nature portfolio

Peer Review File

Evidence for single variant in altermagnetic $RuO_2(101)$ thin films

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This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The authors reported the growth of RuO_2, a material with significant controversy regarding the possibility of altermagnetism, into a single-variant thin film. Securing a thin film of altermagnet with large domains is considered a critical step in exploring potential applications of altermagnetic materials. It is expected that if the authors can present a method to secure single-domain altermagnetic RuO_2 thin films, it would meet the standards of Nature Communications. However, based solely on the current version of the manuscript's data, it is difficult to believe that a single-variant altermagnetic RuO_2 thin film has been successfully achieved, making it challenging to recommend the manuscript for publication in Nature Communications.

The (101) and (-101) lattice planes of RuO_2 are equivalent and share the same lattice parameters, making it experimentally challenging to distinguish between the two. The manuscript suggests the possibility of a single domain based on TEM results, XMLD, and spin-splitting magnetoresistance, but these experimental evidences are not conclusive enough to draw a definitive conclusion.

1.TEM is a local probe that shows only a small part of the sample. Although a wide-area image was presented in the supplementary information, this is still within a region smaller than 100 nm. This data may serve as evidence that the sample has domain sizes larger than 100 nm, but it is insufficient as evidence of a single-variant film.

2.The mechanism for growing single-variant domains involves the epitaxial relationship with the Al_2O_3 substrate, but it is questionable whether this epitaxial relationship is well-applied to the sample. First, the difference between (101) and (-101) domains is the position of the oxygen atoms, as mentioned in the paper (Figure 2). It is uncertain whether the Al_2O_3 surface has a termination like that shown in Figure 2e. According to the mechanism proposed by the authors, if the Al_2O_3 is terminated with Al and O (z = -0.5) atoms, the sample should exhibit the (-101) domain. Without a method to process the Al_2O_3 surface as a single termination (Al & O (z = 0.5)), the domains will naturally exist in a 50:50 ratio. Moreover, the interface in the sample's TEM image is not sharp, making it hard to expect a clear epitaxial relationship at such an interface.

3. For XMLD and spin-splitting magnetoresistance, if the domain size is larger than the beam spot or device size, results similar to those of a single domain can be observed. However, these results still cannot serve as evidence that the entire sample is a single-variant thin film.

Reviewer #2

(Remarks to the Author)
In this study, the authors have successfully fabricated altermagnet $RuO_2(101)$ epitaxial thin films on $Al_2O_3(1\bar{1}02)$ r-plane substrates. They provide evidence for the realization of a single variant in the $RuO_2(101)$ films through a combination of advanced structural analyses, including X-ray diffraction (XRD), atomic-resolution scanning transmission electron microscopy (STEM), and X-ray magnetic linear dichroism (XMLD). Their XMLD results suggest the presence of a finite quadrupole spin moment, which is in qualitative agreement with Density Functional Theory (DFT) calculations. In addition, they report on unidirectional magnetoresistance in $Al_2O_3(1\bar{1}02)/RuO_2(101)$/CoFeB trilayer structures, attributing this effect to directional spin splitting and the resulting anisotropic spin current.

While I find the results to be interesting and potentially important, I cannot recommend the manuscript in its present form for publication in Nature Communications. Several points need to be clarified/addressed, as listed below.

- The authors attribute the formation of the single-variant $RuO_2(101)$ to the alignment of oxygen atoms at the interface with $Al_2O_3(1\bar{1}02)$ substrates based on their STEM observations. While this explanation is plausible, it would benefit from a deeper theoretical understanding of how the substrate lifts the degeneracy of the two variants, energetically favoring one over the other. For instance, further discussion on the role of substrate-induced strain in selecting the $RuO_2(101)$ variant over its mirror would strengthen this finding. This could be approached theoretically by performing strain-modified DFT calculations. The current DFT calculations focus on supporting the XMLD findings related to magnetic anisotropy. It would be beneficial to extend these calculations to include strain effects, as this could provide a comprehensive understanding of strain impacts on bond angles and lengths and could provide insight into variant selection mechanisms.

- Please clarify why ± signs appear before the spin-conserving term which involves only orbital angular momentum operators without directly involving spin.

- The authors demonstrate single-variant $RuO_2(101)$ formation. However, it remains unclear if the reverse orientation, $RuO_2(\bar{1}01)$, can also be selectively grown. Comments on whether or how to achieve this variant would be insightful, as it would relate to understanding what fundamentally lifts the degeneracy between variants.

Minor Suggestions:

- In Equation (1), clarify the notation for “l” in E_{MAE}^{l}, Δ_{ex}^l, and ξ_{l}, as well as the notation for expectation values ⟨ ... ⟩. Specify whether these refer to specific states.

- Spell out “XRD” as X-ray Diffraction when first mentioned.

- The term “unidirectional spin-splitting magnetoresistance (USSMR)” may be somewhat misleading. What is unidirectional in the ferromagnet/altermagnet bilayer is the magnetoresistance, rather than the spin splitting.

Reviewer #3

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #4

(Remarks to the Author)
I read the manuscript by He et al. with interest. However, I have doubts about the conclusions of this work. In my opinion, these doubts should be resolved before one can proceed further.

According to simple models, RuO2 is an altermagnet. These models assume antiferromagnetic arrangements of local moments on Ru sites. To obtain altermagnetic phase numerically in one-electron "ab-initio" calculations, the GGA+U method needs to employed. However, according to one paper [PRB 109, 134424 (2024)], the magnitude of the U-term that leads to magnetic order is higher than the value that describes other electronic properties of RuO2. Furthermore, two muon-spin-rotation studies have found that RuO2 is non-magnetic [PRL 132, 166702 (2024), arXiv:2405.10820 (2024)]. I believe these very recent studies have changed the perception of RuO2, and as a consequence they call for readjustment of the goals of experimental studies on RuO2. These goals have now been shifted from investigating altermagnetic properties of RuO2 into providing the evidence that there exist antiferromagnetic RuO2 samples. Importantly, if RuO2 is antiferromagnetic, it is almost certainly altermagnetic.

He et al. claim to provide the evidence of antiferromagnetism in RuO2 though the XMLD measurements at the Ru M-edge. In a simple model, this means that dipole transitions from the Ru 3p doublet into Ru 4d states at the Fermi level occur (in a crude approximation Ru 5s can be neglected). Optical matrix elements are not directly sensitive to spin, and the spin sensitivity of either XMCD or XMLD is a result of spin-orbit coupling, although the mechanisms are somewhat different in the two methods. Importantly, the measured current results from the Auger cascades and can be assumed fully angle-integrated. To the first approximation an non-vanishing XMLD effect is related to the orientation of the polarization of the light with respect to the orientation of the Ru orbitals at the Fermi level. Ignoring itinerant properties, these Fermi level Ru 4d orbitals can be projected onto the real orbital basis (dz2, dxy, etc), and besides the contribution from the crystal bonds, the coefficients of that decomposition will slightly depend on the orientation of the Neel vector due to spin orbit coupling, potentially leading to magnetic sensitivity of XMLD. (Since Ru 5s are ignored, one does not even have to worry about radial integrals.) XMLD has been used to image antiferromagnetic domains in the PEEM mode, see e.g. Nature Communications 13, 724 (2022) -- but these results are much easier to be correctly interpreted because they are based on the contrast between different areas and on visual characteristic shapes of the domains.

At least three conditions need to be satisfied in order to consider XMLD results of He et al. as a probe of antiferromagnetism in RuO2:

A. The single variant phase of the crystal should be larger than the photon beam spot
B. Magnetic domains should all have the same orientation of the Neel vector
C. XMLD effects non related to magnetism must be eliminated

I think none of the conditions have been fully satisfied in the study by He et al. Correct me if I am wrong, but concerning point A, the largest demonstrated size of the single variant phase is the TEM image in Fig. S4, the size is approx. 100 nm. Therefore, following the XRD results, possible existence of (-101) phase cannot be eliminated in XMLD, where photon beam has been certainly large. Concerning point B, the goal of XMLD results is prove the existence of magnetic order, but B would impose an assumption of one type of magnetic order with moments along [001] -- results are supposed to prove what has been assumed, a logical issue. Also, if Neel vector is oriented along few discrete directions, there might exist geometries where XMLD could be sensitive to magnetic order, but this would need to be analyzed in detail.

Concerning point C, the (101) surface studied by He et al. does not seem to retain much of the symmetry of the bulk crystal. The analysis of the results from Fig. 3 should start with establishing the symmetries of the (101) surface and their relation to symmetries of XMLD signals. I doubt XMLD can be treated in the bulk limit. Furthermore, an unlikely coincidence may lead to thin films having some residual anisotropy due to substrate terraces, deposition angle etc. -- but probably this is not the case here.

Once the surface symmetry is understood, one should assume pure (101) surface or a combination of (101) and (-101) and see which light incidence angle should lead to equivalent XMLD spectra. Perhaps, since domain-from-domain contrast is not available like in XPEEM, one should also consider XMLD process description beyond a simplified single-electron LCAO picture, to see if it introduces additional effects.

My suggestion is that once this is established, and the result points towards the confirmation of magnetic order in these RuO2 films, one can proceed with the analysis of the subsequent part of the paper, conclusions of which are based on the assumption of the existence of the magnetic order.

