

Peer Review File

Manuscript Title: Cavity-mediated thermal control of metal-to-insulator transition in 1T-TaS₂

Redactions – unpublished data

Parts of this Peer Review File have been redacted as indicated to maintain the confidentiality of unpublished data.

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Reviewers' comments:

Referee #1:

Remarks to the Author:

In this work, Jarc et al. studied the insulator-metal transition (IMT) of bulk 1T-TaS₂ when the material is placed inside a mirror cavity. The IMT is probed by time-domain THz spectroscopy, where the authors used the low-frequency THz transmission or the visibility of IR phonon peaks as an indicator of metallicity. Depending on the distance between the two cavity mirrors (which determines the cavity mode frequency) and the mirror alignment (which determines the cavity quality factor), the IMT transition temperature (T_c) can be changed by tens of Kelvins compared to the free-space transition temperature. The size of the temperature hysteresis is also modified.

Overall, I find the results highly interesting, and the fact that T_c can change so dramatically by simply placing the material between a pair of mirrors separated by few millimeters is remarkable. The authors have also performed several tests to show that the results are not due to trivial scenarios. If the proposed mechanisms are true, this cavity design can in principle be applied to many other condensed matter systems to tune their phase transition temperatures.

Given the potential impact of this result, it is also important to exercise some caution to make sure the reported change in T_c is not an experimental artefact. In particular, since the authors do not measure the sample temperature directly but instead rely on the cold finger temperature reading, I am not sure whether it is T_c that changed or it is the temperature difference between the sample and the cold finger that changed. Specifically, the authors should resolve the following anomalies in their data before proceeding to publication:

1. It is well known that the CCDW-NCCDW IMT transition temperature is approximately 180K (cooling) and 220K (warming) in bulk 1T-TaS₂. However, the authors obtained 140K (cooling) and 180K (warming) in Figure 1D. The authors attributed this discrepancy to the fact that the temperature reading is located at the cold finger, so the actual sample temperature (T_{sample}) can be much higher than $T_{\text{coldfinger}}$ due to poor thermal contact and a lack of radiation shielding. Coincidentally, in Figure 4A, when the two cryo-cooled cavity mirrors are very close to each other, the cold finger readings are much closer to the expected value around 180K/220K. Then, a very simple scenario can explain the apparent change in T_c : the cryo-cooled cavity mirrors act as a radiation shield, so T_{sample} is closer to $T_{\text{coldfinger}}$ when the mirrors are closer to the sample. When the authors used 290 K mirrors instead of cryogenic mirrors (Figure S9), the apparent T_c decreases by ~40 K because the 290 K mirrors no longer act as good radiation shields. (By the way,

what is the temperature of the cryo-cooled mirrors? I was not able to find this information in the manuscript.) I encourage the authors to consider the effect of radiation shielding more carefully, especially because the mirror temperatures can dramatically change the apparent sample temperature, as demonstrated in Figure S9.

2. Related to the previous point, it is very strange that Figure S13C and S13D and the inset of Figure 4A show that the sample temperature T_{sample} is lower than the cold finger temperature $T_{\text{coldfinger}}$; in these plots, the quantity $1 - T_{\text{sample}}/T_{\text{coldfinger}}$ is positive. Given the poor thermal contact between the cold finger and the sample, I would expect $T_{\text{sample}} > T_{\text{coldfinger}}$ (it looks like Equation (10.6) has a sign problem). Besides this issue of the relative value of T_{sample} and $T_{\text{coldfinger}}$, based on Figure S13 and Figure 4A inset, the quantity $(1 - T_{\text{sample}}/T_{\text{coldfinger}})$ is too small to explain the T_c change of the sample. Since the authors also acknowledged that the cavity-induced free energy change has a negligible effect in Supplementary Section 9, my understanding is that neither of the two scenarios proposed by the authors (free energy change and Purcell-effect) provides a realistic picture in explaining the dramatic “renormalization” of T_c .

3. Another anomaly in the data is in Figure 4A and 4B, where the free-space data points (at 0 GHz) have a discontinuous jump from the first data point around 10 GHz. One should expect the free-space data points to be smoothly connected to the low-cavity-frequency data points. Why caused the discontinuity? And why is the trend in Figure 4B non-monotonic? How reproducible are the trends in Figure 4A and 4B across different samples? These anomalous behavior seems incongruent with a cavity-induced effect, and can possibly result from experimental artefacts.

To rule out these artefacts and other anomalies mentioned in previous points, I think it is important to consider an independent measurement of the actual sample temperature conducted in situ instead of relying on the cold finger reading. This temperature measurement is challenging for the cavity geometry but still possible, and here are a couple ideas to consider: (i) Raman thermometry (comparing Stokes and anti-Stokes spectra), and (ii) cryo-condensation of gases (using tabulated saturation pressure curves of common gases, whose condensation on the Si₃N₄ layers can be detected by some absorption measurement)

Besides the above concerns about the actual sample temperature, I encourage the authors to consider the following points when re-visiting their experiments and data:

4. The quality of the sample needs to be re-examined because it displays such a large temperature width of the IMT transition in both cooling and warming directions (Figure 1D), which is almost 20 K. For a bulk sample used in the experiment, the first order transition between CCDW and NCCDW is typically very sharp (see reference 32 or any other R-T curve of bulk 1T-TaS₂). The authors argued that the large width stems from inhomogeneity, so the sample must contain an extremely high degree of inhomogeneity or disorder. I wonder whether the large width is also present if the authors perform resistivity-vs-temperature measurement on the same sample or the same batch of samples. On this note, the authors should provide information about how the single crystals are grown (presumably it is a single crystal? The manuscript didn't specify whether a single crystal or a polycrystal is used), which can give further clues about why the transition width is anomalously large.

5. The presentation and analysis of the time-domain THz data are unusual/sloppy. For example, the authors should consider presenting the actual THz conductivity or dielectric function instead of transmission in Figures 1 to 4, especially if the authors would like to compare the low-frequency conductivity. The vertical axis of the time-domain THz trace in the insets of Figure 1D and 3B are also not labeled.

6. The authors seem to have some general misunderstanding of the phase transitions in 1T-TaS₂. For example, the authors mentioned throughout the manuscript that “domain wall fluctuations” are responsible for the metallic behavior of NCCDW while domain walls are locked to the crystal structure in the CCDW phase, therefore making it insulating. What are the “domain walls”? In the C-CDW state, there are no domain walls unless the authors are referring to the “hidden” state of 1T-TaS₂ induced by an optical or electrical pulse, which is clearly not the case here. In the NC-CDW state, I presume the domain walls refer to the discommensurate region, but what are the domain wall fluctuations? It is unclear how the authors arrived at the conclusion that only the discommensurate regions in the NC-CDW phase are conducting while the commensurate regions are insulating. Related to this, how did the authors obtain the 15 GHz central frequency for the “domain wall fluctuations”? Can this number be associated with some known parameters about the low-frequency conductivity of the NC-CDW phase of TaS₂? As another example, the authors mentioned that the metal-to-IC transition is characterized by “polaronic transport”. This description is quite unusual because the resistivity barely changes at 550K (Nature Materials 7, 960 (2008)) and the IC phase remains fairly metallic with no polaronic character. The authors are encouraged to revise these claims and provide relevant references.

Lastly, the authors are encouraged to consider the following points when revising their manuscript:

7. The Landau free energy for 1T-TaS₂ has been well studied since the 1970s (e.g. J. Phys. Soc. Jpn. 43, 1839 (1977), Structural Phase Transitions in Layered Transition Metal Compounds, ed. Motizuki (1986)), so the sketch in Figure 1B looks too simplistic and it is unclear what the order parameter refers to (e.g. the angle between the CDW wavevector and the crystallographic axis?)

8. For each pair of data in Figure 4A, the authors should consider showing the raw temperature sweep curves in the Supplementary Information. Importantly, the authors should demonstrate the reproducibility of the hysteretic curves as a function of temperature (Figure 4C) and mirror separation (Figure 3B) by showing consecutive scans.

9. What is the lateral size of the sample? What is the beam spot size of the THz light at the sample position? The authors are encouraged to specify the information in the Supplementary Material.

10. The authors are encouraged to provide the raw time-domain data corresponding to the processed frequency-domain plots in Figure S2.

In summary, I believe the results are groundbreaking and are suitable for publication in Nature if the authors can show beyond reasonable doubt that cavity-induced T_c change is not an experimental artifact. At the moment, an artifact scenario cannot be safely ruled out especially because the two

theoretical mechanisms provided by the authors are too far off in explaining such a large renormalization of T_c . The central point here is a reliable measurement of the sample temperature, which can be a bit tricky but is nevertheless essential in ruling out the experimental artifact and accounting for the unexplained anomalies in the current dataset.

Referee #2:

Remarks to the Author:

G. Jarc et al. report the observation of a controllable insulator-to-metal transition in bulk 1T-TaS₂ embedded in a Fabry-Pérot THz cavity. The claim is substantiated by the emergence of a renormalized hysteresis in the sample's THz conductivity when the material's electromagnetic environment is modified by the action of the cavity. The authors explore different resonant conditions by mechanically tuning the distance and relative angle between the cavity mirrors. Time-domain THz transmission data suggest that cavity mode frequencies up to 25 GHz promote a transition to the (conducting) near-commensurate CDW phase, whereas cavity mode frequencies around 500 GHz favor the stabilization of the (insulating) commensurate CDW phase.

