

Tuning a magnetic energy scale with pressure and field in UTe_2

Corresponding Author: Professor Johnpierre Paglione

This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.

Version 0:

Decision Letter:

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Dear Professor Paglione,

Thank you for submitting your manuscript, "Tuning a magnetic energy scale with pressure and field in UTe_2 ", to Communications Materials, and please accept our sincere apologies for the unusual time required to review your study, due to end-of-year commitments of our reviewers.

Your manuscript has now been seen by 3 referees, whose comments are appended below. You will see that while they find your work of potential interest, they have raised substantial concerns that must be addressed. In particular, the evidence for non-Fermi liquid and magnetic criticality require a more rigorous and solid foundation, and the reviewers have provided very detailed and constructive indications on how the results and conclusions should be substantiated by further data and analysis.

In light of these comments, we cannot accept the manuscript for publication, but are interested in considering a revised version that addresses all these serious concerns.

We hope you will find the referees' comments useful as you decide how to proceed. Should further experimental data and analysis allow you to address these criticisms, we would be happy to look at a substantially revised manuscript. However, please bear in mind that we will be reluctant to approach the referees again in the absence of major revisions. If the revision process takes significantly longer than three months, we will be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Materials or published elsewhere in the meantime.

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

When submitting your revised manuscript, please include the following:

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

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

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Please do not hesitate to contact me if you have any questions or would like to discuss the required revisions further. Thank you for the opportunity to review your work.

Best regards,
Aldo

Dr Aldo Isidori
Senior Editor
Communications Materials

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

In the manuscript under review, the authors present experimental data for resistivity from UTe₂ samples taken under application of pressure and magnetic field. They observe among known features the upper critical field as function of temperature for the different pressures investigated and an anomaly in the resistivity (maximum) that earlier has been attributed to an energy scale of magnetic fluctuations. The presented data can help to understand the unusual phases of UTe₂ and lead to further understanding of quantum critical behavior in this system as well as the superconducting mechanism. Generally, the work seems suitable for publication in principle, but some details need to be cleared up in order to put the discussion and significance on more solid grounds.

As guidance, a couple of comments as these appear in the manuscript:

- 1) The author highlight in the abstract (line 12) the “clear picture of energy scale evolution”. Can this statement be sustained with actual data or be distilled from it analysis? Which energy scale is measured? What is its value? How is this deduced?
- 2) Related comment on this, line 59: The authors connect the resistance anomaly to “magnetic quantum criticality”. How does the data of the current measurement support the connection to a magnetic criticality? Or is this argument based on the interpretation of data in Ref. [11]? Is there any argument why magnetic fluctuations should lead to a feature that simply shifts with pressure, but broadens with field?
- 3) The authors characterize their samples as exhibiting $T_c=1.6$ K (in line 63) which from the perspective of available samples seems not the highest T_c achieved in this system. As one can see from the literature, samples of different “generations” have common and distinct features (such as the double transition seen in specific heat for example, see Rosa, et al., Communications Materials volume 3, 33 (2022) - published version of Ref. [7]). In order to connect the current study from a material perspective to the available literature, there should be said a bit more about the crystal characterization. For example what is the RRR? Is it known whether crystals from the same batch show double transition in specific heat?
- 4) The authors describe the maximum of the resistivity to be “strongly tuned by magnetic field” (line 96). It seems that the anomaly is in principle a skew peak, thus a general broadening (in temperature) alone might shift its maximum as well (without actually changing the underlying energy scale). Can this be ruled out based on the available data such that there is a real shift of energy scale upon application of field.
- 5) Fig. 3 (panel a): There is some trouble with the two blue triangles which makes it difficult to read the data. The rotation of one of the triangles does not match the legend. Please consider changing color and / or symbol to avoid confusion.
- 6) Fig. 3, caption: The authors write for panels (d-h) “all measured pressures (...) exhibits a sudden slope change”. This should only hold for 4 of the 5 pressures, in panel (h), I cannot see a kink in the T_{sc} . Please adjust. Note this is also explained in the main text, line 153.
- 7) line 111: The authors write “also reveals interesting pressure evolution” and it is not clear what particularly is interesting on the data. Indeed, there comes a longer discussion of the data, but the authors do not stress what is the most unusual behavior. Is it the back-bending, or the increased slope (which actually is needed to go over to a back-bending and re-entrance behavior)?
- 8) line 119: One more precise reference to the figure panel could be useful here.
- 9) All figures: The authors present resistivity data and measured positions of features for different series of pressures or fields. It could be useful to have a consistent set of symbols and colors to better allow for comparison. For example Fig. 3(a) $p=0$ has black square, but Fig. 4 it is $p=5.3$ as in Fig. 2. Even within Fig. 2 the symbols change between panel (f) and (g).
- 10) Discussion section: Can there be anything said about the connection of the energy scale detected in this experiment and the connection to superconductivity? The authors advocate for the peak in the resistivity to be associated with ω_{spin} , an energy scale for spin fluctuations. Is this consistent with the spin fluctuations responsible for superconducting pairing?
- 11) Note that as style for Communications materials, it is advised to have titles for all references which simplifies the recognition of works and among others also simplifies the work of the referees.

Reviewer #2 (Remarks to the Author):

The manuscript "Tuning a magnetic energy scale with pressure and field in UTe₂" by Hyunsoo Kim and coauthors reports on the tuning of a magnetic energy scale in UTe₂, where hydrostatic pressure and a magnetic field applied along the a-axis are used as tuning parameters. A characteristic peak in the c-axis electrical transport is shown to be suppressed by increasing hydrostatic pressure and enhanced by applying a magnetic field along the a-axis. The authors show that this feature in the electrical resistivity coincides with features previously observed in the resistivity and specific heat, demonstrating that resistivity can be used to track the evolution of the underlying energy scale, which is suspected to be of magnetic origin.