Independent on this, I consider this paper an extensive experimental and theoretical characterization of a new type of high-quality RuO2 films and I appreciate this work. My review is written from the perspective of whether the manuscript matches the criteria of Nature Communications.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
It appears the authors have carefully considered the previous reviewer's comments. In particular, the detailed XRD experiments seem sufficient to demonstrate that the thin film is a single variant. I appreciate the authors' efforts over the long review period. I believe this paper will be significant for research and applications in RuO_2-based altermagnetism, and I find it meets the standards of Nature Communications. I recommend its publication.

Reviewer #2

(Remarks to the Author)
The authors have addressed most of my previous comments and have improved the manuscript accordingly. Their DFT calculations on the strain effects and surface termination of $Al_2O_3(1\bar{1}02)$ in stabilizing $RuO_2(101)$ over $RuO_2(\bar{1}01)$ remain technically sound, effectively incorporating formation energy analysis and epitaxial strain effects. However, their assumption of a purely oxygen-terminated $Al_2O_3(1\bar{1}02)$ surface may still be idealized, as mixed terminations (coexistence of O- and Al-terminated regions) are possible due to thermodynamic competition, surface reconstructions, and growth kinetics. If such mixed terminations exist, they could affect the relative stability of RuO_2 variants and potentially modify strain effects at the interface. While their results remain reasonable, a brief discussion on the possibility of mixed terminations and their implications would further strengthen their argument.

Regarding magnetoresistance effects, the authors measured R_{2\omega}. However, in principle, the spin splitting effect should also induce angular dependence of R_{\omega} in linear response, where the spin current polarization and the corresponding magnetic field direction associated with the magnetoresistance extrema are largely determined by the Néel vector of the altermagnet/antiferromagnet. Given its linear-response nature, this effect is expected to be more pronounced than its second-order counterpart. This is a worthwhile aspect to investigate further, and the authors may have already measured it using the same experimental setup for R_{2\omega}. Including (or commenting on) the angular dependence of R_{\omega} in their magnetoresistance measurements would provide a more complete picture of spin transport effects, and with these revisions, the manuscript would be in better shape for publication.

Reviewer #3

(Remarks to the Author)

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #4

(Remarks to the Author)
I would like to thank the Authors for their response and for including additional temperature-dependent XLD measurements in the Supplement. Indeed, such temperature dependence of the XLD could be an evidence of magnetic order in RuO2. However, other factors could also influence the XLD, and without ruling them out, I am not fully convinced that the observed changes are directly linked to antiferromagnetism. For example, I am not sure what is the expected temperature-dependence of XLD at 3p edge in a non-magnetic compound containing Ru or another 4d element. Additionally, since it is not established whether in nature RuO2 is antiferromagnetic (I am referring to recent papers on the topic), its Néel temperature also remains uncertain.

I appreciate the Authors' efforts in thoroughly characterizing their films. However, in my view, a clear and direct connection to magnetism has not yet been demonstrated. Consequently, I am unable to recommend the publication of the paper in Nature Communications.

Version 2:

Reviewer comments:

Reviewer #2

(Remarks to the Author)
I noticed that the preprint cited by the authors on spin-splitting–induced linear magnetoresistance has now been published in Advanced Materials (https://doi.org/10.1002/adma.202507764). It would be appropriate to update the citation to the published version in the revised manuscript.

I have also read Reviewer 4's latest comments and understand the concerns raised. That said, the combination of structural characterization, transport measurements, and temperature-dependent XMLD data appears consistent with magnetic ordering in single-variant RuO_2 thin films.

On this basis, I am inclined to recommend the manuscript for publication in Nature Communications.

Reviewer #3

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reviewer #4

(Remarks to the Author)
I do not have new comments for Authors.

Response letter

Dear Editor and Reviewers,

Thank you for your thoughtful consideration of our manuscript and for the opportunity to submit a major revision to Nature Communications. We appreciate the detailed and constructive feedback provided by the editor and the reviewers, which has significantly helped us to improve the manuscript.

We acknowledge the importance of addressing the issues raised, particularly those concerning the evidence for single-variant epitaxial growth, the implications of potential multi-domain structures for XMLD measurements, and the exclusion of other possible sources of XMLD signals. These are indeed critical points to establish the validity and significance of our findings.

We have already performed substantial additional experimental work to address these concerns:

1. Additional XRD measurements:

As suggested, we performed **additional in-plane scan XRD measurements** with a suitably large size (incident slit of 10 mm) to comprehensively probe the entire sample area. **A single peak feature was observed in the in-plane XRD scan for the RuO_2(002), (10$\bar{1}$) and (200) lattice planes.** Details are shown in the replies to Reviewer #1 below. These XRD results confirm the presence of a single-variant RuO_2(101) film, addressing the primary concern about the epitaxial growth of the films.

2. Symmetry breaking between (101) and the inverse orientation:

By carrying out **additional first-principles calculations**, we have carefully analyzed and discussed the mechanisms, i.e., **formation energies**, that break the symmetry between the (101) orientation and its inverse. These considerations have been incorporated into the revised manuscript and supplementary materials to address the insightful comment raised by Reviewer #2.

3. Assessment of XMLD and domain structures, and exclusion of other XMLD signal sources:

Our new XRD results has verified the epitaxial growth of the single crystal domain/variant RuO_2(101) film. Based on the single crystal domain, we agree with Reviewer #4 mentioned that magnetic domains should all have the same orientation of the Néel vector. We performed the **angular-dependent XMLD measurements** to determine the Néel vector direction. The existence of uniform magnetic domains (all have the same orientation of the Néel vector) can be understood by the formation of the structural single variant. To address the concerns of other XMLD signal sources raised by Reviewer #4, we conducted additional analysis to exclude the structural contributions as X-ray linear dichroism (XLD) from the XMLD signals by **temperature-dependent measurements**, which has strengthened the interpretation of the results and reinforced the evidence for antiferromagnetic order.

We hope that the additional experimental data and revised manuscript comprehensively address the reviewers' concerns. We have prepared a detailed point-by-point response to the reviewers' comments along with the revised manuscript and supplementary materials, highlighting all the changes made in response to their comments/suggestions. If you have any additional comments or specific suggestions, we would be glad to address them as well. We sincerely appreciate your support and guidance throughout the review process.

The response to each comment is given in this letter. Furthermore, the corresponding changes in the main text have been marked by a highlight color in the revised manuscript and supplementary materials. We hope all the concerns from the reviewers can be relieved by our responses, and we are grateful if the revised manuscript can be considered for publication.

Note: Our replies are in blue fonts in the following sections.

Reviewer #1 (Remarks to the Author):

The authors reported the growth of RuO_2, a material with significant controversy regarding the possibility of altermagnetism, into a single-variant thin film. Securing a thin film of altermagnet with large domains is considered a critical step in exploring potential applications of altermagnetic materials. It is expected that if the authors can present a method to secure single-domain altermagnetic RuO_2 thin films, it would meet the standards of Nature Communications. However, based solely on the current version of the manuscript's data, it is difficult to believe that a single-variant altermagnetic RuO_2 thin film has been successfully achieved, making it challenging to recommend the manuscript for publication in Nature Communications.

The (101) and (-101) lattice planes of RuO_2 are equivalent and share the same lattice parameters, making it experimentally challenging to distinguish between the two. The manuscript suggests the possibility of a single domain based on TEM results, XMLD, and spin-splitting magnetoresistance, but these experimental evidences are not conclusive enough to draw a definitive conclusion.

Reply: We sincerely appreciate the reviewer's detailed comments and recognition of the importance of single-variant RuO_2 thin films in advancing altermagnetic materials research. We have carefully considered the reviewer's concerns and have added new experimental and theoretical calculation results to address them in the revised manuscript.

1.TEM is a local probe that shows only a small part of the sample. Although a wide-area image was presented in the supplementary information, this is still within a region smaller than 100 nm. This data may serve as evidence that the sample has domain sizes larger than 100 nm, but it is insufficient as evidence of a single-variant film.

Reply: We acknowledge the reviewer's concern regarding the localized nature of TEM analysis. To provide more direct and compelling evidence for the single variant of the RuO_2 thin film, we have conducted additional wide-area characterization using the X-ray diffraction (XRD) method covering

the region of about 10 mm×10 mm, as also suggested by the editor. XRD can probe the entire sample area and provides comprehensive information about the film's crystallographic properties.

The illustrations in the following Figs. R1-R5 show how to detect a single variant of $RuO_2(101)$ using the XRD method with an X-ray incident slit of 10 mm. In the case of a single variant, as shown in Fig. R1**a**, we can obtain the $RuO_2(002)$ plane diffraction peak at $\varphi = 90^o$ by adjusting the angle of X-ray incidence and reflection based on Bragg's Law. When we rotate the sample $\Delta\varphi = 180^o$ in the plane of sample surface, i.e., $\varphi = 270^o$, no diffraction peaks would occur, because the $RuO_2(200)$ plane does not meet the requirement of Bragg diffraction condition here. Thus, by rotating the sample from 0 to 360º, we will only see one diffraction peak. However, in the case of two variants, as shown in Fig. R1**b**, the diffraction peak of the $RuO_2(002)$ lattice plane can be observed at $\varphi = 90^o$. After rotating the sample to $\varphi = 270^o$, a diffraction peak of the $RuO_2(002)$ plane is also detected because of the reverse variant $RuO_2(\bar{1}01)$. Therefore, we can observe two peaks for the case of two variants by rotating the sample one full 360º circle in the plane of the sample surface. Note that in the first version of the manuscript, Figs. 1f and 1g show the two-fold peaks because of the equivalent planes of (211), $(\bar{2}1\bar{1})$ and (111), $(\bar{1}1\bar{1})$ even in the single-variant case.