The THz cavity design is simpler than the Fabry-Pérot configuration developed by the Rice University group [Q. Zhang et al., Nat. Phys. 12, 1005 (2016); X. Li et al., Nat. Photonics, 324 (2018)] but allows the authors to perform a reliable temperature dependence over a wide temperature range. It is known that similar Fabry-Pérot cavity designs can only lead to a very low Q factor compared to other cavity architectures (the Q factor achieved by the authors is indeed only 3-4 at the explored frequencies). Surprisingly, such a low Q factor seems to be sufficient to induce a sizable change of the material's thermodynamics. The acquired THz transmission data are of high quality, the analysis is rigorous, and possible artifacts are discussed in detail. If confirmed, this discovery would represent a relevant step forward in the cavity-mediated control of quantum material functionalities. However, there are several aspects that the authors should address thoroughly before the paper can be considered fit for publication in Nature.

1) I am not satisfied with the microscopic interpretation provided in the discussion section. The authors propose that their data can be rationalized through a preferential coupling between the cavity modes and charged domain wall fluctuations (modeled as a broad continuum absorption feature around 15 GHz). However, there is no experimental signature that points toward this scenario, and the authors do not perform a systematic scrutiny of other effects that may play a role. The theoretical analysis is phenomenological and the continuum absorption at 15 GHz may not even exist in practice or may have any other origin (e.g., the damped pseudo-Goldstone CDW phason – which is electric dipole allowed in centrosymmetric systems such as bulk 1T-TaS₂ and has large oscillator strength – or other collective deformation modes of the CDW superlattice). The authors should clarify the GHz response of 1T-TaS₂ to draw meaningful conclusions. For example, the authors should measure the temperature-dependent imaginary part of the GHz conductivity in the bare crystal (i.e., outside the cavity), check whether an absorption feature exists in the commensurate phase, and study how its line shape changes with temperature. As the system's metallic response above the transition temperature resembles a Drude-Smith behavior rather than a conventional Drude, GHz collective modes may not be entirely screened even in the metallic phase.

Similar measurements were commonly performed in the 1980s to assign the low-energy collective modes of various one-dimensional CDW systems [G. Grüner, *Rev. Mod. Phys.* 60, 1129 (1988), T. W. Kim et al., *Phys. Rev. B* 40, 5372 (1989)]. As I could not find any reference for 1T-TaS₂ in the spectral range of interest (the closest example being Y. Ma et al., *Phys. Rev. B* 97, 195117), I believe that the authors must include this important piece of information. Finally, performing the same experiments with the material kept in the cavity would provide more conclusive insights into the microscopic mechanism underlying the authors' observations (e.g., strong coupling limit with the phason – which would explain why a low cavity Q factor can generate such a dramatic effect on the hysteresis).

2) The authors should also comment on the shape of the transmitted THz spectrum when the cavity mode frequency is tuned above 200 GHz. Even though cavity resonances at 200 GHz lie well above possible material excitations of interest, it would be important to establish whether specific signatures of light-matter coupling directly emerge in the measured THz spectrum. As the THz probe beam generated by the photoconductive antenna has sufficient intensity over the 0.2-2.5 THz range, this check should be rather straightforward. I could not find these data anywhere in the manuscript.

3) In the nascent field of cavity engineering of quantum materials, some papers could not find their way to publication because the results were not reproducible. How many 1T-TaS₂ samples did the authors measure inside the cavity? Were the results reproducible? It is important to specify this aspect in the main text of the paper and compare spectra acquired from different samples in the Supplementary Information. Finally, the authors should add a few words on the synthesis and preparation process of the sample.

4) While the temperature-dependent hysteresis of Figure 3A reflects the Q factor changes with cavity alignment, the effect leading to the results of Figure 3B is not straightforward to me. Why would the cavity opening and closing cause hysteresis in the integrated THz transmission? Can the authors clarify this aspect more explicitly in the main text of the paper?

Minor points:

1) In the description of the material's low-temperature commensurate CDW phase, I suggest replacing the term "Mott insulator" simply with "insulator." This is to account for the unsettled debate about the nature of this state (i.e., spin-frustrated Mott vs. spin-singlet band insulator). See for example Y. D. Wang et al., *Nat. Commun.* 11, 4215 and many other recent papers. For the same reason, when the authors mention that the ground state of bulk 1T-TaS₂ is driven by the interplay among the Coulomb repulsion, lattice strain, and Fermi surface nesting, they should also include the interlayer hopping.

2) I suggest that the insets of Figure 3A also display the transition temperatures of the free-space material as a reference.

3) The axes of the right panels in Figure 2 are incorrect. The values should span the range 11-15 if the transmission is expressed in %. The same applies to the vertical axes of the two panels in Figure 3 and in Figure 4C.

4) The authors did not cite relevant literature on THz and sub-THz cavities embedding materials with macroscopic quantum effects [e.g., F. Appugliese et al., *Science* 375, 1030 (2022)].

5) All the error bars reported in the figures should be defined in the corresponding legends. I could not find any description of how the error bars were estimated in Figures 3 and 4. The authors should also specify the temperature stability range of their cryostat during a measurement at a fixed temperature. I assume this value to be negligible. However, if it turned out to be significant, all data should show an error bar along the temperature axes.

Referee #3:

Remarks to the Author:

This study reports the control of a phase transition in the dichalcogenide 1T-TaS₂ through the coupling to a GHz Fabry-Perot cavity. The bare material shows a hysteresis curve between 140 and 180 K. In the current experiment, this transition is traced via the change of the low frequency transmission of a terahertz pulse, which traces the presence of free carriers. As shown in Fig. 2, regardless of whether the material is in the metallic or the insulating phase, the insertion of the material into the GHz cavity does not noticeably change the optical response of the system in this frequency regime, which lies substantially above any GHz cavity resonance. Remarkably, however, by tuning the fundamental cavity resonance (i.e. by moving the Fabry-Perot mirrors relative to the sample), the authors raise or lower the temperature range of the hysteresis curve by an astonishing amount - between 110-135 K (at a cavity frequency of around 15 GHz) to 185-210 K (for a cavity frequency above 100 GHz). At any cavity frequency, the temperature range shrinks substantially from 38 K in the bare material to 23-33 K in the cavity-coupled complex.

This cavity-induced shift of the hysteresis behaviour, and in particular its incredible magnitude, is entirely unexpected and I cannot think of any previous work that would even hint at such an amazing impact of low-frequency cavities on correlated materials. The experimental setup is conceptually very simple and the authors carefully rule out alternative explanations of these measurements. It is - together with the experiments by the group of Jerome Faist (*Science* 375, 6584 (2022)) - the only 'clean' demonstration to date of a cavity altering the macroscopic electronic properties of a solid state system. Therefore, I anticipate that this study will be a major breakthrough for the cavity control of quantum materials, and will have a tremendous impact on the future development of the field. However, as I detail below, the authors should provide additional data at specific points, if possible, and clarify their conclusions, to further substantiate their conclusions:

- In particular, in Fig. 3a, the authors explore the transition from the cavity-controlled to the free space regime through the gradual misalignment of the cavity mirrors. Two important questions on this transition should be addressed:

1) It is shown that the misalignment moves the hysteresis curve towards the free space limit. But even at an angle of almost 10 degrees, there is still an enormous deviation from the free space hysteresis curve of around 20 K. Are there technical reasons that restrict the authors from going to larger values angles and explore the vanishing of the remaining discrepancy?

In this regard, the authors only provide measurements of the cavity resonance change in the supplementary material. It would be helpful if they could provide measurements of the cavity Q-factor as a function of the misalignment, and demonstrate that it is indeed the vanishing of the cavity resonance which is responsible for recovering the free space limit.

2) There should be a second route to recover the free space limit - by increasing the distance between the mirrors and sending the resonance frequency to zero (as also sketched in Fig. S11). Therefore, one would expect that, in Fig. 4a, the critical temperatures should approach the free space limit at small cavity frequencies. Yet this is the regime where the authors observe the largest shifts in these critical temperatures. This to me is perhaps the most perplexing of the results presented in this manuscript and it should be discussed carefully. At large frequencies the critical temperatures appear to arrive at some kind of saturation regime, but there is no such limit visible at small frequencies.

Would it be possible to increase the data range of Fig. 4a to even smaller frequencies? How do the authors explain this startling phenomenon?

- Two possible theoretical explanations are provided for the experimental observations - one based on a hybridization scenario, where the cavity couples to domain wall fluctuations in the metallic phase to form new eigenstates and thus change the free energy, and one based on the Purcell effect, which effectively decouples said domain wall fluctuations from the electromagnetic environment when the photonic density of states is reduced at their respective resonance frequency. Moreover, in Section S9 of the supplementary material, the authors argue convincingly that the first scenario is insufficient to explain the observations, as the phase space volume that can be affected by the direct cavity coupling is small compared to the full system. They conclude that "this puts more emphasis on the second mechanism". Thus, they rule out a strong coupling scenario and instead favour a weak coupling scenario. But it is not apparent why the same phase space argument should not apply to this Purcell scenario, too. If strongly coupled cavity modes cannot affect the free energy, why should weakly coupled modes be able to achieve this?

The changes made to the manuscript are listed at the end of each answer.

