3 (41nm) in this study to D4.

6. It is also unclear along which exact direction the current was applied. Trigonal symmetry implies two nonequivalent in-plane directions, such as [100] and [110]. Which of them was used, and does the choice matter?

The crystal direction in which the current is flowing is unknown, as the crystaline orientations of the samples were not characterized. In principle, the two in-plane directions are expected to be equivalent due to the C_3 symmetry, as the conductivity should be a scalar in this case (unless C_3-breaking strain is present in the sample). We have seen no strong evidence of any anisotropy in a previously studied sample with "star" contact geometry (see Ref 8, 10.1021/acs.nanolett.2c04297). In all our devices, the current flows along the longest direction of the flake, which is likely to be a crystal axis, as that would be a preferential exfoliation direction. Furthermore, the APHE measurements (Ref 19) confirm that it is indeed a crystal axis, as the angular origin of the APHE is linked to the crystal structure.

7. Whereas the samples used in this work are described as "high-quality single crystals of PtBi2", I could not find any examples of their characterization, especially Laue images, in this and in any of the preceding/concurrent manuscripts. As a layered material, trigonal PtBi2 can be tricky, with stacking faults and even intergrowths of the different polymorphs. Are these effects fully excluded?

The single crystal characterization can be found in 10.1103/PhysRevMaterials.4.124202 (Ref 18). We note that the sentence quoted by the reviewer refers to the single crystals themselves, not the exfoliated samples that are studied in this manuscript. From our side, the crystal quality of a sample is often evaluated by the residual resistance ratio $RRR = \frac{R(300K)}{R(4K)}$, which gives a measure of the impact of crystal disorder on. In ref 8 (supplementary), we showed that our single crystals can reach $RRR \sim 130$, which is a sign of good crystal quality. ARPES measurements [2], which were taken on cleaved single crystals, also indicate good crystal quality. Exfoliated flakes generally show lower RRR than single crystals (between 5 and 14 for the three samples show in this manuscript).
It is difficult to exclude every possibility of intergrowths of polymorphs or stacking faults. We note that, when exfoliating a crystal, the exfoliation will usually happen preferentially along flaws in the crystal structure, including stacking faults and polycrystaline inclusions, making exfoliated flakes more likely to be single crystaline. Furthermore, no evidence for such flaws were seen with atomic force microscopy (AFM) nor with scanning electron microscopy (SEM). The cubic $PtBi_2$ polymorph is not van der Waals, and should therefore be very visible in exfoliated flakes. Furthermore, the study of the APHE of Ref. 19 suggests that, if these defaults exist, they should not have too strong an impact on the resistance. Indeed, both the hexagonal and the cubic polymorphs of $PtBi_2$ (the most likely ones) have C_2 symmetry, and should therefore not show any APHE. The strength of the signal shown in Ref. 19 is therefore a testament to the majority of the flakes (in volume) being trigonal. Moreover, stacking faults which result in a rotation of the planes with respect to the stacking axis would also be visible in the APHE data: if the sample is made of e.g. 2 orientations (top and bottom, separated by a stacking fault), the APHE should show two sets of $2\pi/3$-periodic oscillations, with different angular origins, which is not observed (if the number of faults was large and the relative angular shift at each fault was random, the C_3 symmetry would be effectively lost and the APHE signal would average out).

8. A significant overlap in the figures between this manuscript and the APHE preprint (arXiv:2410.02353) is undesirable: Fig. 1 (PHE) vs. Fig. 1b (APHE), Fig. 3a (PHE) vs. Fig. 2a (APHE), Fig. 4b (PHE) vs. Fig. 2c (APHE), etc. I could not really understand the scientific logic for splitting the analysis of the same data into two separate manuscripts. In my opinion, it makes the data less accessible and imposes unnecessary difficulties on readers who will have to search through two papers if they are interested in details of transport measurements on t-PtBi2. Since SciPost Physics is a flagship journal, I think that its publications should not have such a strong overlap with other journals' publications in terms of both graphics and content. On the other hand, an adequate merger of the PHE and APHE manuscripts should be very welcome in SciPost Physics.