The authors also analyze the resistivity data for evidence of non-Fermi liquid (FL) behavior, by extracting a power law exponent from a double-logarithmic representation of the temperature dependent data. They argue that non-FL behavior is observed over a wide range of magnetic fields and pressures, and that it may be related to an underlying quantum critical point (QCP) in UTe₂. The power law exponents show obvious deviations from the expected T^2 temperature dependence for the resistivity of a Fermi liquid at low temperatures. However, I question whether a convincing power law can be extracted from this dataset, since it is dominated by the magnetic resistivity peak and features associated with the superconducting transition, and the data only span a temperature range from ~ 0.5 to 14 K. In other compounds, e. g. YbRh₂Si₂, where non-FL liquid was demonstrated in connection with a QCP, a non-FL temperature dependence of the resistivity could be demonstrated over several decades in temperature, and the region of non-FL behavior was clearly centered around the QCP. I also find the color-map representation of the exponents in Fig. 4 misleading, since a wide range of exponents, e. g. from 1.25 to 1.75 is represented by virtually indistinguishable colors. To establish a power law-temperature dependence, I would find it more helpful if the authors showed actual fits of one power-law exponent to the raw resistivity data, rather than plotting the derivative of a double log plot or showing a plot of the fit parameters without plots of the fits.

Finally, the manuscript is lacking a clear conclusion: it is literally absent from the text, which jumps directly from a description of the data to the methods section. The authors need to clearly establish what new information this paper contributes to the field, beyond a phenomenological description of the data. It is encouraging that the resistivity features track the previously reported magnetic energy scale, and establishing resistivity as an accessible and straightforward probe of non-FL behavior and possibly quantum criticality in UTe₂ is valuable, but the arguments need to be clearly laid out in the manuscript. Additional data (e. g. resistivity over a larger temperature range) may also be needed to clearly demonstrate non-FL behavior and a quantum critical point.

In conclusion, I do not recommend publication of the manuscript in its present form, but I suggest a resubmission after addressing these concerns.

Reviewer #3 (Remarks to the Author):

The manuscript describes an investigation of the H-P-T phase diagram of UTe₂, a material with high field reentrant superconductivity and a metamagnetic transition.

The authors measured conductivity along the c-axis under different pressures and magnetic fields that are aligned along the a-axis.

The study is timely and sheds light on the thermodynamic of the different electronic phases that form under the application of thermodynamic variables pressure and magnetic field. While simultaneous application of both pressure and magnetic fields along different crystallographic directions in UTe₂ has been reported previously (as given in refs 13 & 18), the new information obtained in this work needs to be put into a better perspective.

The fact that the magnetoresistivity (Fig 4b) shows a broadening transition may indicate that there is hysteretic behavior in a two-phase region. Has this been studied in more detail?

The tuning aspect (see title) defines surfaces in spaces that contain pressure and magnetic field as coordinates, and some definition of the magnetic energy. It is therefore instructive to show such a surface, for instance conductivity-pressure-magnetic field, power-law-exponents – pressure – magnetic field, etc. Fig 1c could be better represented by such a surface and related projections.

Estimates of the magnetic energy scale that is tuned should be given. What is the order of magnitude of the magnetic energy scale and what are the coupling coefficients to pressure and magnet field?

The manuscript needs a table summarizing the variables that have been obtained from the analysis of the data. This table could be placed into the supplement.

The fonts in the figures are not sufficiently large to allow easy reading. The figures need to be modified.

The pressure is given in kbar, and GPa units should be added as well.

I would be helpful to the readers if the authors provide a schematic of the experimental setup indicating the orientation of the

sample, dimensions, contact geometry, current and electric field direction, etc.

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Version 1:

Decision Letter:

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Dear Professor Paglione,

Thank you for submitting your revised manuscript, "Tuning a magnetic energy scale with pressure and field in UTe_2 ", to Communications Materials. It has now been seen again by 2 of the referees, whose comments are appended below. You will see that while they find your work greatly improved, some points from Reviewer #1 are still requiring your attention and will require an additional round of revisions.

We are definitely interested in publishing your study in Communications Materials as soon as possible, but we would like to consider your response to these concerns in the form of a revised manuscript before we can publish your work.

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

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

When submitting your revised manuscript, please include the following:

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

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

-A clean version of the manuscript. Please select the file type 'Article File'.

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

Best regards,
Aldo

Dr Aldo Isidori
Senior Editor
Communications Materials

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

The referee has examined the full package of rebuttal and revised manuscript/supplement where the authors have made adjustments following the comments from the previous round. Indeed, a number of findings of the referees are not completely new in the literature; proper discussion of this point and interpretation of the new details have been incorporated in the manuscript. Therefore, the referee recommends publication of the manuscript in Communication Materials.

Reviewing the full content, there are some additional comments to be used to improve the manuscript further:

1) page 6, line 107: The authors have added some comment regarding connection to superconductivity as requested. However, the argumentation might not be as straightforward as outlined: Indeed, the observation of decreasing magnetic ORDER and increase of T_c for superconductivity is a signature of competition. However, an increase of (the temperature scale of) magnetic fluctuations as the signatures in resistivity are interpreted might also lead to increased T_c if the cutoff frequency in BCS theory becomes larger and spin fluctuations are mediator of superconducting pairing.

2) Fig. 3, panel (b): Indeed, this figure is to demonstrate that the $H_c(0)$ is larger than Pauli limit. However, at least for zero pressure, there is Δ_0 measurements from STM, so one could state the expected H_P for this case instead of relying on the Δ/T_c ratio entirely. (See also later comment.)

3) page 8 line 161, 164: The authors estimate H_P using the BCS ratio for Δ/T_c . To come up with $\alpha=1.87$ T/K. This is a very rough estimate, maybe even misleading and not applicable especially for UTe_2 which is an unconventional superconductor, has strong spin orbit coupling and is a correlated material. Recently, it has pointed out that exactly this system strongly violates expected BCS ratio, see fig 1 panel h of Nature volume 600, pages 636–640 (2021) where UTe_2 stands out. Indeed, under pressure there is no data for the gap magnitude; a violation of BCS ratio might strongly affect the estimate of H_P .