Notes: the numbering of figures and equations in the following refers to previous versions of the manuscript and the supplemental materials.

We provide below a table indicating the references to the final version of the work.

Previous numbering (used in the Referees'reply)	Current numbering
Fig. S1	Extended Data Fig. 1
Fig. S2	Extended Data Fig. 2
Fig. S3	Extended Data Fig. 3
Fig. S4	Extended Data Fig. 4
Fig. S5	Extended Data Fig. 5A
Fig. S6	Extended Data Fig. 5B,C
Fig. S7	Extended Data Fig. 6
Fig. S8	Extended Data Fig. 7
Fig. S9	Extended Data Fig. 8
Fig. S10	Extended Data Fig. 9
From Fig. S11 to Fig. S33	From Fig. S1 to Fig. S23
From Eq. 3.1 to Eq. 3.19	From Eq. 1 to Eq. 19 (Methods)
From Eq. 10.1 to Eq. 10.7	From Eq. 20 to Eq. 26 (Methods)

Referee #1 (Remarks to the Author):

In this work, Jarc et al. studied the insulator-metal transition (IMT) of bulk 1T-TaS₂ when the material is placed inside a mirror cavity. The IMT is probed by time-domain THz spectroscopy, where the authors used the low-frequency THz transmission or the visibility of IR phonon peaks as an indicator of metallicity. Depending on the distance between the two cavity mirrors (which determines the cavity mode frequency) and the mirror alignment (which determines the cavity quality factor), the IMT transition temperature (T_c) can be changed by tens of Kelvins compared to the free-space transition temperature. The size of the temperature hysteresis is also modified.

Overall, I find the results highly interesting, and the fact that T_c can change so dramatically by simply placing the material between a pair of mirrors separated by few millimeters is remarkable. The authors have also performed several tests to show that the results are not due to trivial scenarios. If the proposed mechanisms are true, this cavity design can in principle be applied to many other condensed matter systems to tune their phase transition temperatures.

Given the potential impact of this result, it is also important to exercise some caution to make sure the reported change in T_c is not an experimental artefact. In particular, since the authors do not measure the sample temperature directly but instead rely on the cold finger temperature reading, I am not sure whether it is T_c that changed or it is the temperature difference between the sample and the cold finger that changed. Specifically, the authors should resolve the following anomalies in their data before proceeding to publication.

We thank the Referee for the positive assessment of the manuscript and for acknowledging the potential impact of our work among the condensed matter community. We greatly appreciate the effort the Referee put in reviewing the manuscript and providing valuable feedbacks.

In fairness, we agree with the referee that given the novelty of the experiment, the surprising and somewhat counterintuitive results, extra caution should be exerted as the strength of the manuscript relies on the robustness of the experimental evidence reported. It is therefore crucial to safely rule out the possibility of any experimental artifacts. This is in fact what led us, already in the first version of the work which we submitted, to perform several tests to exclude potential artifacts and consolidate the measurements. It took us more than one year to design and perform all the tests which convinced us of the genuine nature of the observation and the extent of the supplementary information file associated to the first manuscript submission gives an indication of the carefulness of our assessment.

The issue that the Referee raises about the temperature readout of the experiment is indeed the crucial aspects of the work. As he/she states, all the measurements presented in the manuscript refer to the cold finger temperature, and not directly to the actual temperature of the sample. Given the design of the cavity chamber, the choice of measuring the temperature on the outside of the cavity was due not only to the difficulty of finding an alternative method to constantly monitor the temperature at the sample position while performing the THz measurements, but also to the fact that the introduction of any object in the cavity could potentially perturb the electromagnetic environment and lead to modified experimental conditions.

The possibility raised by the Referee of a mismatch between the cold finger's and the sample's temperature is legitimate and was already considered as one of the two scenarios proposed in the previous version of the manuscript. The reshaping of the electromagnetic density of states within the cavity could in fact affect the thermal load of the sample and thus modify its temperature. In this regard, in **the manuscript previously submitted we focused on delivering the evidence that** irrespectively of which is the physical mechanism leading to the observations (heating or free energy renormalization), **the effect is due to the cavity electrostatics and is not related to incoherent radiative energy exchange between the sample and the environment.** For this reason, we performed experiments in different experimental condition, including hot and cold mirrors, misaligned cavity mirrors, etc...

The reviewer is correct in stating that the lack of a systematic way to monitor the sample's temperature previously prevented us from discriminating whether the observed change in the insulator-metal transition temperature was due to a modified free energy landscape leading to a renormalization of the critical temperature or to a modified thermal emission leading to a temperature shift.

In the current manuscript we also address this question, through a twofold approach:

- We have developed a micrometric Cr-Al junction which can be placed within the membranes (details are given in SM) and performed additional experiments to directly measure the temperature of the sample in different experimental conditions. Nevertheless, we stress that the motivation of our original choice of avoiding placing mechanical object in the cavity not to perturb the cavity is still relevant: the presence of the thermocouple does not allow for a simultaneous THz measurements and will perturb the cavity geometry, so all the temperature measurements within the cavity must be considered as an independent set of data which corroborate the overall scenario, but cannot be compared directly to the THz measurements. Importantly, to test the reproducibility of the experimental findings with different samples, all the temperature measurements have been performed on a different 1T-TaS₂ belonging to a different batch.
- We have carried out finite elements simulations with the COMSOL software (details in SM) to describe how the sample's temperature is modified by incoherent radiation cooling due to purely geometrical factors related to the cavity settings. In particular, we simulated the thermal profile of a free-standing

silicon nitride membrane whose edges have the same temperature as the cold finger. We studied how the temperature in the middle of the membrane (and thus the temperature at the sample position) changes by introducing in the simulations the cavity mirrors and studying different cavity geometries.

The results of these additional tests suggest that the scenario in which the cavity control the thermal load on the sample and therefore its temperature is the most plausible one. The temperature measurements within the cavity consolidate the evidence already collected via THz spectroscopy: we observed the same dependence of the sample's temperature as function of the cavity fundamental mode and cavity alignment.

Importantly, these trends confirm that the results in long cavities are not reproduced in the finite elements simulations, confirming that the cavity-mediated effects cannot be explained by simple geometrical arguments that may lead to a modified temperature at the sample's position via incoherent radiation heating.

We will detail in the following the additional results that we now included in the supplementary materials and provide a point-by-point reply to the Referee's concerns.

1. It is well known that the CCDW-NCCDW IMT transition temperature is approximately 180K (cooling) and 220K (warming) in bulk 1T-TaS₂. However, the authors obtained 140K (cooling) and 180K (warming) in Figure 1D. The authors attributed this discrepancy to the fact that the temperature reading is located at the cold finger, so the actual sample temperature (T_{sample}) can be much higher than $T_{\text{coldfinger}}$ due to poor thermal contact and a lack of radiation shielding. Coincidentally, in Figure 4A, when the two cryo-cooled cavity mirrors are very close to each other, the cold finger readings are much closer to the expected value around 180K/220K. Then, a very simple scenario can explain the apparent change in T_c : the cryo-cooled cavity mirrors act as a radiation shield, so T_{sample} is closer to $T_{\text{coldfinger}}$ when the mirrors are closer to the sample. When the authors used 290 K mirrors instead of cryogenic mirrors (Figure S9), the apparent T_c decreases by ~40 K because the 290 K mirrors no longer act as good radiation shields. (By the way, what is the temperature of the cryo-cooled mirrors? I was not able to find this information in the manuscript.) I encourage the authors to consider the effect of radiation shielding more carefully, especially because the mirror temperatures can dramatically change the apparent sample temperature, as demonstrated in Figure S9.

We thank the Referee for raising this point, which is indeed of the critical aspect of the manuscript. As the Referee states, the possibility of a temperature change in the sample due to the thermal shielding of the cavity mirrors had been already considered in the previous version of the manuscript in which we tested how the temperature of the cavity mirrors modifies the measured transition temperature. The temperature of the cryo-cooled mirrors in the experiments is 95 K and it is the minimum attainable in the current configuration. We have now added this information which, as noted by the Referee, was previously missing in the supplementary materials.

The fabrication of the Cr-Al thermocouple allowed us to repeat these tests and monitor how the temperature of the sample (T_{int}) deviates from the temperature of the cold finger (T_{ext}).

For the sake of clarity, we address step by step the concerns raised by the Referee.

- The referee is correct in the observation that the measured transition temperature in free space is 140 K (180 K) upon cooling (heating), and thus is lower than the nominal critical temperature by about 40 K, is actually due to the poor thermal contact between the cold finger and the sample. This was confirmed by directly measuring T_{int} in free space, i.e. by sealing the Cr-Al junction in thermal contact with the sample and removing the cavity mirrors. We observed that, when $T_{\text{ext}} = 80$ K (minimum achievable temperature), the temperature of the sample is approximately 40 K higher (black triangles in Fig. S11, now included in the revised version of the supplementary materials). The difference $T_{\text{int}} - T_{\text{ext}}$ gradually vanishes when T_{ext} approaches the room temperature. This is easily quantitatively explained assuming that the sample is

subject to radiation heating from the environment and the cooling capacity of the cold finger is limited by the thermal conductance of the membranes.