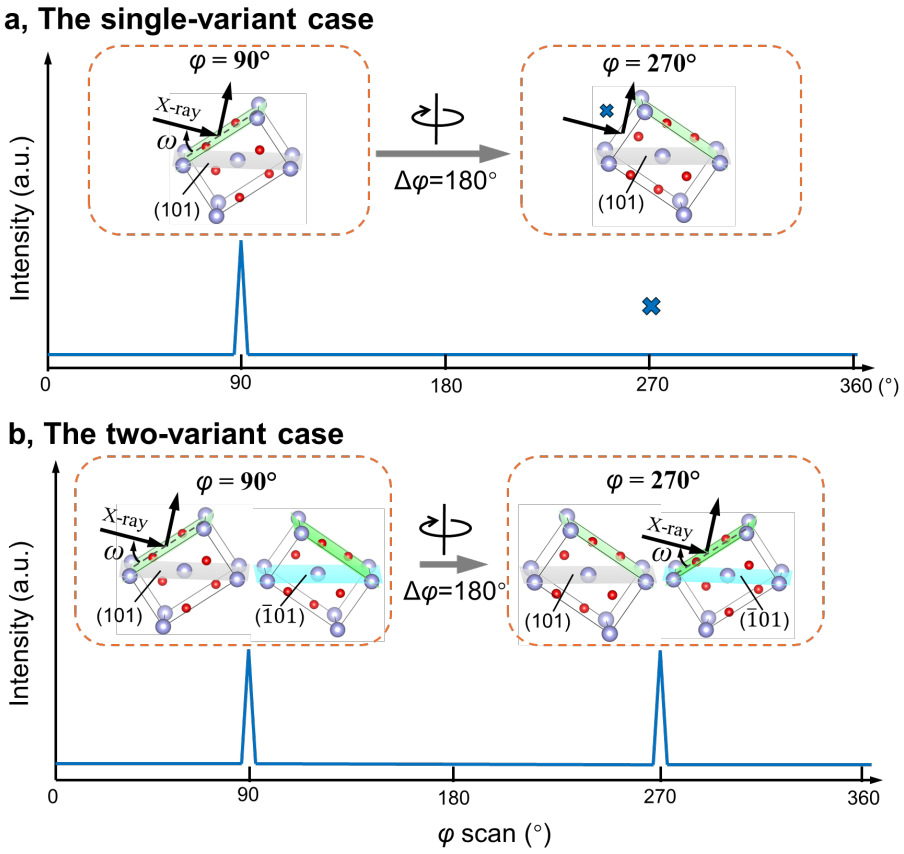

Figure R1. Illustrations of detecting a single variant of RuO_2(101) using the XRD method. **a**, for the single-variant case. **b**, for the two-variant case.

Based on the above description, we have performed detailed XRD measurements for our films. Figure R2 shows an illustration of the setup of XRD measurements. We firstly set up the configuration of X-ray incidence and reflection for RuO_2(002) plane by rotating the sample to $\chi = 34.08°$, $2\theta = 59.62°$, then carried out the in-plane φ scan. The X-ray incident slit used in the measurement is 10 mm in size, which is large enough to obtain information about the crystal structure of the entire thin film. The result of in-plane XRD scan for RuO_2(002) lattice plane of the RuO_2(101) thin film is shown in Fig. R3. A single peak was clearly observed, which clearly indicates a single-variant film. In order

to further confirm this single variant feature, we also performed the in-plane XRD scan measurements for $RuO_2(10\bar{1})$ and (200) lattice planes at $\chi = 68.85^o$, $2\theta = 35.29^o$, and $\chi = 55.28^o$, $2\theta = 40.47^o$, respectively. The XRD patterns are shown in Figs. R4 and R5, respectively. Both cases show a single peak feature. Further, the characteristic single peak was also observed in other samples of $RuO_2(101)$ films deposited at different times. To the best of our knowledge, we think that the above results are sufficient as evidence of a single-variant $RuO_2(101)$ film.

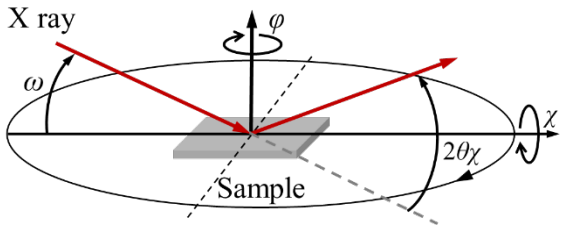

Figure R2. Illustration of the setup of XRD measurements.

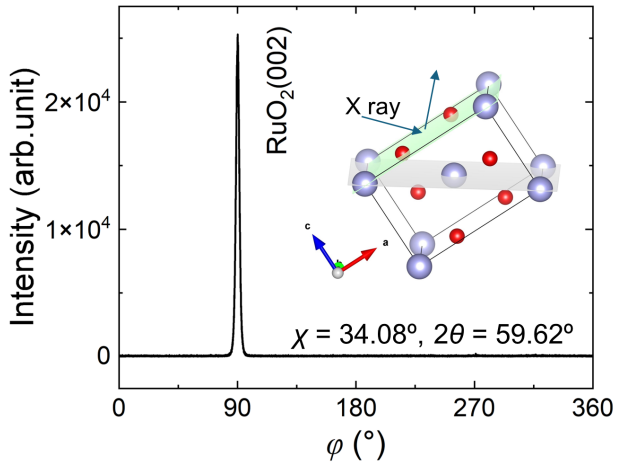

Figure R3. Result of in-plane XRD scan for (002) lattice plane of the $RuO_2(101)$ thin film.

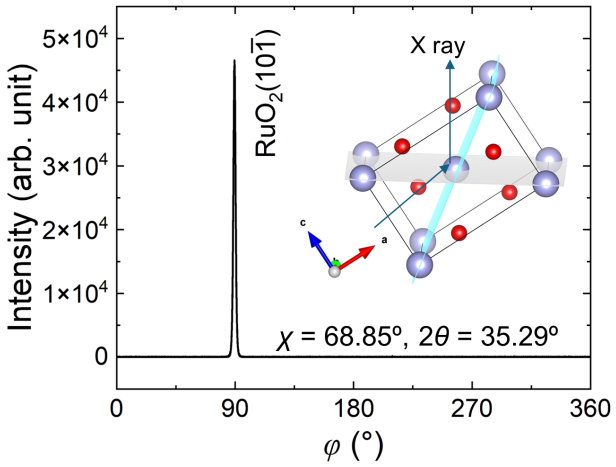

Figure R4. Result of in-plane XRD scan for (10$\bar{1}$) lattice plane of the RuO_2(101) thin film.

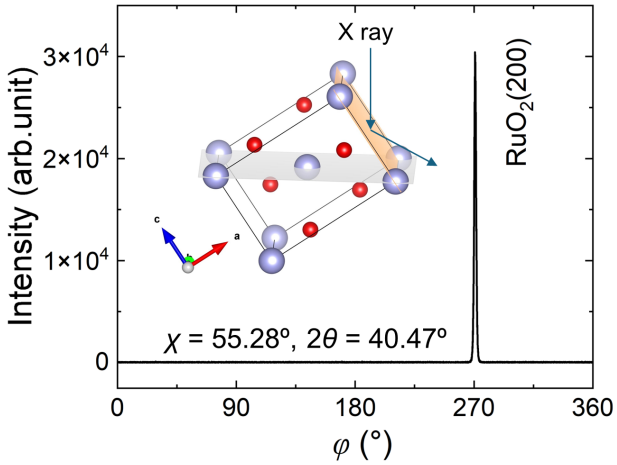

Figure R5. Result of in-plane XRD scan for (200) lattice plane of the RuO_2(101) thin film.

In the revised manuscript, we present the XRD results as new figures (Figs. 1f, 1g, 1h), showing the single diffraction peak nature, which confirm the RuO_2(101) single variant across the entire film. This finding directly supports our claim of achieving the growth of a single-variant RuO_2 thin film, in agreement with the results of TEM, XMLD, and magnetoresistance. The XRD data complements

the localized evidence provided by TEM and strengthens the manuscript. We hope that the reviewer will be satisfied with our new results and explanations.

2.The mechanism for growing single-variant domains involves the epitaxial relationship with the Al_2O_3 substrate, but it is questionable whether this epitaxial relationship is well-applied to the sample. First, the difference between (101) and (-101) domains is the position of the oxygen atoms, as mentioned in the paper (Figure 2). It is uncertain whether the Al_2O_3 surface has a termination like that shown in Figure 2e. According to the mechanism proposed by the authors, if the Al_2O_3 is terminated with Al and O (z = -0.5) atoms, the sample should exhibit the (-101) domain. Without a method to process the Al_2O_3 surface as a single termination (Al & O (z = 0.5)), the domains will naturally exist in a 50:50 ratio. Moreover, the interface in the sample's TEM image is not sharp, making it hard to expect a clear epitaxial relationship at such an interface.