4) page 13, line 267: The authors have in the reply clarified that their samples are from the same batch of samples where double transitions are observed in specific heat. The information that the resistivity sample is from the same batch of samples which show double transitions as in the work by Hayes et al. should be stated somewhere.

5) similar as comment above: It might be advisable to state that the last two columns of Table II (H_{HW} and H_P) are calculated values (where the second has assumed BCS ratio which might be strongly violated in UTe_2).

Caption Fig. 3: Please check whether Panels (d-h) present $H_c2(T)$. Seems that T_c is plotted as function of H , see label on figure.

Typo: "UT2" -> " UTe_2 " in caption of Fig. 2

Reviewer #3 (Remarks to the Author):

The revised manuscript has addressed most of the requests for clarification, and is publishable in its current form.

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Dear Professor Paglione,

Your manuscript titled "Tuning a magnetic energy scale with pressure and field in UTe_2 " has now been seen again by our referees, who have recommended its acceptance in their confidential remarks to the editors. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Materials.

We therefore invite you to edit your manuscript to comply with our journal policies and formatting style in order to maximise the accessibility and therefore the impact of your work.

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Best regards,
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SUMMARY OF THE MAJOR CHANGES MADE:

- We revised the manuscript to comply with the reviewers' comments, which are indicated in magenta in the revised manuscript.
- We incorporated the appendix into the main text and created supplementary information including three figures and four tables.
- We changed symbols and colors in Figures 1, 2, and 3 to keep consistent symbols and colors for the same field and pressure throughout the manuscript.
- Schematics of the sample configuration are added to Figure 1 panel (a) and Figure S1.
- The color scheme of Figure 4 panel (a-e) has been changed to discern the power-law exponent clearly.
- Power-law fitting results (curves) were added to each panel of Figure 5.
- The font size of each figure is increased for easier reading.
- The style of references is changed, which include the title of the reference.

I. REVIEWER #1 (REMARKS TO THE AUTHOR):

In the manuscript under review, the authors present experimental data for resistivity from UTe₂ samples taken under application of pressure and magnetic field. They observe among known features the upper critical field as function of temperature for the different pressures investigated and an anomaly in the resistivity (maximum) that earlier has been attributed to an energy scale of magnetic fluctuations. The presented data can help to understand the unusual phases of UTe₂ and lead to further understanding of quantum critical behavior in this system as well as the superconducting mechanism. Generally, the work seems suitable for publication in principle, but some details need to be cleared up in order to put the discussion and significance on more solid grounds.

As guidance, a couple of comments as these appear in the manuscript:

Response: We thank the reviewer #1 for the accurate summary of our work and overall positive evaluation. We respond to comments point-by-point below.

1) The author highlight in the abstract (line 12) the “clear picture of energy scale evolution”. Can this statement be sustained with actual data or be distilled from it analysis? Which energy scale is measured? What is its value? How is this deduced?

Response: The electronic ground state of UTe₂ is in proximity to the magnetically ordered phase, and therefore strong spin-fluctuation is expected at low temperatures. The energy scale here refers to a magnetic energy scale associated with the spin fluctuation which clearly influences the scattering process in the c -axis transport with a pronounced resistivity local maximum around 12 K. The microscopic origin of the spin fluctuation remains unclear to date although the Kondo effect [1] and the Schottky-like anomaly [2] seem plausible for transport and thermodynamic properties, respectively. The pressure- and field-evolution of this energy scale can be clearly tracked for the first time by transport measurements with $I \parallel c$ and $H \parallel a$ in our work. Such association of an energy scale with a characteristic temperature is commonly made in heavy fermion compounds (e.g. CeCoIn₅ [3]). We deduce the energy scale of ~ 1 meV based on the characteristic temperature $T^* = 12$ K from the observation of the c -axis peak in our transport measurement at ambient pressure, which was also observed in the a -axis transport [1, 4], dc-magnetization [5, 6], nuclear magnetic resonance [7], and thermal expansion [4, 8]. The magnetic nature of the c -axis peak was confirmed from thermodynamic analysis in Ref. [4]. We revised the last sentence of the abstract and the

main text (see below) to point out and address this issue.

Line 50: A similar peak in the electrical transport measurement was identified at 16 K in an unoriented sample [9] and in a sample under applied pressure [10]. Other measurements – including heat capacity [4], linear thermal expansion coefficient [8], nuclear magnetic resonance [11], and thermoelectric power [12] – also exhibit a prominent feature near 12 K, a temperature scale which has been associated with a magnetic energy scale of ~ 1 meV based on thermodynamic measurements [4].

2) Related comment on this, line 59: The authors connect the resistance anomaly to “magnetic quantum criticality”. How does the data of the current measurement support the connection to a magnetic criticality? Or is this argument based on the interpretation of data in Ref. [11]? Is there any argument why magnetic fluctuations should lead to a feature that simply shifts with pressure, but broadens with field?

Response: The magnetic nature of the c -axis peak is now widely accepted [1, 2, 4, 13]. We demonstrate that the peak temperature T^* decreases with pressure, accompanied by increasing T_c and the development of the non-Fermi behavior (violation of T^2 resistivity). This behavior is a hallmark of magnetic quantum criticality. While there is no general agreement on a microscopic model, changes in lattice parameters due to pressure and consequential changes in electronic structure result in the observed phase diagram in UTe_2 . Similar phase diagrams were observed in many strongly correlated systems including heavy fermion [14, 15], cuprate [16], and iron-based [17] superconductors. Regarding the field dependence, the c -axis peak moves to higher temperature and becomes broader with $H \parallel a$ (this work) whereas the application of $H \parallel b$ shifts the peak to lower temperatures and becomes narrower [13]. While the microscopic origin of this field-dependence is not known, it can be explained with a two-level model (Schottky-like anomaly [2]) with anisotropic Zeeman interaction where the energy separation depends on the field orientation, and the inelastic scattering is proportional to the population of the excited spin state. However, this scenario and its connection to QCP need to be verified. To address these issues we include the following sentences in the revised manuscript.