This observation is quantitatively in agreement with finite elements simulations, in which we found that, by fixing the temperature at the edges of the membrane to $T_{\text{ext}} = 180$ K, the temperature in the middle of the membrane (i.e., at the sample position) is higher. **We quantitatively confirm the referee's hypothesis that the mismatch between the nominal and the measured free-space transition temperature is then due to the incoherent thermal load on the sample. An important observation is that the effective phase transition temperature observed with long cavities goes below the values obtained in free space and this cannot be rationalized as simple radiation shielding.**

- We simulated the cavity configuration by placing the membrane between two cavity mirrors at tunable temperature and distance. By fixing both the mirrors' and the cold finger's temperature at 180 K, we simulated how the temperature in the middle of the membrane (which is approximately 220 K in free space) changes as function of the cavity length (Fig. S16). As expected when the cavity fundamental frequency increases (i.e., the mirrors are closer to the sample) the temperature at the sample's position decreases, until it reaches 180 K at the minimum cavity length considered (500 μm , that is 300 GHz). This trend is exactly what the Referee pointed out in Fig. 4A, where the transition temperatures upon cooling and heating approach the nominal ones for higher cavity frequencies. This is expected because, as also the Referee points out, the cryo-cooled mirrors effectively shield the radiation and reduce the thermal load in the middle of the membrane. This trend is also confirmed by the temperature measurements within the cavity (Fig. S18A), in which it is clear that $T_{\text{int}} - T_{\text{ext}}$ is smaller for shorter cavities.

On the other hand, when the temperature of the mirrors is increased to 300 K, we find different behaviors. In the simulations with cavity mirrors at room temperature (Fig. S17), we found that the temperature in the middle of the membrane increases when the mirrors are closer, and it reaches 260 K for the 300 GHz cavity. This means that the mirrors efficiently shield the ambient radiation only when they are at cryogenic temperatures; the mirrors are instead responsible for an increased thermal load on the sample when they are at room temperature.

However, this is not compatible with the results of the THz measurements. In fact, according to a scenario in which the sample's temperature is only affected by the incoherent radiation shielding, we would have expected that, upon closing the cavity with the 290 K mirrors, the apparent transition temperature would have been lower for smaller cavities. We observed instead similar to the one observed with cold mirrors: the measured transition temperatures are higher when the mirrors are closer to the sample, similarly to the trend measured with the mirrors at cryogenic temperatures. This is clear in Fig. S15C, in which we show that the temperature of the mirrors is only responsible for a rigid shift of the measured transition temperatures by about 40 K, but does not affect the overall trend as function of the cavity fundamental frequency.

This observation is confirmed by the new experiments where we measure the temperature within the cavity (Fig. S18C) which show that, regardless of the temperature of the mirrors, the temperature difference between the sample and the cold finger is reduced at higher cavity frequencies (namely, the sample gets colder). **This finally proves that the renormalization of the sample's temperature as function of the cavity length cannot be explained by incoherent thermal heating alone**, which is nevertheless responsible for the overall offset of the transition temperatures with hot and cold mirrors that the Referee correctly highlighted.

- Furthermore, we emphasize that **the radiation shielding cannot explain the renormalization of the measured transition temperature that we observed by changing the alignment of the cavity** (Fig. 3). We simulated the thermal profile of the membrane enclosed in two mirrors having different alignments. We found that, even at the bigger tilting angle studied in the experiment (9.5°), the temperature in the middle of the membrane is not affected by the alignment of the mirrors (Fig. S21). This trend was also confirmed by the temperature measurements within the cavity which show that, by fixing both the cold

finger temperature and the cavity length, the temperature of the membrane is stable against the alignment of the cavity mirrors (Fig. S22). We nevertheless measured a change in the sample's temperature upon tilting the mirrors, which decreases when the misalignment is maximum (Fig. S22). We stress again that the temperature of the mirrors does not affect this trend, which - except for the rigid shift discussed above - was retrieved also with the mirrors at room temperature (Fig. S19).

Finally, we considered the effect that the ambient radiation shielding may have on the sample's temperature by shielding the chamber with metallic foils. We measured the local temperature of the sample as a function of the cavity fundamental mode, and we observed no difference in the measured sample's temperature with and without the shield (Fig. S24). This test confirms that **the geometrical screening of the ambient radiation does not play a significant role in the renormalization of the sample's temperature.**

- **Finally, the important observation is that the effective phase transition temperature observed with long cavity goes below the values obtained in free space and this cannot be rationalized as simple radiation shielding.**

To conclude, **the additional measurements and simulations that we performed suggest that the radiation shielding, albeit being responsible for an overall offset in the measured sample's temperature, cannot explain the cavity-mediated effects that we reported, that are genuinely due to the cavity electrostatics.**

We included all the measurements discussed here in the supplementary materials. In order to improve the readability of the manuscript we have reorganized the supplementary materials. We have divided the SM in two parts. In the first part, a general description of the tests done, the calculation results, and methodologies is given, while in the second part we address specific questions that the reader may have regarding the issues discussed in the manuscript. Furthermore, we replaced Fig. 4B in the revised manuscript: instead of the difference of the critical temperature upon cooling and heating as function of the cavity fundamental mode, we added the direct measurement of the temperature of the sample performed with the Cr-Al junction for different cavity lengths. This measurement is in fact key for concluding a cavity mediated heating of the sample is indeed the most likely scenario explaining the experimental evidence.

2. Related to the previous point, it is very strange that Figure S13C and S13D and the inset of Figure 4A show that the sample temperature T_{sample} is lower than the cold finger temperature $T_{\text{coldfinger}}$; in these plots, the quantity $1-T_{\text{sample}}/T_{\text{coldfinger}}$ is positive. Given the poor thermal contact between the cold finger and the sample, I would expect $T_{\text{sample}} > T_{\text{coldfinger}}$ (it looks like Equation (10.6) has a sign problem). Besides this issue of the relative value of T_{sample} and $T_{\text{coldfinger}}$, based on Figure S13 and Figure 4A inset, the quantity $(1-T_{\text{sample}}/T_{\text{coldfinger}})$ is too small to explain the T_c change of the sample. Since the authors also acknowledged that the cavity-induced free energy change has a negligible effect in Supplementary Section 9, my understanding is that neither of the two scenarios proposed by the authors (free energy change and Purcell-effect) provides a realistic picture in explaining the dramatic "renormalization" of T_c .

We thank the Referee for spotting out this mistake. We apologize for the sign problem in previous Equation 10.6, which has been now fixed in the revised supplementary materials (currently, Equation 3.19). As correctly stated by the Referee, the temperature of the sample is expected to be higher than the temperature of the cold finger.

To facilitate the discussion of the results obtained in the Purcell-like model, we plotted in Fig. S7C and Fig. S7D (former Fig. S13), the temperature ratio $T_{\text{int}}/T_{\text{ext}}$ (rather than $1-T_{\text{int}}/T_{\text{ext}}$) as a function of the cavity fundamental frequency. We observe that the calculated temperature ratio correctly predicts that i) for higher cavity frequencies (i.e., smaller cavities), T_{int} approaches T_{ext} ; ii) at a fixed cavity length, $T_{\text{int}}/T_{\text{ext}}$ is larger for lower cold finger temperatures.

Both trends are consistent with the direct measurement of the sample’s temperature within the cavity. In Fig. R1 we plot the temperature of the sample for different cavity fundamental frequencies. These measurements are the same ones displayed in Fig. S11A and Fig. 4B, but here we have preferred to plot T_{int} against T_{ext} (rather than the temperature difference $T_{\text{int}}-T_{\text{ext}}$) for facilitating the comparison with the model. We observe that the sample and the cold finger have the same temperature (approaching the dashed black diagonal) when T_{ext} is close to room temperature; moreover, the deviation of T_{int} from T_{ext} is smaller (regardless of the value of T_{ext}) for higher cavity frequencies.

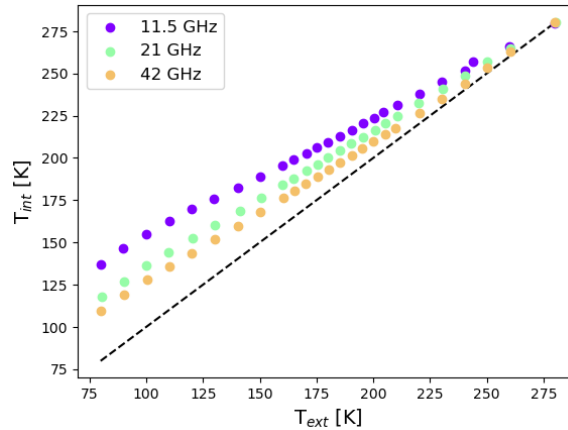


Figure R1: Measured temperature of the sample (T_{int}) against temperature of the cold finger (T_{ext}) at three different cavity lengths.

We emphasize once again that all these features can be qualitatively understood in terms of the proposed Purcell-like scenario. From a quantitative point of view, we stress that the calculated temperature ratio depends on the thermal coupling $Q_{\text{ext-int}}$ between the cold finger and the sample (Fig. S7D), which nevertheless does not affect the dependence of $T_{\text{int}}/T_{\text{ext}}$ on the cavity fundamental frequency. According to the model, the renormalization of the sample’s effective temperature is expected to be larger when the thermal coupling between the sample and the cold finger is poorer. Quantitative discrepancies between the model and the experimental data can be thus ascribed to the choice of the model parameters.