Reply: We thank the reviewer for raising these critical points about the epitaxial relationship between $RuO_2(101)$ and the $Al_2O_3(1\bar{1}02)$ substrate, as well as the termination of the $Al_2O_3(1\bar{1}02)$ surface. Below, we address the concerns in detail.

To clarify the stability of the $Al_2O_3(1\bar{1}02)$ surface, we conducted first-principles density functional theory (DFT) calculations using the VASP-PAW method. We optimized the surface structures of the $Al_2O_3(1\bar{1}02)$ for four different terminations, i.e., Type 1 and Type 2 with O and Al terminations, as shown in Figs. R6a-d. The Type 1 with O termination corresponds to the $Al_2O_3(1\bar{1}02)$ surface terminated with Al and O ($z = +0.5$) atoms while the Type 2 with O termination is for the $Al_2O_3(1\bar{1}02)$ surface terminated with Al and O ($z = -0.5$) atoms. The calculations reveal that the oxygen-terminated Type 1 surface has the lowest surface energy density shown in Fig. R6 and is the most stable termination. In the experiments, we treated the $Al_2O_3(1\bar{1}02)$ substrates in a muffle furnace at 1000°C

for one hour in an atmospheric environment. This preparation step also supports the formation of a stable oxygen-terminated surface.

We further calculated the stability of the $RuO_2(101)$ and $RuO_2(\bar{1}01)$ variants by stacking $RuO_2(101)$ and $RuO_2(\bar{1}01)$ cells on the oxygen-terminated Type-1 $Al_2O_3(1\bar{1}02)$ surface, corresponding to Fig. 2f $Al_2O_3(1\bar{1}02)/RuO_2(101)$ and Fig. 2g $Al_2O_3(1\bar{1}02)/RuO_2(\bar{1}01)$ in the manuscript. In these calculations, a supercell containing 162 atoms (Al, O, and Ru) was used, with 4 monolayers (ML) of RuO_2 modeled as either $RuO_2(101)$ or $RuO_2(\bar{1}01)$. After structural relaxation, the cross-sectional atomic distributions in the two cases are shown in Fig. R7. We found that $Al_2O_3(1\bar{1}02)/RuO_2(101)$ has a lower total formation energy density by 1.2 J/m^2 than that for $Al_2O_3(1\bar{1}02)/RuO_2(\bar{1}01)$. Here, the area density of formation energy is defined as $\gamma_{\text{formation}} = [E_{\text{total}} - (N_{\text{Ru}}\mu_{\text{Ru}} + N_{\text{Al}}\mu_{\text{Al}} + N_{\text{O}}\mu_{\text{O}})]/A$, where N_{Ru}, N_{Al} and, N_{O} are the numbers of Ru, Al, and O atoms, μ_{Ru}, μ_{Al} and μ_{O} are the total energies per atom for bulk hcp-Ru, bulk fcc-Al, and O_2 molecule, and A is the unit cell area of $Al_2O_3(1\bar{1}02)/RuO_2(101)$ interface. This result confirms that the $Al_2O_3(1\bar{1}02)/RuO_2(101)$ is the more favorable configuration, aligning with experimental observations. Also, we agree with the reviewer that the interface in the sample's TEM image is not perfectly sharp as a mono-atomic layer. However, we can find that, across the interface, the atomic terraces of the $Al_2O_3(1\bar{1}02)$ substrate have a height of two layers of Al atoms, which is consistent with the Type 1 oxygen-terminated case (Fig. R6a).

Overall, based on the DFT calculations, experimental TEM observations, and substrate preparation process, we conclude that the $Al_2O_3(1\bar{1}02)$ surface is Type 1 with oxygen-termination, allowing the growth of a single-variant $RuO_2(101)$ thin film. The interface structure and energy calculations further support that $RuO_2(101)$ is a more stable and preferred orientation than $RuO_2(\bar{1}01)$.

These explanations are added in the main manuscript and Supplemental Materials.

We hope that these new results and explanations have addressed the reviewer's concerns regarding the epitaxial relationship and substrate termination.

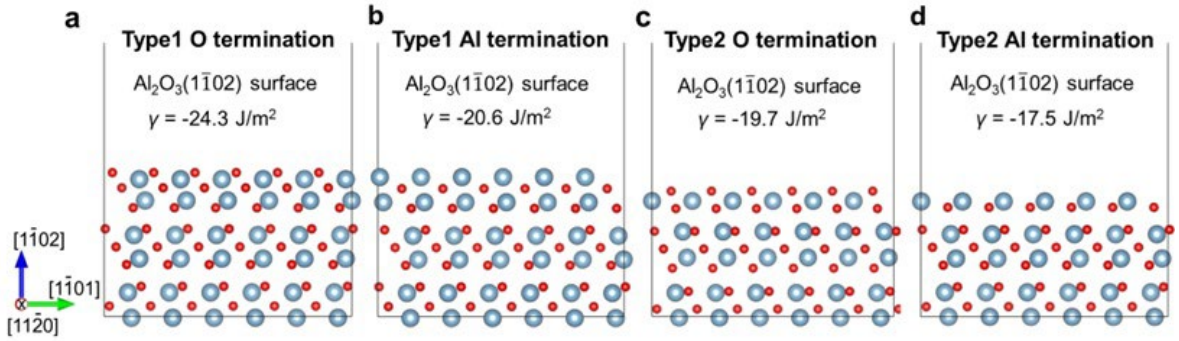

Figure R6. Illustrations of structures of $Al_2O_3(1\bar{1}02)$ surface. **a**, Type 1 with O termination, **b**, Type 1 with Al termination, **c**, Type 2 with O termination, **d**, Type 2 with Al termination. Red and light blue dots represent oxygen and aluminum atoms, respectively. The formation energy density for each case is also shown.

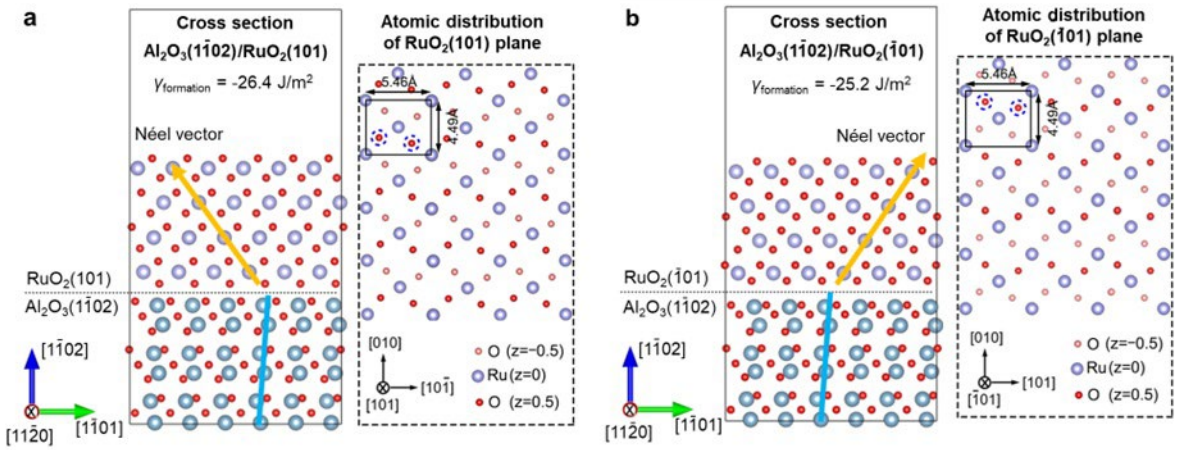

Figure R7. Illustrations of cross-sectional and in-plane crystal structures and atomic distributions of **a**, $Al_2O_3(1\bar{1}02)/RuO_2(101)$ and **b**, $Al_2O_3(1\bar{1}02)/RuO_2(\bar{1}01)$. Gray dots represent Ru atoms. Red and light blue dots are for oxygen and aluminum atoms, respectively. The formation energy density for each case is also shown.

3. For XMLD and spin-splitting magnetoresistance, if the domain size is larger than the beam spot or device size, results similar to those of a single domain can be observed. However, these results still cannot serve as evidence that the entire sample is a single-variant thin film.

Reply: We thank the reviewer for highlighting this important point. The beam spot size in XMLD measurements is approximately the order of 100 μm in diameter and the size of Hall bar device for spin-splitting magnetoresistance measurement is 10 μm × 25 μm. Based on the reviewer's comment, we agree with the reviewer that the localized characteristics of XMLD and spin-splitting magnetoresistance measurements may lead to results resembling those of a single domain even if the entire sample is not a single-variant thin film. To address this concern comprehensively, we have performed additional wide-area XRD in-plane scan measurements. The details are shown in the reply to the above Comment #1.

The XRD method provides a macroscopic evaluation of the sample and allows us to probe the crystallographic properties of the entire film. The results confirm the successful detection of the RuO_2(101) single variant across the entire sample area. This wide-area characterization provides evidence supporting our claim that the film is indeed a single-variant thin film. The XRD results complement the local observations from XMLD and spin-splitting magnetoresistance and address the concern that the localized nature of these measurements might not fully represent the entire sample. We have included these new data and their analysis in the revised manuscript.

We hope this additional evidence has resolved the reviewer's concern and strengthened the validity of our conclusions.