Both manuscript indeed share the same raw data, although they are analyzed in different ways (as pointed out in our reponse to the first comment, and as now included in the introduction section). Since the two APHE and PHE are distinct effects, requiring different analyses, and do not share a common origin, we decided to publish a separate study for each effect to avoid confusion between them and in our analyses. Analysing both effects independently is possible as there is a difference of about two orders of magnitude in amplitude between the two effects. While the raw data is presented in a similar fashion between the two manuscripts (to make comparison easier), no graphs are shared between the main texts. The shared graphs in the SM (which were only the presentation of the raw data at each field/temperature) have been cut out of the present version of the manuscript. Fig. 1 (PHE) was not modified, as it is a simple schematic showing the PHE as expected in experiments (as a π-periodic oscillation), and it cannot be modified to be fundamentally different from the corresponding part of Fig. 1b (APHE), apart from simple cosmetic changes.

REPORT #3

We thank the reviewer for their comments, which gave us the opportunity to improve our manuscript. Below, we provide a point-by-point response to their questions. We implemented multiple changes to the manuscript in order to comply with the modifications requested, by adding more information about the samples (in methods and SM), showing the measurements from additional contacts (in SM), providing a more detailed discussion of the Weyl nodes to the PHE, and clarifying the link between this study and Reference 19 (in the introduction). Modifications to the main text are highlighted in blue.

Report Veyrat et al. report on electrical transport in nanostructures of bulk t-$PtBi_2$, a Weyl semimetal with multiple Weyl points and potential topological nodal lines in its band structure. The key finding is a planar Hall effect (PHE) linked to the topological nature of its electronic structure. At this point, only few semimetals hosting line nodes have been identified. Hence, the results of this work are of high importance for a better understanding of the fundamental behavior in such materials. I therefore would lean toward publishing this work after a revision.

Questions/remarks:

How do the nanostructured samples compare to the bulk samples? Does the RRR vary for the investigated samples? How was the quality of these samples confirmed?

We have not yet investigated the planar Hall effect in bulk crystals, and so cannot comment on this. In a previous study (Ref. 8, 10.1021/acs.nanolett.2c04297, in the SM), we reported on magnetotransport measurements of bulk crystals with a $RRR \sim 130$. The RRR for the three samples presented in the present manuscript are also reported, as 8.7 for D1 (70nm), 13.6 for D2 (126nm) and 4.7 for D3 (41nm). The lower RRR for exfoliated crystals than the bulk counterpart is common in such studies (see e.g. 10.1063/5.0137604 on nanostructures and 10.1038/s41467-018-05730-3 on single crystals, which show the general reduction in RRR with decreasing thickness). The amplitude of the magnetoresistance (with out-of-plane field) of the exfoliated structures is also lower than for bulk crystal, which might be due to an apparent correlation between high RRR and high magnetoresistance (as can be seen by comparing the different reports in the litterature).
The quality of the crystals was studied in greater details in 10.1103/PhysRevMaterials.4.124202 (Ref 18). In this present manuscript, the claim of "high quality" pertained to the crystals (i.e. bulk samples) from which the samples were exfoliated.
More details was added about the samples in the new Methods section. AFM images of the samples were added to the supplementary materials.

Have the authors considered high-mobility effects such as current jetting as mentioned in the discussion section? In other words, would the aspect ratio of the measured samples be sufficient for ruling out such effects. One way to check is the local homogeneity of the current distribution. With their multi terminal devices it would be easy to show that for example in sample D2 all 6 Hall pairs exhibit the same magnetoresistance as well as Hall signals. What are the mobilities and charge carrier densities in this material?

Due to the large number of bands involved in transport, it has proven difficult to estimate the mobilities and densities of the cariers in this material. More details on the transport characterization can be found in our previous report on this material (Ref. 8, 10.1021/acs.nanolett.2c04297), expecially in the SM. For sample D2, multiple pairs of contacts were investigated, and they all show the same hallmarks of PHE. Data was shown for the pair showing the least amount of noise. In all studied nanostructures, the current source and drain were designed very wide to specifically to reduce issues related to possible current jetting. In any case, the magnetoresistance measured in our samples is too small to induce current jetting effects. Furthermore, we note that due to the invasiveness of the contacts, it is not possible to quantitatively compare the different transverse signals (see 10.1039/d0nr04402d, Ref. 20)
Two additional figures were added to the supplementary materials showing the field and temperature measurements for all four pairs of longitudinal contacts, and both pairs of transverse contacts, measured on $D2$. All pairs of contacts show the exact same dependence, confirming that current jetting cannot be the sole origin of our signal.