Line 56: While there are varying views on the nature of magnetic fluctuations and their observation in UTe_2 [18–20], previous studies consistently point to this energy scale as being magnetic in origin, allowing the tracking this feature in c -axis transport to be used as a unique tool to study the evolution of the magnetic fluctuation spectrum in UTe_2 .

Line 130: The observed field dependence of T^* is in stark contrast to the case with $H \parallel b$ [13], which

narrows and shifts the peak to lower temperatures on increasing field. Together with previous work [13, 21], our results may support a recent $5f^2$ crystal-field model [2] that predicts the Schottky-like features observed in transport and thermodynamic quantities, assuming an anisotropic Zeeman interaction where the energy separation depends on the field orientation, and inelastic scattering that is proportional to the population of the excited spin states.

Line 191: Whereas Fermi liquid behavior is routinely manifested in normal metals as a quadratic temperature dependence of resistivity due to electron-electron scattering, a sub-quadratic exponent is well-known signature of unconventional scattering usually attributed to strong spin fluctuations near a magnetic quantum critical point [14–17, 22, 23]. Below, we investigate the field and pressure evolution of the power-law temperature dependence of ρ_c in UTe_2 .

3) The authors characterize their samples as exhibiting $T_c=1.6$ K (in line 63) which from the perspective of available samples seems not the highest T_c achieved in this system. As one can see from the literature, samples of different “generations” have common and distinct features (such as the double transition seen in specific heat for example, see Rosa, et al., Communications Materials volume 3, 33 (2022) - published version of Ref. [7]). In order to connect the current study from a material perspective to the available literature, there should be said a bit more about the crystal characterization. For example what is the RRR? Is it known whether crystals from the same batch show double transition in specific heat?

Response: We thank the reviewer for pointing out the published reference, and Ref. [7] in the original manuscript has been updated. As the reviewer pointed out, T_c of UTe_2 can be as high as 2 K [24], and some UTe_2 samples exhibit two transitions in the heat capacity measurement [25]. While we cannot confirm the resistivity sample would exhibit the double transition feature in heat capacity, the samples with the similar $T_c \approx 1.6$ K from the same batch consistently show two features at the transition as reported in our previous work by Hayes et al. [25]. The RRR of the sample used in the current study is $R(280\text{K})/R(2.2\text{K}) = 4.3$ or $R(280\text{K})/R(0\text{K}) = 5.6$. Here $R(0\text{ K})$ was determined by extrapolating with a power-law function, $R(T) = R(0\text{K}) + AT^n$. We have included this information in the Methods section.

Line 265: The RRR of the sample used in the current study is $R(280\text{K})/R(2.2\text{K}) = 4.27$ or $R(280\text{K})/R(0\text{K}) = 5.57$. Here $R(0\text{ K})$ was determined by extrapolating with a power-law function, $R(T) = R(0\text{K}) + AT^n$.

4) The authors describe the maximum of the resistivity to be “strongly tuned by magnetic field” (line 96). It seems that the anomaly is in principle a skew peak, thus a general broadening (in

temperature) alone might shift its maximum as well (without actually changing the underlying energy scale). Can this be ruled out based on the available data such that there is a real shift of energy scale upon application of field.

Response: We agree that the observed peak is skewed or asymmetrical and the determination of the true maximum can be ambiguous. However, the comparison between 0 and 18 T data taken at 14.2 kbar in Fig. 2(e), clearly indicates the shift in peak and, hence, energy scale. We also want to point out Ref. [13] where the peak shifts to lower temperatures with increasing $H \parallel b$, which confirms the field tuning of the magnetic energy scale. To address this issue, we added the following in the revised manuscript.

Line 119: We note that the asymmetric nature of the c -axis peak, which is likely due to a convolution of scattering rates with different profiles (e.g., spin fluctuations and phonons), makes the precise determination of its field evolution difficult. The similar broadening caused by increasing field and decreasing pressure suggests an intrinsic characteristic temperature scale that can be used for analysis. Furthermore, its near equal evolution with the measured susceptibility feature in Fig. 1(c) suggests it has a common origin.

5) Fig. 3 (panel a): There is some trouble with the two blue triangles which makes it difficult to read the data. The rotation of one of the triangles does not match the legend. Please consider changing color and / or symbol to avoid confusion.

Response: We thank the reviewer for pointing out the confusion due to similar symbols. We changed the symbol types and colors, and now they are easier to discern.

6) Fig. 3, caption: The authors write for panels (d-h) “all measured pressures (...) exhibits a sudden slope change”. This should only hold for 4 of the 5 pressures, in panel (h), I cannot see a kink in the T_{sc} . Please adjust. Note this is also explained in the main text, line 153.

Response: We agree with the reviewer that there is no noticeable slope change for 14.2 kbar data. We excluded panel (h) in mentioning the slope change.

7) line 111: The authors write “also reveals interesting pressure evolution” and it is not clear what particularly is interesting on the data. Indeed, there comes a longer discussion of the data, but the authors do not stress what is the most unusual behavior. Is it the back-bending, or the increased slope (which actually is needed to go over to a back-bending and re-entrance behavior)?

Response: We agree with the reviewer that we do not stress enough what is interesting about the

pressure evolution. We revised the sentence as follows.

Line 142: The application of pressure also affects the upper critical field $H_{c2}(T)$ by inducing an unusual low-temperature non-monotonic increase of $H_{c2}(T)$ at pressures of 5.3 kbar and higher, and re-entrant behavior at 14.2 kbar as shown in Fig. 3(a).

8) line 119: One more precise reference to the figure panel could be useful here.

Response: We thank the reviewer for pointing out this issue. We added the reference Fig. 3(c) to the sentence in the revised manuscript (Line 151).

9) All figures: The authors present resistivity data and measured positions of features for different series of pressures or fields. It could be useful to have a consistent set of symbols and colors to better allow for comparison. For example Fig. 3(a) $p=0$ has black square, but Fig. 4 it is $p=5.3$ as in Fig. 2. Even within Fig. 2 the symbols change between panel (f) and (g).

Response: We thank the reviewer for pointing out the inconsistent symbols in the manuscript. We changed the symbol types and colors, and now they are consistent throughout the revised manuscript.