Reviewer #2 (Remarks to the Author):

In this study, the authors have successfully fabricated altermagnet $RuO_2(101)$ epitaxial thin films on $Al_2O_3(1\bar{1}02)$ r-plane substrates. They provide evidence for the realization of a single variant in the $RuO_2(101)$ films through a combination of advanced structural analyses, including X-ray diffraction (XRD), atomic-resolution scanning transmission electron microscopy (STEM), and X-ray magnetic linear dichroism (XMLD). Their XMLD results suggest the presence of a finite quadrupole spin moment, which is in qualitative agreement with Density Functional Theory (DFT) calculations. In addition, they report on unidirectional magnetoresistance in $Al_2O_3(1\bar{1}02)/RuO_2(101)$/CoFeB trilayer structures, attributing this effect to directional spin splitting and the resulting anisotropic spin current. While I find the results to be interesting and potentially important, I cannot recommend the manuscript in its present form for publication in Nature Communications. Several points need to be clarified/addressed, as listed below.

Reply: We thank the reviewer for his/her thoughtful comments and for finding our results interesting and potentially important. We understand that several points need clarification, and we are committed to addressing these concerns comprehensively by providing additional theoretical calculations for a deeper understanding of the substrate-induced selection mechanism. Below, we provide detailed responses to the points raised and outline the revisions made to the manuscript.

- The authors attribute the formation of the single-variant $RuO_2(101)$ to the alignment of oxygen atoms at the interface with $Al_2O_3(1\bar{1}02)$ substrates based on their STEM observations. While this explanation is plausible, it would benefit from a deeper theoretical understanding of how the substrate lifts the degeneracy of the two variants, energetically favoring one over the other. For instance, further discussion on the role of substrate-induced strain in selecting the $RuO_2(101)$ variant over its mirror would strengthen this finding. This could be approached theoretically by performing strain-modified DFT calculations. The current DFT calculations focus on supporting the XMLD findings related to

magnetic anisotropy. It would be beneficial to extend these calculations to include strain effects, as this could provide a comprehensive understanding of strain impacts on bond angles and lengths and could provide insight into variant selection mechanisms.

Reply: We thank the reviewer for their insightful comment. To address this issue, we first performed first-principles DFT calculations using the VASP-PAW method to evaluate the formation energies of different terminations of the $Al_2O_3(1\bar{1}02)$ surface, i.e., Type 1 and Type 2 with O and Al terminations, as shown in Figs. R8a-d. The calculations indicate that the Type-1 oxygen-terminated surface has the lowest formation energy density, making it the most stable termination, consistent with experimental STEM results.

Subsequently, we assessed the stability of $RuO_2(101)$ and $RuO_2(\bar{1}01)$ variants on the $Al_2O_3(1\bar{1}02)$ surface with Type-1 oxygen termination. In these calculations, we modeled 4 monolayers (ML) of RuO_2 with oxygen-terminated interfaces, using a supercell comprising 162 atoms of Al, O, and Ru for both cases. Figures R9a and b show the resulting atomic configurations for both cases. It is found that the formation energy of the $Al_2O_3(1\bar{1}02)/RuO_2(101)$ configuration is 1.2 J/m² lower than that of $Al_2O_3(1\bar{1}02)/RuO_2(\bar{1}01)$, indicating that $RuO_2(101)$ variant is energetically favored. Here, the area density of formation energy is defined as $\gamma_{\text{formation}} = [E_{\text{total}} - (N_{\text{Ru}}\mu_{\text{Ru}} + N_{\text{Al}}\mu_{\text{Al}} + N_{\text{O}}\mu_{\text{O}})]/A$, where N_{Ru}, N_{Al}, and N_{O} are the numbers of Ru, Al, and O atoms, respectively. μ_{Ru}, μ_{Al}, and μ_{O} are the energies for each corresponding atom, and A is the unit cell area of $Al_2O_3(1\bar{1}02)/RuO_2(101)$. This result confirms that the degeneracy between $RuO_2(101)$ and $RuO_2(\bar{1}01)$ is lifted at the interface, with $RuO_2(101)$ being more energetically stable.

Regarding the role of strain effect, we have taken into account the strain-induced lattice modifications in RuO_2 arising from its epitaxial growth on $Al_2O_3(1\bar{1}02)$ in the current calculation. In the calculations, the RuO_2 is adapted to the $Al_2O_3(1\bar{1}02)$ lattice constants, resulting in a strain of

+5.2% along the [010] direction and −6.6% along the $[10\bar{1}]$ direction. This strain alters the bond angles and lengths within the RuO_2 lattice, resulting in the lower surface energy and stabilization of the $RuO_2(101)$ variant. In the $Al_2O_3(1\bar{1}02)/RuO_2(\bar{1}01)$ configuration, as shown in Fig. R9b, the oxygen bonds at the interface are stretched and weakened, making this configuration less stable than the $Al_2O_3(1\bar{1}02)/RuO_2(101)$ configuration. Based on our calculations, the $Al_2O_3(1\bar{1}02)/RuO_2(\bar{1}01)$ structure exhibits a higher formation energy after relaxation, supporting the preference for the $RuO_2(101)$ variant, which is also consistent with our experimental observations.

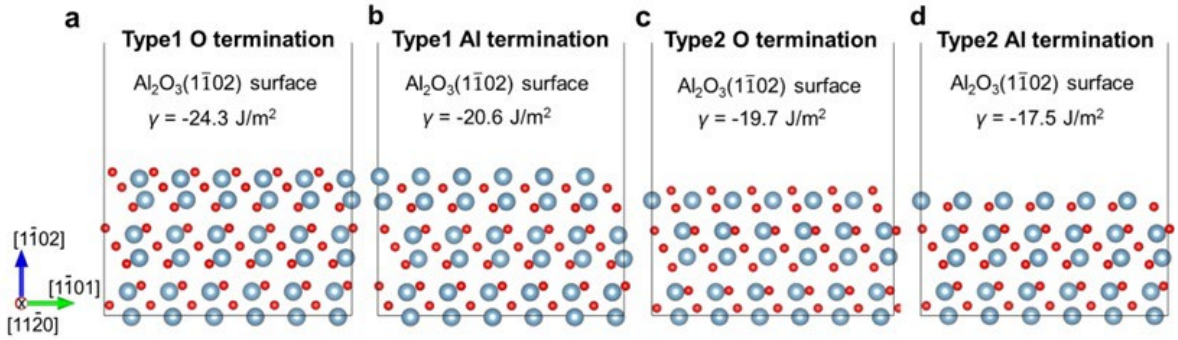

Figure R8. Illustrations of structures of $Al_2O_3(1\bar{1}02)$ surface. **a**, Type 1 with O termination, **b**, Type 1 with Al termination, **c**, Type 2 with O termination, **d**, Type 2 with Al termination. Red and light blue dots represent oxygen and aluminum atoms, respectively. The formation energy density for each case is also shown. (Figures are the same as Fig. R6.)

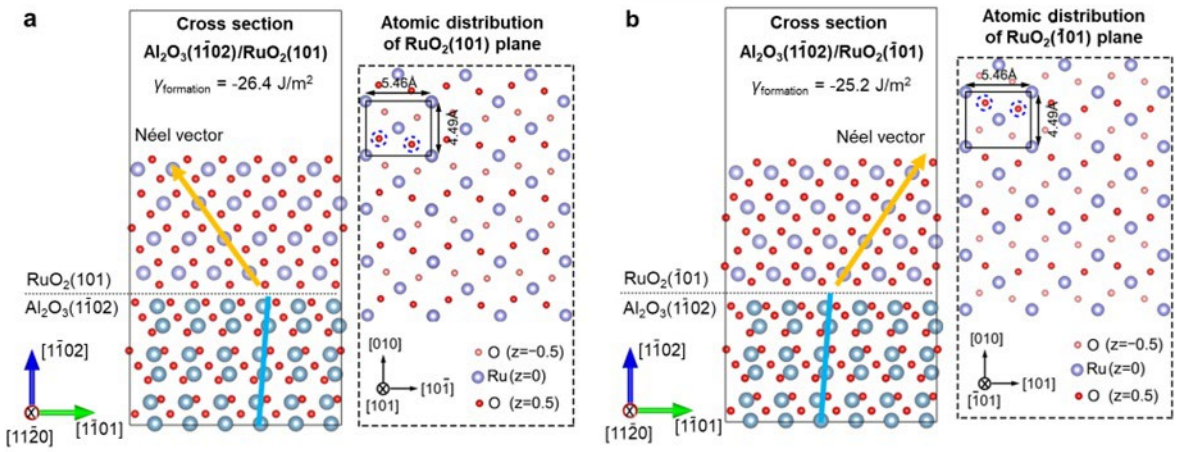

Figure R9. Illustrations of cross-sectional and in-plane crystal structures and atomic distributions of **a**, $Al_2O_3(1\bar{1}02)/RuO_2(101)$ and **b**, $Al_2O_3(1\bar{1}02)/RuO_2(\bar{1}01)$. Gray dots represent Ru atoms. Red and light blue dots are for oxygen and aluminum atoms, respectively. The formation energy density for each case is also shown. (Figures are the same as Fig. R7.)

- Please clarify why ± signs appear before the spin-conserving term which involves only orbital angular momentum operators without directly involving spin.