Importantly, the temperature measurements that we performed by means of the Cr-Al junction provided strong evidence that the cavity mediated heating (Purcell-like) model is in qualitative agreement with the experimental evidence.

We fixed the sign problem in the equation highlighted by the Referee and change the vertical axes in Fig. S7C and S7D. We included the temperature measurements within the cavity (Fig. S11) which are qualitatively consistent with the predictions of the second scenario.

3. Another anomaly in the data is in Figure 4A and 4B, where the free-space data points (at 0 GHz) have a discontinuous jump from the first data point around 10 GHz. One should expect the free-space data points to be smoothly connected to the low-cavity-frequency data points. Why caused the discontinuity? And why is the trend in Figure 4B non-monotonic? How reproducible are the trends in Figure 4A and 4B across different samples? This anomalous behavior seems incongruent with a cavity-induced effect and can possibly result from experimental artefacts.

We thank the Referee for pointing this out. This is indeed a crucial observation that deserves to be thoroughly discussed.

In principle, the Referee is right, and it would be reasonable that the free-space data points should be smoothly connected to the lowest cavity frequency data, as also Referee#3 pointed out. In fact, longer cavities would be expected to couple more weakly to the material and asymptotically approach the free-space configuration. However, the data in Fig. 4A suggest that this is not the case for cavity frequencies smaller than ~15 GHz, in which the measured transition temperature is smaller than the free-space one upon heating and cooling the sample. This means that, depending on the cavity-mediated scenario that we consider, higher frequency cavities are either responsible for a significant decrease of the critical temperature with respect to the free-space case or they prompt a renormalization of the temperature of the sample, which would then be hotter in long cavities than in free space. The trend is reversed for cavity frequencies greater than ~15 GHz.

In this regard, both the finite elements simulations and the temperature measurements within the cavity helped to clarify this point. The intuitive picture that longer cavities data should be connected to free-space data without discontinuous jumps is in fact confirmed by the simulations. In Fig. S16C it is clear that, when the membrane is enclosed into cryogenic mirrors, the temperature in the middle of the membrane is lower than the free-space configuration. This trend is consistent also for higher frequency cavities, for which the temperature at the sample's position monotonically approaches the cold finger's temperature. As already discussed above, this result can be understood in terms of a more efficient screening of the thermal radiation that is ensured by moving the cavity mirrors closer to the membrane.

By sealing the Cr-Al junction within the membranes (without the sample), we confirmed this general trend. In Fig. S11B it is clear that the temperature of the membrane enclosed in the cavity is always lower than the one measured in free space, regardless of the cavity frequency.

However, we measured a different trend for the temperature of the sample (Fig. S11A). In fact, when the junction is mounted within the membranes and in thermal contact with the sample, the sample's temperature for the lowest cavity frequency (11.5 GHz) is higher than the free-space case. The trend is then reversed for the 21.4 and 42.8 GHz cavities. The temperature measurements within the cavity thus confirm the non-monotonic trend that we already reported via THz spectroscopy. In order to highlight the robustness of this evidence, we now included in the revised manuscript (new Fig. 4B) the direct measurement of the sample's temperature within the cavity at different fundamental frequencies. Please note that we removed the plot that was previously displayed in Fig. 4B, namely the difference between heating and cooling of the measured critical temperatures as a function of the cavity frequency. This information is in fact already implicitly provided in Fig. 4A.

We stress that the non-monotonic behavior for the temperature inside the cavity, for which longer cavities are heating the sample with respect to free space temperature while shorter cavities are cooling it (Fig. S11A), is not observed in an experiment without the sample (Fig. S11B) which instead gives a monotonic behavior in which the presence of the cavity reduces the sample temperature through screening. The non-monotonic trend in temperature is indeed an effect which depends on the presence of the sample in the cavity.

Finally, we highlight once again that the temperature measurements have been carried out on a different samples, belonging to a different batch. The fact that we qualitatively observed the same trend as the one measured via THz spectroscopy in a different sample assures the reproducibility of the experimental findings.

The evidence collected so far does not allow to fully understand the origin of this anomalous behavior of the sample. In this regard, measurements with cavity lengths larger than the one used in the present work could help to shed light on this effect. Unfortunately, we could not test yet these configurations because of the geometrical constraints of the vacuum chamber, which needs to be completely redesigned to enable measurements with longer cavities.

Importantly, the last aspect that we want to highlight is that, although anomalous, the non-monotonic trend reported in Fig. 4A - and confirmed by the temperature measurements in current Fig. 4B - is a further proof that the cavity-mediated effects reported in the work are genuinely due to the cavity electrodynamics. In fact, as also the COMSOL simulations demonstrated, incoherent radiation heating could not explain this effect and would instead cause a temperature renormalization that is opposite in sign to the one experimentally observed.

We performed temperature measurements on a sample belonging to a different batch and confirmed the same non-monotonic trend observed via THz spectroscopy (comparison Fig. 4A and 4B). We included Fig. S11 and S20 which confirm that this is a sample-dependent effect.

To rule out these artefacts and other anomalies mentioned in previous points, I think it is important to consider an independent measurement of the actual sample temperature conducted in situ instead of relying on the cold finger reading. This temperature measurement is challenging for the cavity geometry but still possible, and here are a couple ideas to consider: (i) Raman thermometry (comparing Stokes and anti-Stokes spectra), and (ii) cryo-condensation of gases (using tabulated saturation pressure curves of common gases, whose condensation on the Si₃N₄ layers can be detected by some absorption measurement)

We thank the Referee for these suggestions. We acknowledge the importance of the additional temperature measurements that the Referee required. Indeed, as detailed above, the local measurement of the sample's temperature turned out to be crucial to substantiate our experimental findings.

Before assembling the Cr-Al junction that we actually used to monitor the sample's temperature, we considered the ideas suggested by the Referee. Initially, Raman thermometry was our first choice since it has the obvious advantage of leaving unperturbed the cavity geometry. We attempted Raman measurements in cavity in collaboration with the group of Prof. Paul van Loosdrecht in Cologne. The main limitations were posed by the cavity geometry which required the implementation of macro-Raman measurements. Unfortunately, the beam intensity needed to obtain a sufficient signal-to-noise ratio was high enough to melt the membranes. Raman measurements were thus unfeasible in the current cavity setup.

For this reason, we designed the micrometric Cr-Al junction, that we successfully embedded in the membranes. The results of the temperature measurements of the sample are in qualitative agreement with the THz measurements and confirm the main trends reported in the manuscript: the sample's temperature depends on the cavity fundamental frequency and the cavity alignment. We also demonstrated that the effects are peculiar of the sample and are not due to a modification of the membrane's local temperature. In particular, the temperature measurements confirmed the non-monotonic trend of T_{int} (but not T_{membrane}) as function of the cavity frequency (Fig. S11), which surprisingly shows that the temperature of the sample enclosed in long cavities is higher than its temperature in free space. This trend is reversed for cavity frequencies greater than ~15 GHz, where the sample's temperature is lower with respect to the free-space configuration. Remarkably, the renormalization of the sample's temperature due to the cavity alignment – observed in both THz and temperature measurements - cannot be explained by simple geometrical arguments and is related to the cavity electrodynamics.

Finally, it is worth noting that minor quantitative differences between THz and temperature measurements may be due to the presence of the thermocouple itself which could introduce an additional heat load. However, this effect is expected to be small because of the tiny diameter of the thermocouple's wires (~10 μm).

Besides the above concerns about the actual sample temperature, I encourage the authors to consider the following points when re-visiting their experiments and data:

4. The quality of the sample needs to be re-examined because it displays such a large temperature width of the IMT transition in both cooling and warming directions (Figure 1D), which is almost 20 K. For a bulk sample used in the experiment, the first order transition between CCDW and NCCDW is typically very sharp (see reference 32 or any other R-T curve of bulk 1T-TaS₂). The authors argued that the large width stems from inhomogeneity, so the sample must contain an extremely high degree of inhomogeneity or disorder. I wonder whether the large width is also present if the authors perform resistivity-vs-temperature measurement on the same sample or the same batch of samples. On this note, the authors should provide information about how the single crystals are grown (presumably it is a single crystal? The manuscript didn't specify whether a single crystal or a polycrystal is used), which can give further clues about why the transition width is anomalously large.

We thank the Referee for pointing this out. We now included a dedicated section in the supplementary materials to the synthesis of the samples. The samples used in the experiments are single crystals and have been grown using the vapor phase transport method.

Comparisons of transition widths require precise knowledge of all experimental parameters. Apart from the impurity and interstitial S concentration, the width depends on the rate of temperature change during the measurement, temperature inhomogeneity and substrate strain. To obtain a sharp transition temperature with thick single crystals, the rate of cooling or heating needs to be < 0.1 K/minute. Often in the literature the cooling rate is not sufficiently slow to obtain sharp transitions.

A more important factor in thin films is substrate strain, which plays a large role in the transition, and can shift and broaden the transition temperature substantially (<http://dx.doi.org/10.7567/APEX.7.103201>). When mounted on membranes, additionally, homogeneity of the temperature (particularly in-plane) may also broaden the transition.

None of these effects have any bearing on the shifts of T_C discussed in this paper. In fact, the well-documented sensitivity of the discommensuration melting transition temperature to external influences speaks in favor of the observed effects being genuine.