10) Discussion section: Can there be anything said about the connection of the energy scale detected in this experiment and the connection to superconductivity? The authors advocate for the peak in the resistivity to be associated with ω_{spin} , an energy scale for spin fluctuations. Is this consistent with the spin fluctuations responsible for superconducting pairing?

Response: The observed decreasing c -axis peak is closely related to the superconductivity in UTe_2 . As demonstrated in Fig. 1, T_c increases with pressure while T^* decreases (magnetic energy scale), which is consistent with a superconductivity-magnetism competition picture, supporting evidence for magnetically mediated superconductivity. We note that this types of T - p phase diagram were first observed in magnetically ordered Ce-based heavy fermion compounds where the application of pressure suppresses the magnetic order at a quantum critical point [22], which is often hidden in a superconducting dome. This picture now serves as a hallmark of magnetically ordered superconductivity proposed for heavy fermion [14, 15], cuprate [16] and iron-based [17] superconductors. To address this issue, we included the following in the revised manuscript.

Line 105: The observed relationship between a decreasing magnetic energy scale and strengthened superconductivity is consistent with a competitive relation, supporting the picture of magnetically mediated superconductivity in UTe_2 .

11) Note that as style for Communications materials, it is advised to have titles for all references

which simplifies the recognition of works and among others also simplifies the work of the referees.

Response: We thank the reviewer for pointing out the required bibliography style. We changed the style in accordance with Communications Materials, and now references include the title.

II. REVIEWER #2 (REMARKS TO THE AUTHOR):

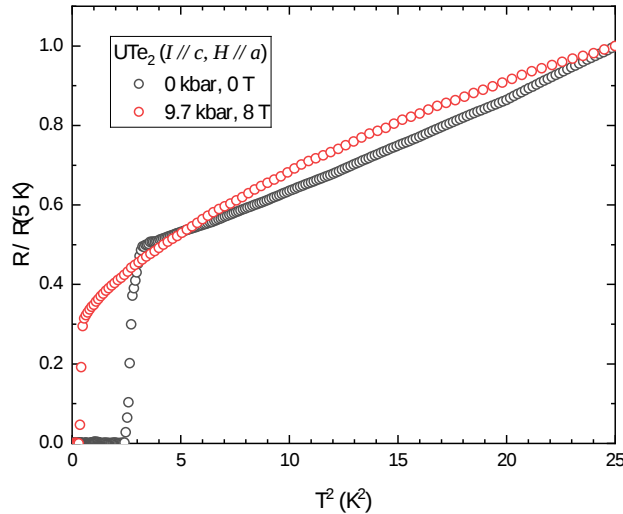
The manuscript “Tuning a magnetic energy scale with pressure and field in UTe₂” by Hyunsoo Kim and coauthors reports on the tuning of a magnetic energy scale in UTe₂, where hydrostatic pressure and a magnetic field applied along the a-axis are used as tuning parameters. A characteristic peak in the c-axis electrical transport is shown to be suppressed by increasing hydrostatic pressure and enhanced by applying a magnetic field along the a-axis. The authors show that this feature in the electrical resistivity coincides with features previously observed in the resistivity and specific heat, demonstrating that resistivity can be used to track the evolution of the underlying energy scale, which is suspected to be of magnetic origin.

Response: We thank the reviewer #2 for the accurate summary of our work and valuable comments. We respond to comments point-by-point below.

The authors also analyze the resistivity data for evidence of non-Fermi liquid (FL) behavior, by extracting a power law exponent from a double-logarithmic representation of the temperature dependent data. They argue that non-FL behavior is observed over a wide range of magnetic fields and pressures, and that it may be related to an underlying quantum critical point (QCP) in UTe₂. The power law exponents show obvious deviations from the expected T^2 temperature dependence for the resistivity of a Fermi liquid at low temperatures. However, I question whether a convincing power law can be extracted from this dataset, since it is dominated by the magnetic resistivity peak and features associated with the superconducting transition, and the data only span a temperature range from 0.5 to 14 K. In other compounds, e. g. YbRh₂Si₂, where non-FL liquid was demonstrated in connection with a QCP, a non-FL temperature dependence of the resistivity could be demonstrated over several decades in temperature, and the region of non-FL behavior was clearly centered around the QCP.

Response: We agree with the reviewer that the temperature range is somewhat limited for reliable power-law analysis at certain pressure and field ranges. However, we convincingly present the evolution of the power-law when we compare $R(T)$ with various pressure and field conditions as shown in Fig. 5. For a direct comparison of quadratic and linear $R(T)$ curves, we added a figure in the supplement (Fig. S3) and also below. We note that this issue arises because of the

superconducting transition with a relatively high T_c and the c -axis peak at low temperatures while our measurements cover sufficient ranges for both temperature and field. In such cases, the least square fitting results will sensitively depend on the bound temperature values, and the double-logarithm approach can be a more reliable method. Nonetheless, we selected the pressure and field values where a relatively wider temperature range is available, followed by performing the least square fitting to confirm the exponent deduced from the double-logarithm method. A similar approach was employed for UTe_2 in the previous works by Thomas et al. with $I \parallel a$ [8], Ran et al. with I in the ab -plane [10], and others [13, 21]. Our analysis and presentation can be directly compared to the power-law behavior reported in the aforementioned references, and, therefore, we believe the publication of our results is worthwhile for the purpose of direct comparisons. To address this issue, we incorporated the following sentences in the revised manuscript.



Line 196: Because of limited temperature range between T_c and T^* , we employ a double-logarithm fitting routine commonly used [10, 26] to produce contour plot phase diagrams to display the general trends as well as power-law fits to $R(T) = R(0) + AT^n$ to study the trends with field and pressure.

Line 217: We also performed least-squares fits on selected $R(T)$ curves by fitting our data to the relation $R(T) = R(0) + AT^n$ with $T \leq T^*/2$ well-away from the c -axis peak, selecting pressure and field values with the widest temperature ranges available.