Reply: Thank you for the comment. The ± signs appear because the direction of the orbital moment depends on the spin-quantum axis. Specifically, the Ru site with a negative spin moment exhibits a negative orbital moment, while the Ru site with a positive spin moment displays a positive orbital moment. Consequently, the sign of the orbital moment anisotropy's contribution to the magnetic anisotropy energy (MAE) depends on whether the Ru site has a positive or negative spin moment.

- The authors demonstrate single-variant $RuO_2(101)$ formation. However, it remains unclear if the reverse orientation, $RuO_2(\bar{1}01)$, can also be selectively grown. Comments on whether or how to

achieve this variant would be insightful, as it would relate to understanding what fundamentally lifts the degeneracy between variants.

Reply: Thank you for the insightful comment. As shown in the reply to the Comment #1, as well as the reply to the comment #2 of Reviewer #1, the selective formation of the $RuO_2(101)$ variant is attributed to the stability of the oxygen-terminated $Al_2O_3(1\bar{1}02)$ substrate based on our first-principles density functional calculations. These calculations also reveal that the $Al_2O_3(1\bar{1}02)/RuO_2(101)$ is more stable than that for the $Al_2O_3(1\bar{1}02)/RuO_2(\bar{1}01)$, indicating that the degeneracy is lifted by the interface structure. While our current study focuses on elucidating the stability of the $RuO_2(101)$ variant, achieving selective growth of $RuO_2(\bar{1}01)$ would require engineering the substrate termination or interface conditions to favor the orientation. This is an intriguing direction for future research, and we greatly appreciate the reviewer's suggestion.

Minor Suggestions:

- In Equation (1), clarify the notation for "I" in E_{MAE}^{I}, Δ_{ex}^I, and ξ_{I}, as well as the notation for expectation values ⟨ ... ⟩. Specify whether these refer to specific states.

Reply: We thank the reviewer for pointing this out. In the revised manuscript, we have added explanations clarifying that "*I*" refers to the Ru atomic sites (Ru1, Ru2) and provided definitions for the expectation values ⟨ ... ⟩ in the main text.

- Spell out "XRD" as X-ray Diffraction when first mentioned.

Reply: We thank the reviewer for carefully reviewing our manuscript. We have spelled out "XRD" as "X-ray Diffraction" when first mentioned in the revised manuscript.

- The term “unidirectional spin-splitting magnetoresistance (USSMR)” may be somewhat misleading. What is unidirectional in the ferromagnet/altermagnet bilayer is the magnetoresistance, rather than the spin splitting.

Reply: We appreciate the reviewer’s comment regarding the terminology. To address this, we have revised the manuscript by changing the term from “unidirectional spin-splitting magnetoresistance (USSMR)” to “spin-splitting magnetoresistance (SSMR).” This change ensures clarity and avoids any potential misunderstanding, aligning with the reviewer's clarification. We have updated all relevant sections in the manuscript accordingly and hope the changes improve the accuracy and precision of the terminology.

Reviewer #3 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reply: We sincerely thank the reviewer and their co-reviewer for their time, effort, and thoughtful comments/suggestions on our manuscript, which have helped us to improve our manuscript.

Reviewer #4 (Remarks to the Author):

I read the manuscript by He et al. with interest. However, I have doubts about the conclusions of this work. In my opinion, these doubts should be resolved before one can proceed further.

Reply: We thank the reviewer for his/her time in reviewing our manuscript. We appreciate the opportunity to address any doubts and clarify our conclusions. In the revised manuscript, we have provided additional experimental data, theoretical calculations, and discussions to resolve the concerns raised. We hope these revisions will address the reviewer's doubts and clarify our findings.

According to simple models, RuO2 is an altermagnet. These models assume antiferromagnetic arrangements of local moments on Ru sites. To obtain altermagnetic phase numerically in one-electron "ab-initio" calculations, the GGA+U method needs to employed. However, according to one paper [PRB 109, 134424 (2024)], the magnitude of the U-term that leads to magnetic order is higher than the value that describes other electronic properties of RuO2. Furthermore, two muon-spin-rotation studies have found that RuO2 is non-magnetic [PRL 132, 166702 (2024), arXiv:2405.10820 (2024)]. I believe these very recent studies have changed the perception of RuO2, and as a consequence they call for readjustment of the goals of experimental studies on RuO2. These goals have now been shifted from investigating altermagnetic properties of RuO2 into providing the evidence that there exist antiferromagnetic RuO2 samples. Importantly, if RuO2 is antiferromagnetic, it is almost certainly altermagnetic.

He et al. claim to provide the evidence of antiferromagnetism in RuO2 though the XMLD measurements at the Ru M-edge. In a simple model, this means that dipole transitions from the Ru 3p doublet into Ru 4d states at the Fermi level occur (in a crude approximation Ru 5s can be neglected). Optical matrix elements are not directly sensitive to spin, and the spin sensitivity of either XMCD or XMLD is a result of spin-orbit coupling, although the mechanisms are somewhat different in the two methods. Importantly, the measured current results from the Auger cascades and can be assumed fully angle-integrated. To the first approximation a non-vanishing XMLD effect is related to the orientation of the polarization of the light with respect to the orientation of the Ru orbitals at the Fermi level.

Ignoring itinerant properties, these Fermi level Ru 4d orbitals can be projected onto the real orbital basis (dz2, dxy, etc), and besides the contribution from the crystal bonds, the coefficients of that decomposition will slightly depend on the orientation of the Neel vector due to spin orbit coupling, potentially leading to magnetic sensitivity of XMLD. (Since Ru 5s are ignored, one does not even have to worry about radial integrals.) XMLD has been used to image antiferromagnetic domains in the PEEM mode, see e.g. Nature Communications 13, 724 (2022) -- but these results are much easier to be correctly interpreted because they are based on the contrast between different areas and on visual characteristic shapes of the domains.

Reply: We sincerely thank the reviewer for the comprehensive and insightful comments.

We acknowledge the recent studies [PRB 109, 134424 (2024); PRL 132, 166702 (2024); arXiv:2405.10820 (2024)] highlighting the challenges in establishing RuO_2 as an altermagnet. These works show the sensitivity of the magnetic ground state to the value of the U term used in GGA+U calculations, as well as experimental results indicating the absence of long-range magnetic order in their RuO_2 samples. However, as the reviewer pointed out, if RuO_2 were antiferromagnetic, it would exhibit altermagnetic properties. Our study aims to contribute to this ongoing discussion by providing experimental evidence for antiferromagnetic order in RuO_2(101) samples using XMLD at the Ru M-edge, alongside structural and spin transport characterizations.

We appreciate the reference to XMLD imaging of antiferromagnetic domains in PEEM mode [Nature Communications 13, 724 (2022)]. Because of the limitation of our facilities, this work does not involve direct imaging of domains. We agree with the reviewer that domain imaging could further validate these findings. We plan to pursue such investigations in future studies, although no domain images are likely obtained for the present single variant samples. In addition, to the best of our knowledge, PEEM only detects photoelectrons emitted along normal directions with a grazing

incident beam so it can identify in-plane antiferromagnetic domains in CuMnAs, while it has limitations in providing orbital state information. Orbital states could not be analyzed solely through domain imaging. Our XMLD measurements, with angular dependence, allow us to probe antiferro-(alter-) magnetic properties concerning the Neel vector direction of RuO_2(101). We also agree with the reviewer that the XMLD signal arises from spin-orbit coupling rather than directly measuring spin itself. As the reviewer mentioned, the XMLD signal includes contributions from structural orbital symmetries. To clarify these points, we have provided temperature-dependent measurements, which have been included in the Supplemental Materials. We have also added the related explanations in the revised manuscript and the following replies.

At least three conditions need to be satisfied in order to consider XMLD results of He et al. as a probe of antiferromagnetism in RuO2:

A. The single variant phase of the crystal should be larger than the photon beam spot

B. Magnetic domains should all have the same orientation of the Neel vector

C. XMLD effects non related to magnetism must be eliminated

I think none of the conditions have been fully satisfied in the study by He et al. Correct me if I am wrong, but concerning point A, the largest demonstrated size of the single variant phase is the TEM image in Fig. S4, the size is approx. 100 nm. Therefore, following the XRD results, possible existence of (-101) phase cannot be eliminated in XMLD, where photon beam has been certainly large.

Reply: Thank you for raising these critical points. Regarding point A, we have performed additional in-plane scan XRD measurements over a 10 mm ×10 mm area to evaluate the uniformity of the single-variant RuO_2(101). The detailed results are shown in the above reply to the Comment 1 of Reviewer #1. These new XRD results confirmed the absence of the ($\bar{1}$01) variant across the entire sample area,

ensuring the single-variant nature of the film over a significantly larger scale than indicated by the TEM images. Based on these results, the XMLD measurements are appropriate for discussing the magnetic properties of the single-variant RuO_2(101) phase. We have revised the manuscript to reflect this point.

We hope this addresses your concerns regarding point A, and we thank you for pointing out the important point.

Concerning point B, the goal of XMLD results is prove the existence of magnetic order, but B would impose an assumption of one type of magnetic order with moments along [001] -- results are supposed to prove what has been assumed, a logical issue. Also, if Neel vector is oriented along few discrete directions, there might exist geometries where XMLD could be sensitive to magnetic order, but this would need to be analyzed in detail.

