

Monolayer van der Waals kagome lattice for high-temperature flat band physics

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This manuscript has been previously reviewed at another journal. This document only contains information relating to versions considered at Nature Physics.

Version 0:

Reviewer comments:

Reviewer #2

(Remarks to the Author)

The authors have substantially addressed my main concern by adding gate-dependent nano-ARPES and gate-dependent transport measurements. These new results directly demonstrate that the kagome flat band can be tuned to the Fermi level and that the partially filled flat-band regime can be accessed experimentally. This significantly strengthens the manuscript beyond the original ARPES and DFT/theory results. The additional comparisons with other flat-band platforms and the polarization-dependent ARPES measurements also satisfactorily address my technical comments.

My remaining concern is that the newly observed insulating state at partial filling should be described cautiously. The present transport data suggest possible correlation effects, but they do not yet establish the expected novel phases such as a fractional quantum anomalous Hall state. I therefore recommend that the authors avoid overclaiming and clearly distinguish experimentally demonstrated tunability/partial filling from theoretically proposed FQAH physics. Subject to this moderation of the claims, I would recommend the publication of this paper in Nature Physics.

Reviewer #3

(Remarks to the Author)

The authors have revised the manuscript and included ARPES and transport measurements of gated devices. The new data show that it is possible to move the flat band up to the Fermi level and observe metallic behaviour. Still there seems to be a substantial distance between the prediction of interesting correlated phases at partial filling of the flat band in Figure 5 and these first measurements. In particular I found strange that there is little discussion to what extent the insulating state observed upon hole doping relates to the phase diagram in Figure 5 and the filling of the band. Beyond the characterisation of the electronic structure of this family of materials, it is the observation of this insulating state that will be of immediate interest to the research community.

There seems to be an inconsistency in the transport data presented in Figure 4 panels (d) and (e).

In panel (d), at $V_{BG} = -90$ V and $T = 70$ K, the resistance is labelled as "Flat band partially filled (Metalled)" for all values of the top gate V_{TG} . In panel (e) the corresponding horizontal cut at $T = 70$ K would suggest that the system is still in an insulating state.

Version 1:

Reviewer comments:

Reviewer #2

(Remarks to the Author)

The authors have satisfactorily addressed all my comments. I therefore recommend this paper for publication in Nature

Physics.

Reviewer #3

(Remarks to the Author)

The authors have addressed the concerns of both referees. At this point, it remains unclear to what extent the insulating state relates to the theoretical predictions. Hopefully, this system will get enough attention to show in the near future whether it is a suitable platform for a FQAH state at high temperature.

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Response to the Report of Reviewer #2:

General remarks: The authors have substantially addressed my main concern by adding gate-dependent nano-ARPES and gate-dependent transport measurements. These new results directly demonstrate that the kagome flat band can be tuned to the Fermi level and that the partially filled flat-band regime can be accessed experimentally. This significantly strengthens the manuscript beyond the original ARPES and DFT/theory results. The additional comparisons with other flat-band platforms and the polarization-dependent ARPES measurements also satisfactorily address my technical comments.

My remaining concern is that the newly observed insulating state at partial filling should be described cautiously. The present transport data suggest possible correlation effects, but they do not yet establish the expected novel phases such as a fractional quantum anomalous Hall state. I therefore recommend that the authors avoid overclaiming and clearly distinguish experimentally demonstrated tunability/partial filling from theoretically proposed FQAH physics. Subject to this moderation of the claims, I would recommend the publication of this paper in *Nature Physics*.

Our reply: We thank the reviewer for finding our revision satisfactory and for recommending the publication of our manuscript in *Nature Physics*.