Line 228: To directly visualize the power-law evolution, we plot $R(T)$ under selected fields and pressures that exhibit the most striking non-Fermi liquid behavior in Fig. 5. It is clear that the T^2 behavior at ambient pressure (Fig. 5(a)) evolves rapidly as a function of pressure and magnetic

field to exhibit a nearly linear behavior under $p = 9.7$ kbar and $\mu_0 H = 8$ T (Fig. 5(d)). Our power-law results with a unique current-field configuration can be directly compared to that reported in previous works [10, 13, 21, 26] and complete the picture of the power-law evolution associated with quantum criticality in UTe_2 .

I also find the color-map representation of the exponents in Fig. 4 misleading, since a wide range of exponents, e. g. from 1.25 to 1.75 is represented by virtually indistinguishable colors. To establish a power law-temperature dependence, I would find it more helpful if the authors showed actual fits of one power-law exponent to the raw resistivity data, rather than plotting the derivative of a double log plot or showing a plot of the fit parameters without plots of the fits.

Response: We thank the reviewer for pointing out the presentation issue in Fig. 4(a-e). We changed the color scheme, and they now clearly resolve the effective exponent changes. We agree with the reviewer that the display of actual power-law fitting functions will be helpful. We already performed the least square fitting with AT^n , and the fitting results are summarized in Fig. 4(f-i). In the revised manuscript, we added the fitting curves to Fig. 5, which shows representative $R(T)$ data demonstrating the evolution of Fermi-liquid to non-Fermi liquid behavior.

Finally, the manuscript is lacking a clear conclusion: it is literally absent from the text, which jumps directly from a description of the data to the methods section. The authors need to clearly establish what new information this paper contributes to the field, beyond a phenomenological description of the data. It is encouraging that the resistivity features track the previously reported magnetic energy scale, and establishing resistivity as an accessible and straightforward probe of non-FL behavior and possibly quantum criticality in UTe_2 is valuable, but the arguments need to be clearly laid out in the manuscript. Additional data (e. g. resistivity over a larger temperature range) may also be needed to clearly demonstrate non-FL behavior and a quantum critical point. In conclusion, I do not recommend publication of the manuscript in its present form, but I suggest a resubmission after addressing these concerns.

Response: We thank the reviewer for the suggestion of adding a concluding paragraph. The revised manuscript now includes a paragraph describing new information and our contribution to the understanding of normal state and superconducting properties of UTe_2 . The paragraph is also available below.

Line 236: In conclusion, we investigated the tuning of a magnetic energy scale with applied hydrostatic pressure and magnetic field along the a -axis as complementary (and opposing) tuning parameters in the heavy-fermion superconductor UTe_2 in the vicinity of a long range magnetic

ordering. The strong spin fluctuation manifest a pronounced local maximum at low temperatures, which is identified as a peak in the c -axis electrical transport. Upon increasing pressure, the characteristic c -axis peak moves to a lower temperature before vanishing near the critical pressure of about 15 kbar (1.5 GPa). The application of a magnetic field shifts the peak to a higher temperature and broadens the peak under all studied pressure values. The power-law behavior of the c -axis transport is also significantly affected by the application of pressure and field where the non-Fermi liquid behavior is most pronounced around 9.7 kbar and about 7 T, exhibiting nearly linear resistivity in temperature and an enhanced pre-factor. These signatures in c -axis transport are a revealing indication of an incipient magnetic order that has a strong influence on the physical properties and possibly superconductivity. Our results provide the pressure-and field-evolution of the magnetic energy scale relevant to quantum criticality in UTe_2 and shed light on its influence on unconventional superconductivity.

III. REVIEWER #3 (REMARKS TO THE AUTHOR):

The manuscript describes an investigation of the H-P-T phase diagram of UTe_2 , a material with high field reentrant superconductivity and a metamagnetic transition.

The authors measured conductivity along the c -axis under different pressures and magnetic fields that are aligned along the a -axis. The study is timely and sheds light on the thermodynamic of the different electronic phases that form under the application of thermodynamic variables pressure and magnetic field. While simultaneous application of both pressure and magnetic fields along different crystallographic directions in UTe_2 has been reported previously (as given in refs 13 & 18), the new information obtained in this work needs to be put into a better perspective.

Response: We thank the reviewer #3 for the positive evaluation of our work and accurate summary. As the reviewer mentioned, transport properties of UTe_2 have been studied under various configurations of pressure and field, and Refs. [27] and [28] are prominent examples. However, both references report the transport properties with the current along the a -axis whereas we study the transport properties with the current along the c -axis. Recently, Knebel et al. reported the c -axis transport with $H \parallel b$ where the field evolution of the peak is qualitatively different. Therefore, we provide unique pressure- and field-tuning of the magnetic energy scale, which sheds light on the role of magnetism in unconventional superconductivity in UTe_2 . We added the following in the introduction section to emphasize this.

Line 60: Transport properties of UTe_2 have been studied under various orientations of pressure

and field [10, 27, 28]. In this work, we study its transport properties in a unique configuration with $I \parallel c$ and $H \parallel a$ where pressure- and field-tuning of the magnetic energy scale provide insight into the role of magnetism in unconventional superconductivity in UTe_2 . Furthermore, we elucidate the presence of quantum criticality in its rich phase diagram.