We also highlight that the temperature width that we measured does not depend on the observable (i.e., THz transmission) used to track the phase transition. We derived the real part of the optical conductivity from the raw THz fields measured in free space upon heating and cooling the sample (Fig. S28 and S29) and found that the low-energy conductivity (which tracks the degree of metallicity in the sample) undergoes the phase transition with the same temperature width reported in Fig. 1C.

Furthermore, in order to prove that this is not an anomaly related to a specific sample, we carried out temperature dependent THz measurements in free space in a different sample (having similar thickness) belonging to the same batch. As shown in Fig. S32, apart from a tiny change in the critical temperature, we did not observe any substantial change in the temperature width of the transition.

We included a sample preparation section (in supplementary materials), mentioned in the main manuscript that the strain may also contribute to the temperature width of the transition, compared the temperature-dependent conductivity and transmission (Fig. S28 and S29), and performed free-space measurements on a different sample (Fig. S32).

5. The presentation and analysis of the time-domain THz data are unusual/sloppy. For example, the authors should consider presenting the actual THz conductivity or dielectric function instead of transmission in Figures 1 to 4, especially if the authors would like to compare the low-frequency conductivity. The vertical axis of the time-domain THz trace in the insets of Figure 1D and 3B are also not labeled.

We thank the Referee for this comment. We acknowledge that the analysis of our time-domain THz data may seem unusual. However, the choice of discussing transmission instead of conductivity/dielectric function was intentionally made to minimize data manipulation.

The profound reason behind this choice is that, in order to obtain a quantitatively reliable estimation of the optical conductivity of the sample within the cavity, it is also necessary to consider the conductivity of the empty cavity (i.e., the membranes and the cavity mirrors) as a reference. On the one hand, this means that for every measurement performed in a specific cavity setting (cavity length, cavity alignment and temperature), a corresponding measurement without the sample should be available. On the other hand, this does not seem the right way to approach the study of light-matter hybrids. In fact, by isolating the response of the sample from the one of the empty cavity, information on the collective light-matter system may be lost. In order to avoid possible errors in the quantitative determination of the THz conductivity of the sample inside the cavity, we thus preferred to directly plot the THz transmission.

To address the Referee's concern, we compared in Fig. S28 the THz transmission measured in free space upon heating and cooling the sample with the calculated real part of the THz optical conductivity. Furthermore, we plot in Fig. S29 the hysteretic curves obtained by integrating the low-frequency THz spectrum of both the transmission and the optical conductivity. This comparison clarifies that, at energies below the lowest-lying phonon, the transmission is indeed a good indicator of the metallicity of the system.

Finally, we apologize for not labeling the vertical axes of the time-domain THz traces in the insets of Fig. 3B. We have fixed this issue in the revised manuscript. Regarding Fig. 1D, it displays just a qualitative trend of the free energy and the units of measure are arbitrary.

We added Fig. S28 and S29 and labeled the axes in Fig. 3B.

6. The authors seem to have some general misunderstanding of the phase transitions in 1T-TaS₂. For example, the authors mentioned throughout the manuscript that “domain wall fluctuations” are responsible for the metallic behavior of NCCDW while domain walls are locked to the crystal structure in the CCDW phase, therefore making it insulating. What are the “domain walls”? In the C-CDW state, there are no domain walls unless the authors are referring to the “hidden” state of 1T-TaS₂ induced by an optical or electrical pulse, which is clearly not the case here. In the NC-CDW state, I presume the domain walls refer to the discommensurate region, but what are the domain wall fluctuations? It is unclear how the authors arrived at the conclusion that only the discommensurate regions in the NC-CDW phase are conducting while the commensurate regions are insulating. Related to this, how did the authors obtain the 15 GHz central frequency for the “domain wall fluctuations”? Can this number be associated with some known parameters about the low-frequency conductivity of the NC-CDW phase of TaS₂? As another example, the authors mentioned that the metal-to-IC transition is characterized by “polaronic transport”. This description is quite unusual because the resistivity barely changes at 550K (Nature Materials [7, 960 \(2008\)](#)) and the IC phase remains fairly metallic with no polaronic character. The authors are encouraged to revise these claims and provide relevant references.

We apologize for the confusion that the description of the phase transitions in 1T-TaS₂ may have generated. We believe that this is due to a poor choice of the wording that, following the Referee's suggestions, we have now revised. However, we would like to stress that the focus of our work is reporting the cavity-mediated effects on the insulator-to-metal transition. In this respect, we are aware that the experimental findings that we report do not allow to shed light on the physical mechanisms that drive the phase transitions in 1T-TaS₂, in which many other degrees of freedom (not considered in our description) are known to play a major role.

The word “domain wall fluctuations” was used throughout the manuscript to indicate a material's mode whose strong response to ac-currents could justify a significant coupling to the cavity modes. We stress that this choice,

along with the identification of the resonance at 15 GHz, was entirely phenomenological and led by i) the experimental observation in our data of a resonance centered approximately in that frequency range and ii) the fact that the conductivity measured in the MHz range seems to increase above 10 MHz (<https://doi.org/10.1103/PhysRevB.97.195117>, <https://doi.org/10.1016/j.ssc.2020.113946>, papers that we now included in the reference list), suggesting the presence of a resonance at higher frequencies. The lack in literature of conductivity data in the GHz range makes it difficult to be sure about both the actual presence of a resonance and its physical origin. However, we stress that this does not jeopardize the validity of the phenomenological free-energy model and its conclusions, as any active mode in that frequency range could potentially couple to the cavity.

To avoid unnecessary confusion, we cleaned the wording in the section regarding the physics behind the phase transitions in 1T-TaS₂ and added the reference suggested by the Referee. Moreover, we removed the word “domain wall fluctuations” throughout the manuscript to further emphasize that the model’s predictions are independent of the physical meaning of the active material’s mode and have instead a more general validity. We reckon that these changes allow to put more emphasis on the experimental observations that are at the core of the manuscript and, at the same time, leave open to further investigations the underlying microscopic description.

Lastly, the authors are encouraged to consider the following points when revising their manuscript:

7. The Landau free energy for 1T-TaS₂ has been well studied since the 1970s (e.g. J. Phys. Soc. Jpn. 43, 1839 (1977), Structural Phase Transitions in Layered Transition Metal Compounds, ed. Motizuki (1986)), so the sketch in Figure 1B looks too simplistic and it is unclear what the order parameter refers to (e.g. the angle between the CDW wavevector and the crystallographic axis?)

We thank the Referee for this comment. We agree that the sketch of the Landau free energy in Fig. 1B may look too simplistic for a material such as 1T-TaS₂. The aim of the sketch was in fact to show the rationale of the experiment, without entering into the details of the sample’s specific properties that, as the Referee also recognizes, are complex and widely studied in the literature. For clarity, the order parameter in the previous sketch referred to the charge density modulation.

In order to avoid confusion, we have revisited the structure of the manuscript. We included a general introduction on the effects that embedding a quantum material in an optical cavity may have on its macroscopic properties.

Without limiting our discussion only to the insulator-to-metal phase transition in 1T-TaS₂, we provide two possible scenarios that may unfold: i) a selective cavity-mediated renormalization of the thermodynamic potentials, leading to modified control parameters of a given phase transition in the material; ii) a cavity-mediated modification of the material’s emission spectrum, leading to a modified effective temperature. We stress that these two scenarios have general validity and the simple sketch of the free energy landscape across a phase transition (current Fig. 1A, bottom left panel) should be considered representative of the broad class of phase transitions that may occur in quantum materials.

We hope that this explains our choice of sketching a simple free energy diagram and that the changes made in the revised manuscript meet the favor of the Referee.

8. For each pair of data in Figure 4A, the authors should consider showing the raw temperature sweep curves in the Supplementary Information. Importantly, the authors should demonstrate the reproducibility of the hysteretic curves as a function of temperature (Figure 4C) and mirror separation (Figure 3B) by showing consecutive scans.

We have included the temperature sweep curves for each pair of data in Fig. 4A in the supplementary materials (Fig. S27). We have also demonstrated the reproducibility of the hysteretic curves as a function of mirror separation and temperature by separately showing consecutive scans.

We plotted in Fig. S30 the THz fields upon opening and closing the cavity mirrors for three consecutive scans, corresponding to the integrated data in Fig. 3B. The time between a scan and the next is approximately 10 minutes: the waiting time before starting the acquisition is ~5 minutes and the acquisition itself lasts ~5 minutes more. The hysteretic curves (Fig. S30B) show similar behavior, indicating that the results are stationary state and not related to the dynamics of the phase transition.

We have also included the separate scans for the hysteretic curves in Fig. 4C. In Fig. S31, we compare three consecutive scan (~10 minutes) acquired by heating and cooling the sample in two different cavity lengths (337 and 16.7 GHz). The hysteretic curves are equivalent in all the scans.

We included Fig. S27, S30, S31 in the supplementary materials.

9. What is the lateral size of the sample? What is the beam spot size of the THz light at the sample position? The authors are encouraged to specify the information in the Supplementary Material.

We apologize for not specifying this information in the manuscript. The lateral size of the sample is approximately 4 mm x 4 mm, while the spot size of the THz beam at the sample's position is approximately 1.5 mm.

We have now included these experimental details in the revised supplementary materials.