Reply: Thank you for the insightful comment. We consider that uniform magnetic domains are closely related to the structural single-variant nature of the film. As addressed in point A, from the XRD results, we have confirmed the single-crystal domain of RuO_2(101), ensuring structural uniformity. Based on this structural uniformity, it is expected that the magnetic domains exhibit uniform Néel vector orientation, as noted by the reviewer that the magnetic domains should all have the same orientation of the Néel vector. Also, our first-principles calculation section shows the magnetic order with moments along [001]. Regarding the sensitivity of XMLD to magnetic order and geometries, we performed angular-dependent XMLD measurements in our experiments. The changes in XMLD intensity and sign observed in the angular dependence of XMLD measurement provide the information about the Néel vector orientation along the [001] direction. This result avoids relying purely on assumptions and instead allows us to infer the Néel vector orientation from the measured

XMLD experimental data, which is also consistent with the magnetic anisotropy along [001] direction from our first-principles calculations.

We appreciate the reviewer highlighting the importance of analyzing potential geometries for XMLD sensitivity. We have revised the manuscript to further clarify this point. We hope this addresses the reviewer's concerns.

Concerning point C, the (101) surface studied by He et al. does not seem to retain much of the symmetry of the bulk crystal. The analysis of the results from Fig. 3 should start with establishing the symmetries of the (101) surface and their relation to symmetries of XMLD signals. I doubt XMLD can be treated in the bulk limit. Furthermore, an unlikely coincidence may lead to thin films having some residual anisotropy due to substrate terraces, deposition angle etc. -- but probably this is not the case here. Once the surface symmetry is understood, one should assume pure (101) surface or a combination of (101) and (-101) and see which light incidence angle should lead to equivalent XMLD spectra. Perhaps, since domain-from-domain contrast is not available like in XPEEM, one should also consider XMLD process description beyond a simplified single-electron LCAO picture, to see if it introduces additional effects.

Reply: Thank you for raising this point. We appreciate the opportunity to clarify and elaborate on our analysis concerning point C.

Regarding the symmetry of the RuO_2(101) surface: We examine the symmetry of the RuO_2(101) surface, where the Néel vector is along the [001] orientation (z axis). The surface exhibits yz and zx orbital symmetries along the surface normal direction, where x and y are along [100] and [010] directions, respectively. Our XMLD results in Fig. 3 indicate that the XMLD signal arises when the beam electric field couples mainly with the magnetic $3z^2-r^2$ orbital states. While XMLD is surface-sensitive, the angular dependence of XMLD measurements catches up with the bulk RuO_2 magnetic

properties, as the Néel vector orientation aligns with the observed angular dependence. We have added the discussion on this point in the manuscript to clarify this relationship.

For the possibility of residual anisotropy due to external factors (e.g., substrate terraces or deposition angle): Such effects could potentially influence the XMLD signals; however, as the reviewer noted, these effects are unlikely in this case. The uniformity of the RuO_2(101) variant across the large-scale sample was confirmed by extensive in-plane XRD measurements, as detailed in the previous replies. The observed XMLD signals exhibit obvious angular dependence further supporting the intrinsic magnetic origin of the signals. Furthermore, to address the contributions of structural and magnetic components, we conducted temperature-dependent XMLD measurements. As shown in Fig. R10, the temperature dependence of XMLD is evident, with measurements taken at 80, 200, 300, and 400 K. Given that the Néel temperature of RuO_2 is approximately 400 K, the XMLD intensity at 400 K is significantly reduced compared to 80 K. This difference is attributed to the magnetic contribution, as structural components are temperature-independent. These results provide evidence that the XMLD signals include a significant magnetic contribution, and the related description and discussion are added in the revised manuscript and supplementary materials.

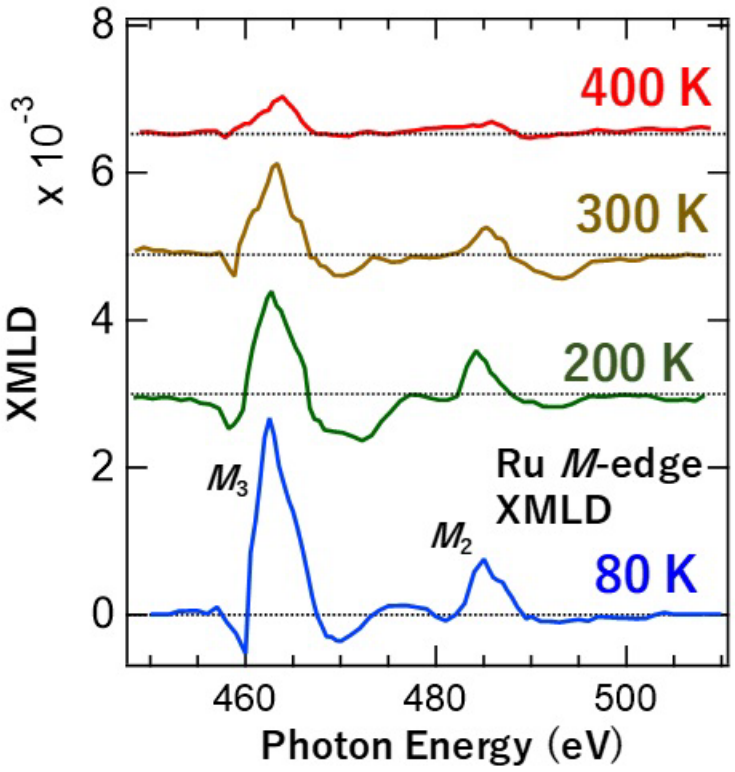

Fig. R10. Analysis of XMLD spectral line shapes measured at 80, 200, 300, and 400 K.

Regarding the inclusion of both (101) and ($\bar{1}$01) phases: As detailed in the responses to earlier points, our additional XRD measurements confirm the absence of the ($\bar{1}$01) phase across the entire sample area, ensuring that the XMLD spectra arise solely from the (101) variant. This can eliminate an equivalent XMLD spectrum resulting from a mixture of (101) and ($\bar{1}$01) domains.

About a more detailed description of the XMLD process: We acknowledge that a simplified single-electron LCAO picture may not fully capture all effects influencing XMLD signals. While our current analysis focuses on the primary angular dependence and symmetry considerations, we agree that a more comprehensive description of the XMLD process could provide additional insights. This may be relevant for capturing subtle effects introduced by orbital hybridization or spin-orbit coupling

at the surface. We have included a discussion in the revised manuscript highlighting the need for further theoretical work to refine the XMLD process description for a complex system.

We hope the above explanation addresses the reviewer's concerns. Thank you for the valuable comments, which have allowed us to improve our manuscript.

My suggestion is that once this is established, and the result points towards the confirmation of magnetic order in these RuO2 films, one can proceed with the analysis of the subsequent part of the paper, conclusions of which are based on the assumption of the existence of the magnetic order. Independent on this, I consider this paper an extensive experimental and theoretical characterization of a new type of high-quality RuO2 films and I appreciate this work. My review is written from the perspective of whether the manuscript matches the criteria of Nature Communications.

Reply: Thank the reviewer for his/her thoughtful and constructive comments. Also, we greatly appreciate the reviewer for the consideration of the experimental and theoretical works on the high-quality RuO_2 film and its potential significance.

We fully agree with the suggestion that confirming the magnetic order is a foundational step for the subsequent analysis and conclusions. In the revised manuscript, we have explicitly determined the structural orientation using XRD and TEM, ensuring a robust foundation for the subsequent magnetic characterization. We have provided additional clarifications and supporting evidence regarding the XMLD results and their connection to the magnetic order in RuO_2. Specifically, we have emphasized the angular and temperature-dependent XMLD measurements. These efforts aim to establish the existence of antiferromagnetic (altermagnetic) order in the single variant RuO_2(101) thin films. We sincerely thank the reviewer for the valuable comments and suggestions, which have helped us refine our work, and for consideration of the manuscript's alignment with the criteria of *Nature Communications*.

Response letter

We sincerely thank the reviewers for insightful comments to our manuscript. Based on the comments raised by the reviewers, we have revised the manuscript accordingly. The response to each comment is given in this letter. We hope that our revised manuscript and replies now dispel the concerns of reviewers. We would be grateful if the revised manuscript could be considered for timely publication. If you have any additional comments or specific suggestions, we would be glad to address them as well.

The corresponding changes in the main text have been marked by a highlight color in the revised manuscript and supplementary materials.

Thank you very much for your kind consideration.

Note: Our replies are in blue fonts in the following sections.

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

It appears the authors have carefully considered the previous reviewer's comments. In particular, the detailed XRD experiments seem sufficient to demonstrate that the thin film is a single variant. I appreciate the authors' efforts over the long review period. I believe this paper will be significant for research and applications in RuO_2-based altermagnetism, and I find it meets the standards of Nature Communications. I recommend its publication.

Reply: We thank the reviewer for the kind and encouraging feedback. We appreciate your recognition of our additional XRD measurements and your precious time during the review process.

Reviewer #2 (Remarks to the Author):

The authors have addressed most of my previous comments and have improved the manuscript accordingly. Their DFT calculations on the strain effects and surface termination of $Al_2O_3(1\bar{1}02)$ in stabilizing $RuO_2(101)$ over $RuO_2(\bar{1}01)$ remain technically sound, effectively incorporating formation energy analysis and epitaxial strain effects. However, their assumption of a purely oxygen-terminated $Al_2O_3(1\bar{1}02)$ surface may still be idealized, as mixed terminations (coexistence of O- and Al-terminated regions) are possible due to thermodynamic competition, surface reconstructions, and growth kinetics. If such mixed terminations exist, they could affect the relative stability of RuO_2 variants and potentially modify strain effects at the interface. While their results remain reasonable, a brief discussion on the possibility of mixed terminations and their implications would further strengthen their argument.