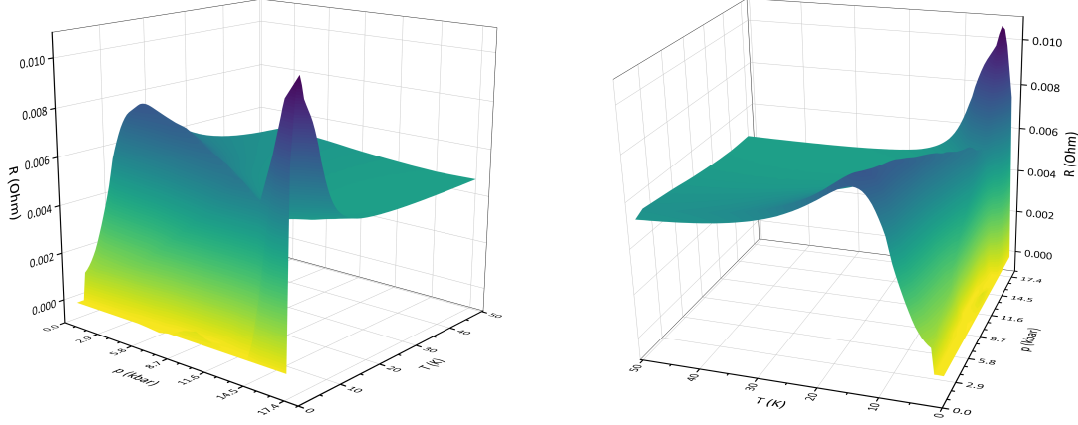
The fact that the magnetoresistivity (Fig 4b) shows a broadening transition may indicate that there is hysteretic behavior in a two-phase region. Has this been studied in more detail?

Response: As the reviewer pointed out, the transport property in UTe_2 can exhibit hysteresis behavior. We want to point out that we observed hysteresis behavior just above T_c at 6 T under 1.4 GPa with the electric current in the ab -plane (see Ref. [10]). Naturally, we looked for similar behavior in this configuration ($I \parallel c$) by measuring the resistance of both warming and cooling over the entire temperature range we studied, but we did not observe any hysteresis behavior in this case. To address the issue of possible hysteresis, we have included the following sentence in the revised manuscript.

Line 83: We note that the hysteresis behavior, previously observed in Ref. [10], was not observed in this configuration in any H - p - T space studied in this work.

The tuning aspect (see title) defines surfaces in spaces that contain pressure and magnetic field as coordinates, and some definition of the magnetic energy. It is therefore instructive to show such a surface, for instance conductivity-pressure- magnetic field, power-law-exponents – pressure – magnetic field, etc. Fig 1c could be better represented by such a surface and related projections.

Response: We thank the reviewer for the comments about efficient presentations of data. We have included 3D plots showing the pressure evolution of resistance as a function of temperature in the supplementary information (also here. See below), which clearly presents the trend. We believe the field- and pressure-evolution of the power-law behavior and the characteristic temperatures including T_c and T^* are sufficiently presented in Figs. 2 and 3. serve that purpose.



Estimates of the magnetic energy scale that is tuned should be given. What is the order of magnitude of the magnetic energy scale and what are the coupling coefficients to pressure and magnet field?

Response: The magnetic energy scale is of an order of 1 meV inferred from the observed *c*-axis peak around 12 K [1], magnetic susceptibility anomaly [5, 6], and thermodynamic response [4], which are presented in Fig. 1(c). The microscopic origin of these observations is still under debate although a Schottky-like anomaly due to crystal electric field splitting of *f*-bands as proposed in Ref. [2]. We also refer to our response to the reviewer #1's comment. The coupling coefficient of the magnetic energy with pressure can be deduced from our experiments presented in Fig. 1, and it is 0.6 K/kbar at ambient pressure, which is consistent with a thermodynamic consideration [4]. The coupling with the magnetic field can be inferred from Fig. 2(h), which is 0.6 K/T and 0.83 K/T at 5.3 kbar and 14.5 kbar, respectively. We added the following statement in the revised manuscript to include information about the various coupling coefficients.

Line 137: The coupling coefficient of the magnetic energy with pressure can be deduced from our experiments presented in Fig. 1, and it is 0.6 K/kbar at ambient pressure, which is consistent with a thermodynamic consideration [4]. The coupling coefficient with the magnetic field can be inferred from Fig. 2(h), which is 0.6 K/T and 0.83 K/T at 5.3 kbar and 14.5 kbar, respectively.

The manuscript needs a table summarizing the variables that have been obtained from the analysis of the data. This table could be placed into the supplement.

Response: We created four tables (Table I, II, III, IV) summarizing key variables obtained from analysis in the supplementary information.

The fonts in the figures are not sufficiently large to allow easy reading. The figures need to be modified.

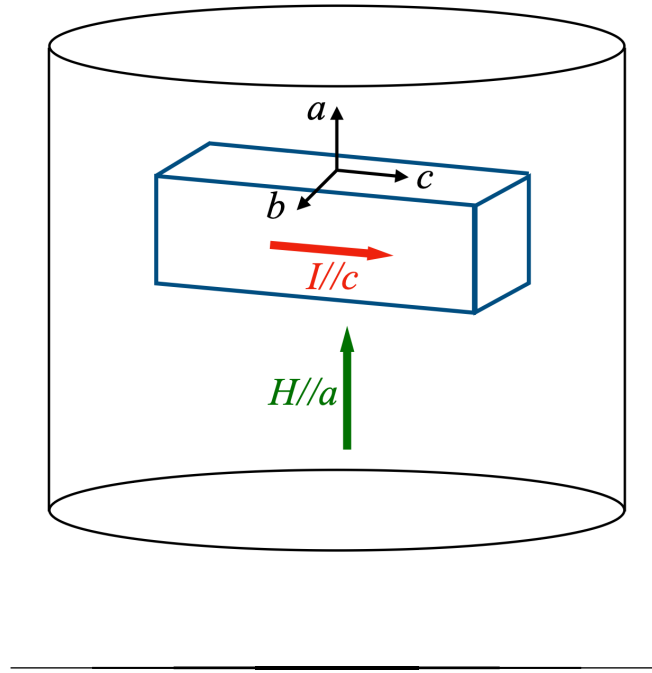
Response: We agree with the reviewer, and we used larger fonts throughout new figures for easier reading.

The pressure is given in kbar, and GPa units should be added as well.

Response: We note that $1 \text{ GPa} = 10 \text{ kbar}$. We added this conversion in the main text and expressed the pressure in both scales in the revised manuscript.

I would be helpful to the readers if the authors provide a schematic of the experimental setup indicating the orientation of the sample, dimensions, contact geometry, current and electric field direction, etc.

Response: We agree with the reviewer, and a schematic of the measurement configuration is added to Figure 1 panel (a) as well as to the supplementary information (Fig. S1) in the revised manuscript. Fig. S1 is also presented below.