10. The authors are encouraged to provide the raw time-domain data corresponding to the processed frequency-domain plots in Figure S2.

Following the Referee's suggestion, we have now added the raw time-domain data in Fig. S2. For each frequency-domain plot in Figure S2B, we included the time-domain THz fields, which show the multiple reflections within the tunable Fabry-Pérot cavity and the cavity dissipations responsible for the attenuation of subsequent reflections.

In summary, I believe the results are groundbreaking and are suitable for publication in Nature if the authors can show beyond reasonable doubt that cavity-induced T_c change is not an experimental artifact. At the moment, an artifact scenario cannot be safely ruled out especially because the two theoretical mechanisms provided by the authors are too far off in explaining such a large renormalization of T_c . The central point here is a reliable measurement of the sample temperature, which can be a bit tricky but is nevertheless essential in ruling out the experimental artifact and accounting for the unexplained anomalies in the current dataset.

We thank again the Referee for their insightful comments. We deeply appreciate the careful evaluation of our work and the suggestions for the improvement. We hope that the new evidence provided, along with additional datasets and the changes made to the revised manuscript, will address the Referee's concerns and substantiate the robustness of the experimental findings which remain at the core of the present work.

Referee #2 (Remarks to the Author):

G. Jarc et al. report the observation of a controllable insulator-to-metal transition in bulk 1T-TaS₂ embedded in a

Fabry-Pérot THz cavity. The claim is substantiated by the emergence of a renormalized hysteresis in the sample's THz conductivity when the material's electromagnetic environment is modified by the action of the cavity. The authors explore different resonant conditions by mechanically tuning the distance and relative angle between the cavity mirrors. Time-domain THz transmission data suggest that cavity mode frequencies up to 25 GHz promote a transition to the (conducting) near-commensurate CDW phase, whereas cavity mode frequencies around 500 GHz favor the stabilization of the (insulating) commensurate CDW phase.

The THz cavity design is simpler than the Fabry-Pérot configuration developed by the Rice University group [Q. Zhang et al., Nat. Phys. 12, 1005 (2016); X. Li et al., Nat. Photonics, 324 (2018)] but allows the authors to perform a reliable temperature dependence over a wide temperature range. It is known that similar Fabry-Pérot cavity designs can only lead to a very low Q factor compared to other cavity architectures (the Q factor achieved by the authors is indeed only 3-4 at the explored frequencies). Surprisingly, such a low Q factor seems to be sufficient to induce a sizable change of the material's thermodynamics. The acquired THz transmission data are of high quality, the analysis is rigorous, and possible artifacts are discussed in detail. If confirmed, this discovery would represent a relevant step forward in the cavity-mediated control of quantum material functionalities. However, there are several aspects that the authors should address thoroughly before the paper can be considered fit for publication in Nature.

We thank the Referee for providing a detailed and constructive feedback on our manuscript and for acknowledging the relevance of our work in the community.

Before moving into the details, we would like to inform the Referee that the revised manuscript contains additional measurements. In particular, by sealing a micrometric junction within the membranes, we were able to directly measure the sample's temperature and thus monitor its cavity-mediated changes. The purpose of these measurements was to discriminate between the two theoretical interpretations that we proposed in the manuscript. The temperature measurements qualitatively confirmed the same anomalous trend measured via THz spectroscopy and thus further validated our experimental evidence. Importantly, the additional measurements suggested that the Purcell-like scenario is the dominant effect, as we measured an appreciable cavity-controlled renormalization of the sample's temperature with respect to the cold finger's one. We included in the supplementary materials further experimental tests and also finite elements simulations (ran with the COMSOL software) corroborate the evidence.

Overall, the temperature measurements (which have been performed on a sample belonging to a different batch) are consistent with the previous results. The body of evidence that we collected hints at a scenario in which the presence of the cavity triggers a renormalization of the emission spectrum, and thus of the internal temperature, of the sample.

We provide below a point-by-point answer to the Referee's concerns.

1) I am not satisfied with the microscopic interpretation provided in the discussion section. The authors propose that their data can be rationalized through a preferential coupling between the cavity modes and charged domain wall fluctuations (modeled as a broad continuum absorption feature around 15 GHz). However, there is no experimental signature that points toward this scenario, and the authors do not perform a systematic scrutiny of other effects that may play a role. The theoretical analysis is phenomenological and the continuum absorption at 15 GHz may not even exist in practice or may have any other origin (e.g., the damped pseudo-Goldstone CDW phason – which is electric dipole allowed in centrosymmetric systems such as bulk 1T-TaS₂ and has large oscillator strength – or other collective deformation modes of the CDW superlattice). The authors should clarify the GHz response of 1T-TaS₂ to draw meaningful conclusions. For example, the authors should measure the temperature-dependent imaginary part of the GHz conductivity in the bare crystal (i.e., outside the cavity), check

whether an absorption feature exists in the commensurate phase, and study how its line shape changes with temperature. As the system's metallic response above the transition temperature resembles a Drude-Smith behavior rather than a conventional Drude, GHz collective modes may not be entirely screened even in the metallic phase. Similar measurements were commonly performed in the 1980s to assign the low-energy collective modes of various one-dimensional CDW systems [G. Grüner, *Rev. Mod. Phys.* **60**, 1129 (1988), T. W. Kim et al., *Phys. Rev. B* **40**, 5372 (1989)]. As I could not find any reference for 1T-TaS₂ in the spectral range of interest (the closest example being Y. Ma et al., *Phys. Rev. B* **97**, 195117), I believe that the authors must include this important piece of information. Finally, performing the same experiments with the material kept in the cavity would provide more conclusive insights into the microscopic mechanism underlying the authors' observations (e.g., strong coupling limit with the phason – which would explain why a low cavity Q factor can generate such a dramatic effect on the hysteresis).

We thank the Referee for this comment and appreciate their concerns. We agree with the Referee that it would be very beneficial to include the temperature-dependent conductivity in the GHz range. To this aim, we contacted different collaborators to try to perform such measurements but, unfortunately, we could not get access to the expertise needed to measure the response in the GHz range.

However, we will argue in the following that the lack of this information does not jeopardize our manuscript as the main focus of the manuscript remains the experimental observation of a reversible cavity-control of the metal-to-insulator transition in 1T-TaS₂, regardless of the microscopic mechanisms that drive the huge temperature renormalization that we observed. As the reviewer correctly highlights, the free-energy model that we proposed is phenomenological and does not aim at a microscopic description of the coupling mechanism between 1T-TaS₂ and cavity, but rather at giving a phenomenological picture for which low frequency coupling could give rise to a similar effect. In this respect, the nature of the specific resonance coupled to the cavity is beyond the scope of the paper. The theoretical interpretations that we proposed aim at showing that with some reasonable assumptions for the response of the material, we could predict the experimentally observed trends and stimulate further investigations in this direction.

In this respect, the idea of modelling a resonance centered at 15 GHz was stimulated by two main arguments: i) the experimental observation of a resonance in that region upon detuning the cavity (former Fig. 4B); ii) the fact that other conductivity measurements in the MHz range (as the one indicated by the Referee) indicates that the dielectric response increases at frequencies larger than 10 MHz (<https://doi.org/10.1103/PhysRevB.97.195117>, <https://doi.org/10.1016/j.ssc.2020.113946>) which would put a high and low frequency constraints to the resonance observed.

Finally, it is important to note that the free-energy model that we developed gives a qualitatively similar trend irrespective of the details and shape of the low frequency resonance and of the microscopic nature of the material's excitation coupled to the cavity. The only requirement is that its dielectric response is strong enough to justify a coupling to the cavity mode. So, as also the Referee suggested, the involvement of domain wall fluctuations is not conclusive and other degrees of freedom (the CDW phason or in general other collective modes with sufficient oscillator strength) may play a role.

In order to clarify those aspects, we revised the manuscript, clarified the spirit of the model, and removed the microscopic assignment of the excitation in the GHz range involved in the coupling. This choice emphasizes the general validity of the theoretical model.

2) The authors should also comment on the shape of the transmitted THz spectrum when the cavity mode frequency is tuned above 200 GHz. Even though cavity resonances at 200 GHz lie well above possible material excitations of interest, it would be important to establish whether specific signatures of light-matter coupling directly emerge

in the measured THz spectrum. As the THz probe beam generated by the photoconductive antenna has sufficient intensity over the 0.2-2.5 THz range, this check should be rather straightforward. I could not find these data anywhere in the manuscript.

We thank the Referee for this comment. The Referee's remark is correct and there is evidence that when the cavity is tuned in resonance with the phonon modes in the dielectric phase strong coupling features are observed. In fairness, we started this set of experiments aiming at studying the strong vibrational coupling between optical cavities and phonon modes across the metal-insulator transition temperature. We soon realized that the larger effect on the apparent transition temperature giving access to a cavity control of the metal insulator transition is observed for low-frequency cavities and we focused the attention of the current manuscript on this aspect.

In order to show evidence of the coupling between 1T-TaS₂ and high-frequency cavities, we share confidentially with the Referees some unpublished results of the study conducted by tuning the cavity to higher frequency.

As the Referee correctly highlighted, some signatures of strong light-matter coupling directly emerge in the transmitted THz spectrum. In particular, we explored the regime in which the cavity is tuned in resonance with the phonons of the commensurate charge-density-wave (C-CDW) phase. As shown in Fig. [REDACTED]. The strong coupling between the three phonons (left panel) and the single cavity mode (right panel) results in the formation of four non-degenerate hybrid states: an upper and a lower polariton (UP, LP) and two middle polaritons (P1, P2).