Reply: We sincerely thank the reviewer for the thoughtful evaluation and for recognizing our efforts to improve the manuscript.

As the reviewer pointed out a brief discussion on the possibility of mixed terminations and their implications, we have followed the reviewer's suggestion and added the related discussions in the revised manuscript. The possibility of mixed surface terminations, due to thermodynamic competition and kinetic factors, may affect the stabilization of $RuO_2(101)$ versus $RuO_2(\bar{1}01)$ variants. Once two variants exist, one would detect two peaks in the in-plane XRD φ scan (Figs. 1f-1h) and disappearance of angular shift in spin-splitting magnetoresistance measurements (Fig. 5c). Therefore, surface treatment of the $Al_2O_3(1\bar{1}02)$ substrates (e.g. 1000 °C baking treatment) and precise control of growth conditions (e.g. O_2 atmosphere) are crucial for achieving single-variant $RuO_2(101)$ thin films. With this discussion, we have a more comprehensive understanding of the epitaxial interface.

We thank the reviewer again for this constructive suggestion, which has helped strengthen the manuscript.

Regarding magnetoresistance effects, the authors measured R_{2¥omega}. However, in principle, the spin splitting effect should also induce angular dependence of R_{¥omega} in linear response, where the spin current polarization and the corresponding magnetic field direction associated with the magnetoresistance extrema are largely determined by the Néel vector of the altermagnet/antiferromagnet. Given its linear-response nature, this effect is expected to be more pronounced than its second-order counterpart. This is a worthwhile aspect to investigate further, and the authors may have already measured it using the same experimental setup for R_{2¥omega}. Including (or commenting on) the angular dependence of R_{¥omega} in their magnetoresistance measurements would provide a more complete picture of spin transport effects, and with these revisions, the manuscript would be in better shape for publication.

Reply: We sincerely thank the reviewer for this insightful and constructive comment. We agree that the angular dependence of R_{ω} in linear response is an important aspect to explore, especially as it may reflect the orientation of the Néel vector in altermagnets and provide complementary information to the second harmonic $R_{2\omega}$ measurements.

As the reviewer pointed out, the first harmonic R_{ω}, known as the conventional resistance, represents the linear response of the samples to the current [Avci et al., Nat. Phys. **11**, 570 (2015)]. Based on previous studies, in heavy metal/ferromagnetic bilayers [e.g., Choi et al., Phys. Rev. B **96**, 174412 (2017)] and YIG/heavy metal systems [e.g., Nakayama et al., PRL **110**, 206601 (2013)], the angular dependence of R_{ω}, when rotating the magnetic field in the plane perpendicular to the current, is typically attributed to the

spin Hall magnetoresistance (SMR) effect, while the second harmonic signal $R_{2\omega}$ originates from the unidirectional spin Hall magnetoresistance (USMR) effect [Avci et al., Nat. Phys. **11**, 570 (2015)].

Regarding the mechanisms behind these effects, it is reported that the SMR involves spin current generation (via spin Hall effect) in the heavy metal layer (i.e., spin source layer), the interaction between the spin current and the adjacent FM layer, and the back flow of the spin current into the heavy metal layer which increases the conductivity of the heavy metal due to the inverse spin Hall effect (ISHE) [Nakayama et al., PRL **110**, 206601 (2013), Althammer et al., PRB **87**, 224401 (2013)]. However, the USMR is due to the SHE-induced spin accumulation and modulation of the HM/FM resistance with respect to the relative orientation between the spin current polarization and the magnetization of FM layer, similar to the conventional GMR effect [Avci et al., Nat. Phys. **11**, 570 (2015)].

In our RuO_2/CoFeB bilayer system, we focused on the second harmonic $R_{2\omega}$ while we did not collect R_{ω} data along with $R_{2\omega}$ because of the limitation of our measurement setup. Also, based on the above explanations on the mechanisms, we initially considered that the first harmonic R_{ω} in our altermagnet/FM bilayers could be ascribed to the combined action of both the spin-splitting effect and the inverse spin-splitting effect, which appears to be more complicated than the $R_{2\omega}$ component.

We have also noticed that a recent preprint [arXiv:2412.18220] systematically investigated the angular dependence of the longitudinal resistance R_{xx} which can be considered as the first harmonic resistance. They also observed an angle shift linked to the Néel vector of the altermagnet in the angular dependence of the longitudinal resistance

R_{xx}. This independent work, consistent with our findings, supports the existence of spin-splitting magnetoresistance in RuO_2.

In the revised manuscript, we have added the related discussions to clarify this point. We thank the reviewer again for this valuable suggestion, which has helped improve the clarity of our work.

Reviewer #3 (Remarks to the Author):

I co-reviewed this manuscript with one of the reviewers who provided the listed reports. This is part of the Nature Communications initiative to facilitate training in peer review and to provide appropriate recognition for Early Career Researchers who co-review manuscripts.

Reply: We thank the reviewer for their involvement in the review process. We are grateful for your time and effort in evaluating our work.

Reviewer #4 (Remarks to the Author):

I would like to thank the Authors for their response and for including additional temperature-dependent XLD measurements in the Supplement. Indeed, such temperature dependence of the XLD could be an evidence of magnetic order in RuO2. However, other factors could also influence the XLD, and without ruling them out, I am not fully convinced that the observed changes are directly linked to antiferromagnetism. For example, I am not sure what is the expected temperature-dependence of XLD at 3p edge in a non-magnetic compound containing Ru or another 4d element. Additionally, since it is not established whether in nature RuO2 is antiferromagnetic (I am referring to recent papers on the topic), its Néel temperature also remains uncertain.

I appreciate the Authors' efforts in thoroughly characterizing their films. However, in my view, a clear and direct connection to magnetism has not yet been demonstrated. Consequently, I am unable to recommend the publication of the paper in Nature Communications.

Reply: We sincerely thank the reviewer for the thoughtful comments. The magnetism in RuO_2 is indeed controversial from some previous reports, but we respectfully differ with the reviewer's conclusion that our results do not clearly demonstrate antiferromagnetic ordering in our RuO_2 samples.

First, we have confirmed the Néel temperature in our RuO_2 samples. The resistivity measurements (Fig. S2 in Supplementary and a new Fig.1j in Main text) reveal a Néel temperature close to 390 K, which is consistent with previous reports (e.g., Fig. 2b and Fig. 4c in Nat. Electron. 5, 735-743 (2022)). Then, the temperature-dependent XMLD measurements conducted over a wide temperature range (80 K to 400 K), shown in Fig. S7a of Supplementary, indicate a monotonic decrease in signal amplitude with increasing temperature, and a significant suppression at 400 K. Since 400 K exceeds the Néel temperature of RuO_2, the spectral intensity taken at 400 K consists of almost XLD contribution. The XMLD data and resistivity measurements are consistent and support the antiferromagnetic order in our RuO_2 samples.

Second, regarding the possibility of structural contributions to XLD: to the best of our knowledge, the structural components of XLD are generally temperature insensitive (except for thermal broadening) unless there is a structural phase transition. Based on our results (e.g., temperature-dependent resistivity), we do not observe such a structural transition in our RuO_2(101) films. Therefore, the strong temperature dependence of XMLD shown in Fig. S7a is primarily attributed to the magnetic component. Additionally,

regarding the reviewer mentioned “not sure what is the expected temperature-dependence of XLD at 3p edge in a non-magnetic compound containing Ru or another 4d element”, we have tried to search the database for all compounds containing 4d elements that adopt the rutile structure. However, we are unable to identify a suitable non-magnetic reference compound with the rutile structure and containing another 4d element that could serve as a direct comparison for evaluating the temperature dependence of XLD at the 3p edge. Given the crystal structural constraints and the lack of ideal candidate materials at present, we plan to explore this direction in our future work to provide deeper insight into the temperature dependence of XLD in such systems, which lies beyond the scope of the present manuscript.

Third, we have also noticed that recent independent works (arXiv:2412.17016, arXiv:2412.17013) report the observation of antiferromagnetic order in RuO_2 using XMLD in different orientations ((100) and (110)), including Néel vector switching was detected via XMLD. These results further validate our interpretation that RuO_2 can host magnetic order under appropriate deposition conditions. In the revised manuscript, we have added the discussion that sample-to-sample variations (e.g., growth conditions, uniformity, Ru or O vacancies, crystal orientation) may result in contradictory reports on magnetism in RuO_2.

Overall, our combined structural, transport, and XMLD analyses provide mutually consistent and direct evidence of a single variant in altermagnetic RuO_2(101) thin films; for example, the observed spin-splitting magnetoresistance (SSMR) effect cannot be interpreted unless the RuO_2 film has the Néel vector along the [001] direction. We hope this work will advance the ongoing discussion and provide critical insight into altermagnetic materials.

We have included the above discussions in the revised manuscript. We hope these clarifications have addressed the reviewer's concerns. With these revisions, we would be grateful if the reviewer would be satisfied and recommend our revised manuscript for publication.