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SUMMARY OF THE MAJOR CHANGES MADE:

- We revised the manuscript to comply with the reviewer #1' comments, which are indicated in magenta in the revised manuscript.
- The caption and axis label of Figure 3 have been revised for clarification.

RESPONSE TO REVIEWER'S REMARKS:

I. REVIEWER #1 (REMARKS TO THE AUTHOR):

The referee has examined the full package of rebuttal and revised manuscript/supplement where the authors have made adjustments following the comments from the previous round. Indeed, a number of findings of the referees are not completely new in the literature; proper discussion of this point and interpretation of the new details have been incorporated in the manuscript. Therefore, the referee recommends publication of the manuscript in Communication Materials.

Response: We thank the reviewer for recommending the publication of our work.

Reviewing the full content, there are some additional comments to be used to improve the manuscript further:

Response: We appreciate the additional comments and respond to them by point-to-point below.

1) page 6, line 107: The authors have added some comment regarding connection to superconductivity as requested. However, the argumentation might not be as straightforward as outlined: Indeed, the observation of decreasing magnetic ORDER and increase of T_c for superconductivity is a signature of competition. However, an increase of (the temperature scale of) magnetic fluctuations as the signatures in resistivity are interpreted might also lead to increased T_c if the cutoff frequency in BCS theory becomes larger and spin fluctuations are mediator of superconducting pairing.

Response: We appreciate the reviewer's comment. As the reviewer pointed out T_c should go up with T^* if T^* represents the energy scale of spin fluctuations directly associated with the pairing mediation. However, T^* is likely a competing magnetic energy scale which is inversely correlated with the fluctuation strength. Prominent examples of such an inverse correlation include the

heavy fermion, copper-and iron-based superconductors where the competing picture manifests a similar phase diagram. We revised the manuscript as follows to clarify that T^* is associated with a "magnetic energy scale" of the magnetic fluctuation spectrum.

Line 67: We changed "fluctuation" to "magnetic."

Line 105: We added the following to the end of the sentence: "...a common magnetic origin with an energy scale associated (inversely) with the strength of the fluctuation spectrum."

2) Fig. 3, panel (b): Indeed, this figure is to demonstrate that the $H_c(0)$ is larger than Pauli limit. However, at least for zero pressure, there is Δ_0 measurements from STM, so one could state the expected H_P for this case instead of relying on the Δ_0/T_c ratio entirely. (See also later comment.)

Response: We thank the reviewer for pointing out the measured $\Delta_0 \approx 0.25$ meV by an STM study [1]. The directly calculated H_P with the experimental Δ_0 is $H_P \approx 3$ which is in good agreement with the weak-coupling limiting case presented in Table II because $2\Delta_0/k_B T_c \approx 3.623$ in UTe₂. We note the theoretical value for the weak-coupling BCS superconductor is $2\Delta_0/k_B T_c \approx 3.528$. To incorporate the comment, we have revised the manuscript as follows.

Line 165: We added to end of sentence "...[32], which is consistent with the gap energy of $\Delta \sim 0.25$ meV observed in STM experiments [33]."

3) page 8 line 161, 164: The authors estimate H_P using the BCS ratio for Δ_0/T_c . To come up with $\alpha=1.87$ T/K. This is a very rough estimate, maybe even misleading and not applicable especially for UTe₂ which is an unconventional superconductor, has strong spin orbit coupling and is a correlated material. Recently, it has pointed out that exactly this system strongly violates expected BCS ratio, see fig 1 panel h of Nature volume 600, pages 636–640 (2021) where UTe₂ stands out. Indeed, under pressure there is no data for the gap magnitude; a violation of BCS ratio might strongly affect the estimate of H_P .

Response: We agree that there is no direct measure of gap magnitude under pressure to date, but there is also no data to indicate it may drastically deviate from zero pressure, which is nearly BCS as noted above. We also want to point out that the energy scale noted in Figure 1 in [Nature 600, 636 (2021)] is the so-called resonance energy scale from inelastic neutron scattering and is not a direct measure of the superconducting energy gap. This value needs to be distinguished from the values from STM and other spectroscopic measurements.

4) page 13, line 267: The authors have in the reply clarified that their samples are from the same batch of samples where double transitions are observed in specific heat. The information that the

resistivity sample is from the same batch of samples which show double transitions as in the work by Hayes et al. should be stated somewhere.

Response: We agree with the reviewer and have added the following sentence at the end of the first Methods paragraph.

Line 259: Samples used in this study were obtained from the same synthesis batch as used in the study of Hayes *et al.* [42].

5) similar as comment above: It might be advisable to state that the last two columns of Table II (H_HW and H_P) are calculated values (where the second has assumed BCS ratio which might be strongly violated in UTe2).

Response: As noted above, the H_P values obtained using T_c values are reasonable given the gap energy obtained by STM experiments. However, we do agree with the reviewer on noting about the theoretical estimates of H_{HW} and H_P and have revised the caption.

Caption Fig. 3: Please check whether Panels (d-h) present $H_{c2}(T)$. Seems that T_c is plotted as function of H , see label on figure.

Response: The red symbols in each panel do represent $H_{c2}(T)$ when projected to the horizontal axis. The legend indicates them as T_{sc} because we plot T_{sc} as a function of $\mu_0 H$. To eliminate the confusion, we added $H_{c2}(T)$ to the legend, and the label of the vertical axis has been changed to T_{sc} .

Typo: “UT2” $\rightarrow$ “UTe2” in caption of Fig. 2

Response: We thank the reviewer for pointing out the error. We have corrected it.

II. REVIEWER #3 (REMARKS TO THE AUTHOR):

The revised manuscript has addressed most of the requests for clarification, and is publishable in its current form.

Response: We thank the reviewer for recommending the publication of our work in Communications Materials.

[1] L. Jiao, S. Howard, S. Ran, Z. Wang, J. O. Rodriguez, M. Sigrist, Z. Wang, N. P. Butch, and V. Madhavan, Nature **579**, 523 (2020).