We fully agree with the reviewer's final comment: While the newly observed insulating state at partial filling points to the onset of correlation effects, its precise nature and connection to novel phases in the theoretical kagome phase diagram – such as the FQAH phase – should be discussed with appropriate caution. We have directly addressed the reviewer's suggestion by making the following modifications in the revised manuscript:

- 1) We added two new sentences in Line 272 and Line 278 explicitly noting that identifying the microscopic nature of the observed insulating state will require further investigations using complementary probes, such as optical and tunneling spectroscopies.
- 2) In Line 282, we added a transition sentence between the experimental and theoretical sections to more clearly separate the experimental observations from the theoretical prediction, as suggested by the reviewer.
- 3) While the prediction of high-temperature FQAH state in the 2D kagome flat band is an important aspect of our manuscript, we agree with the reviewer that placing excessive emphasis on this particular phase could be misleading. We therefore moderated our discussion of the potential FQAH phase by:
 - modifying a statement regarding fractional ground states in the introduction (Line 88).
 - removing a prospective paragraph on the FQAH phase from the discussion (Line 313).

We thank the reviewer again for the constructive and insightful feedback provided throughout the review process.

Response to the Report of Reviewer #3:

General remarks: The authors have revised the manuscript and included ARPES and transport measurements of gated devices. The new data show that it is possible to move the flat band up to the Fermi level and observe metallic behaviour. Still there seems to be a substantial distance between the prediction of interesting correlated phases at partial filling of the flat band in Figure 5 and these first measurements. In particular I found it strange that there is little discussion to what extent the insulating state observed upon hole doping relates to the phase diagram in Figure 5 and the filling of the band. Beyond the characterisation of the electronic structure of this family of materials, it is the observation of this insulating state that will be of immediate interest to the research community.

Our reply: We thank the reviewer for acknowledging the new gate-dependent measurements added in the revised manuscript. These new data – directly motivated by the reviewer’s previous comments – successfully confirm experimental access to the partially filled flat band regime and enable the observation of a new insulating state.

At the same time, the reviewer is fully correct that the connection between the newly observed insulating state and specific phases in the theoretical kagome phase diagram is not firmly established in the present study. While it is certainly possible that the onset of correlation effects evidenced by our transport data is related to one of the correlated phases predicted for kagome flat bands, such as flat band ferromagnetism, we believe identifying the microscopic nature of this state will require dedicated optical and/or tunneling spectroscopy investigations, as has been the case for Moiré-based flat band systems. As such studies lie beyond the scope of the present work, we choose to remain cautious in our interpretation and refrain from attributing the observed insulating state to any specific correlated phase yet.

In response to the reviewer’s comment, we explicitly clarified these points in the revised manuscript as follows:

- 1) In Line 272, 278, and 282, we added three new sentences to explicitly note that, while our work demonstrates experimental access to flat band physics in a 2D kagome system, exploring the full landscape of correlated phases will require further experimental efforts. In particular, we noted that complementary investigations using optical and tunneling spectroscopies will be necessary to fully understand the microscopic nature of the observed insulating state.

Nevertheless, we fully agree with the reviewer that the observation of an insulating state within the partially filled kagome flat band is likely to generate immediate interest to the research community. We thank the reviewer for acknowledging the significance of this result.

Comment 1: There seems to be an inconsistency in the transport data presented in Figure 4 panels (d) and (e). In panel (d), at $V_{BG} = -90$ V and $T = 70$ K, the resistance is labelled as “Flat band partially filled (Metallized)” for all values of the top gate V_{TG} . In panel (e) the corresponding horizontal cut at $T = 70$ K would suggest that the system is still in an insulating state.

Our reply: We very much thank the reviewer for bringing this point to our attention. We agree with the reviewer that the label “Flat band partially filled (metallized)” in Fig. 4d was indeed misleading, as not all of the partially filled regime is metallic, as correctly noted by the reviewer.

Specifically, even though the flat band becomes partially filled, in the very low doping limit, transport may still exhibit insulating temperature dependence due to disorder-dominated conduction. Only upon sufficient doping to screen impurity potentials, the system begins to exhibit true metallic transport.

As the onset of flat band filling does not necessarily coincide with the onset of metallic transport, the label “Flat band partially filled (metallized)” mentioned by the reviewer indeed raises confusion. We have therefore removed the term “(metallized)” from the Fig. 4d and also modified the caption accordingly (Line 711 and Line 716).

We thank the reviewer again for this helpful comment, which has improved the clarity and accuracy of the presentation.