It is not immediately clear how the observed transport signature would be related to the band structure. An illustration of the parts of the FS responsible for the observations and a description of the theory behind that would be helpful for a better understanding.

In Weyl semimetals, the planar Hall effect appears as a result of Berry curvature effects, and is therefore directly linked to the Weyl nodes (which are sources / sinks of Berry curvature). The simple case was studied by Burkov (Ref 2, 10.1103/PhysRevB.96.041110) and Nandy et al. (Ref 3, 10.1103/PhysRevLett.119.176804), and can be seen as a direct consequence of the chiral anomaly (the transfer of chiral charges between Weyl nodes of opposite chiralities under colinear electric and magnetic fields). In our case however, with numerous Weyl nodes sitting at different energies, it is extremely difficult to quantitatively link the PHE to particular Weyl nodes rather than to others (except between zero-field and field-generated Weyl nodes), which is why we avoid making any particular claims in the manuscript.
We have enhanced the discussion section to add further comments on the relative contribution of the different Weyl nodes: As the strength of the PHE grows with the square of the Fermi velocity of the Weyl cones, one could speculate that, at least in the weak field limit, the zero-field Weyl nodes should give a stronger contribution to the PHE than the Weyl nodes due to conversion of nodal lines, as one could naively expect the Fermi velocity of these field-generated nodes to be roughly proportional to the magnetic field to recover line degeneracies in the $B \to 0$ limit. Therefore, the field-generated Weyl nodes might not contribute much to the PHE. Further research is therefore needed to determine the relative contribution of the different Weyl nodes in this system.
We have also added a new figure (Figure 1) showing the position of the 12 Weyl nodes closest to the Fermi level, and some band structure cuts.

In a previous manuscript (Reference 19) the authors show some of the data presented in this work. Is the newer manuscript a continuation/follow-up/ replacement of the previous work?

This question is discussed at greater lengths in the answer to Report #2. While the present manuscript uses the same raw data as Ref 19, in the latter the PHE was actually treated as a background and removed in the analysis to uncover the anomalous PHE (which is the subject of Ref 19). The two papers are therefore complementary, as they study two simultaneous yet distinct effects in the same samples. The study was separated into two manuscripts as the two effects have different origins and require different analysis. The link between the two manuscripts was made clear in the revised version of the manuscript, at the end of the introduction: We note that, in a complementary study based on the same raw data[1], we evidence and thourougly analysed another effect known as the anomalous PHE (APHE). This effect, which is two orders of magnitude weaker PHE than the PHE, does not originate from Weyl nodes already existing without external magnetic field, but rather from a conversion of topological nodal lines (which we also predict in that separate study) into very distant Weyl nodes (in k-space). Because of their different origin and different analysis required for both signals, as well as the fact the APHE constitutes but a small perturbation compared to the PHE signal, the two effects are studied separately in each study and the latter is simply ignored in the present study.

Requested change
Improve the experimental details about the samples.
Provide more evidence about the quality of the samples.
Include more evidence that would prove homogeneous current distribution throughout the samples.
Improve on the theoretical description that links the observed PHE to the electronic structure of the material.
Provide a detailed explanation about the relevance of this work as compared to Reference 19.

[1] A. Veyrat, K. Koepernik, L. Veyrat, G. Shipunov, S. Aswartham, J. Qu, A. Kumar, M. Ceccardi, F. Caglieris, N. P. Rodríguez, R. Giraud, B. Büchner, J. van den Brink, C. Ortix, and J. Dufouleur, Dissipationless transport signature of topological nodal lines (2024), arXiv:2410.02353.
[2] A. Kuibarov, O. Suvorov, R. Vocaturo, A. Fedorov, R. Lou, L. Merkwitz, V. Voroshnin, J. I. Facio, K. Koepernik, A. Yaresko, G. Shipunov, S. Aswartham, J. V. D. Brink, B. Büchner, and S. Borisenko, Nature **626**, 294 (2024).