The THz transmission measurements in the coupled system (blue curve in Fig. [REDACTED]) confirm the presence of the hybrid polaritonic mixing, which disperse with the cavity detuning (Fig. [REDACTED]).

Furthermore, we have evidence that the even cavities tuned at lower frequency ($0.2 \text{ THz} < \omega < 1.1 \text{ THz}$), and thus in resonance with the Drude excitation of the material, can strongly couple to the system and consequently modify the THz transmitted spectrum. In particular, we observed that the coupling gives rise to modified cavity dissipative rates which are sensitive to the 1T-TaS₂ phase.

[REDACTED]

[REDACTED]

We have decided not to include those high frequency studies in the current manuscript because the large renormalization of the effective phase transition happens with low-frequency cavities and is not affected by the strong coupling with optically active phonon modes.

3) In the nascent field of cavity engineering of quantum materials, some papers could not find their way to publication because the results were not reproducible. How many 1T-TaS₂ samples did the authors measure inside the cavity? Were the results reproducible? It is important to specify this aspect in the main text of the paper and compare spectra acquired from different samples in the Supplementary Information. Finally, the authors should add a few words on the synthesis and preparation process of the sample.

We agree with the Referee that the reproducibility of the data across different samples is of key importance for validating our findings.

To this aim, we have tested different samples belonging either to the same batch as the sample measured in the submitted manuscript or to a different batch. The additional measurements that we carried out, and that will be detailed below, confirmed that all the samples display similar trends.

First of all, a legitimate point that may be raised (as also Referee#1 pointed out in question#4) is that the details of the hysteretic behavior observed even in free space (such as the critical temperature and the temperature width of the transition) may depend on the quality of the sample. In this regard, it should be noted that the details of the transition may strongly depend on the features of the individual sample, such as the strain due to the lateral extension of the sample and the presence of inhomogeneities.

We tested the free-space THz transmission upon heating and cooling two different samples belonging to the same batch. The results of the measurements are reported in Fig. S32, where we compared the transmission integrated in the range 0.2-1.5 THz for sample 1 (the one used in the measurements reported in the main manuscript) and

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sample 2 (another sample having similar thickness). Except for a small change in the critical temperature, we did not observe any substantial difference in the hysteretic curves measured in the two samples.

Secondly, we confirmed that all the cavity-mediated effects that we reported are reproducible in a sample belonging to a different batch. In particular, as anticipated above, we directly measured the sample's temperature within the cavity by means of micrometric Cr-Al junction sealed within the membranes. In this respect, the choice of using an alternative observable - which nevertheless confirmed the effects already measured via THz spectroscopy - further proved the general validity of our results.

As mentioned above and thoroughly discussed in the updated supplementary materials, the temperature measurements were decisive in discriminating between the two scenarios proposed in the manuscript. In fact, we revealed that the experimental evidence can be rationalized in terms of a Purcell-like effect that is responsible for a cavity-mediated modification of the thermal emissivity of the sample. This means that the change in the critical temperature that we observed via THz spectroscopy can be equally reflected in a temperature shift, which is detectable via local temperature measurements.

The main experimental results that we reported in the manuscript, and that are reproducible via the additional temperature measurements that we provide, are:

- i) A renormalization of the effective critical temperature as function of the cavity fundamental mode (Fig. 4A). In particular, we observed that the measured critical temperature (both upon cooling and heating the sample), is lower in longer cavities and approach the nominal value for shorter cavity lengths. Importantly, the free-space data points are not smoothly connected to the larger cavities, but there is a discontinuous jump that we further emphasized in the revised manuscript by choosing a different marker for the free-space data.
The same trend was qualitatively reproduced via temperature measurements in a different sample. In Fig. 4B, we included the results of the measurements. In particular, the inset of Fig. 4B clearly shows the same discontinuous jump of the free-space data already observed via THz. We stress that this trend is peculiar of the sample and is not observed when the thermocouple only measures the temperature variation of the bare membranes (Fig. S11B).
- ii) A renormalization of the effective critical temperature as function of the cavity alignment (Fig. 3A and Fig. S33). In particular, we observed that the critical temperature is lower (i.e., deviates more from the nominal value) when the cavity quality factor is higher. In the same fashion, we found that, also in the other sample, the temperature renormalization is more prominent when the cavity is perfectly aligned (Fig. S22).

To conclude, the experimental findings are found to be qualitatively reproducible in samples belonging to different batches. Eventual small quantitative differences may be attributed to a different thickness of the samples or to the thermal load possibly introduced by the thermocouple mounted in the cavity.

Finally, all the samples that we used are single crystals and have been grown using the vapor phase transport method.

We included a new extensive section in the supplementary materials to provide details on the sample preparation process. We added Fig. S32 and all the temperature measurements performed on a sample belonging to a different batch.

4) While the temperature-dependent hysteresis of Figure 3A reflects the Q factor changes with cavity alignment, the effect leading to the results of Figure 3B is not straightforward to me. Why would the cavity opening and

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closing cause hysteresis in the integrated THz transmission? Can the authors clarify this aspect more explicitly in the main text of the paper?

We thank the Referee for pointing this out. We understand that the presentation of the data in Fig. 3B might have been confusing. We will clarify here the content of the figure. Moreover, we modified the main text of the paper and the figure itself accordingly. In particular, we swapped the colors of the hysteretic curves in the main panel for consistency and changed the order of the THz fields in the insets in order to reflect more clearly how the measurements were performed. We insert below in Fig. R4 the updated figure.

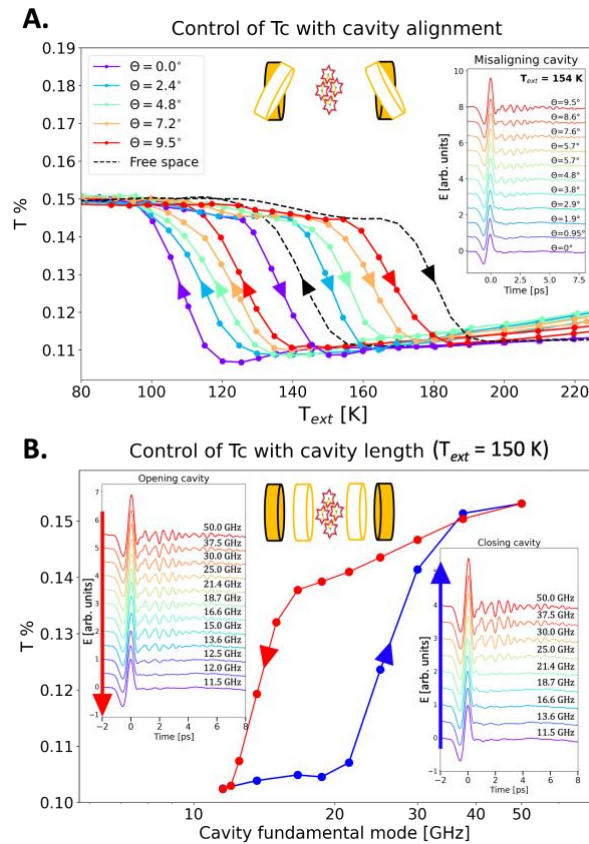


Figure R4: Updated Fig. 3 in the revised manuscript.

The data have been acquired by fixing the temperature of the cold finger at 150 K and recording the THz field transmitted by the cavity, as plotted in the insets. The first data point acquired is the one corresponding to the 11.5 GHz cavity (purple curve in the right inset) which shows that the system is in the metallic state. By integrating the low-frequency THz transmission, this corresponds to the leftmost blue point in the main panel. Upon closing the cavity (blue arrow), the system turns into an insulator (rightmost blue point) at a given cavity frequency (~25 GHz). The opposite behavior is observed when, starting from the smaller cavity (50 GHz, red curve in the left inset), the cavity length is increased. Upon closing the cavity (red arrow), the metallic state is retrieved (leftmost red point in the main panel).

The hysteretic behavior arises from the fact that the critical cavity frequency that marks the metal-to-insulator transition is different in the two directions. We can rationalize this evidence based on the temperature measurements that we carried out within the cavity, which confirmed that the cavity boundary conditions are [Type here]

indeed responsible for a renormalization of the emission spectrum of the sample and thereby coherently control its temperature through cavity electrodynamics. Fig. S11A shows that the local temperature of the sample is increased for long cavities; conversely, the sample is cooled down below the free-space reference for smaller cavity lengths. In this framework, closing the cavity can be considered as an effective temperature scan: by starting from a “hot” metallic state at 11.5 GHz, the system is cooled down when the cavity frequency is increased up to 50 GHz and it turns into an insulator. Upon opening the cavity, the local temperature of the sample is increased, and the system goes back to the metallic state. The cavity frequency-dependent hysteresis thus maps the well-known temperature dependent hysteresis of the sample.

We modified Fig. 3B and added the following paragraph in the main text to clarify this aspect.

“The cavity frequency-dependent hysteresis in Fig. 3B can be rediscussed in light of the temperature measurements reported in Fig. 4B. By keeping fixed the cold finger temperature the local temperature of the sample decreases upon increasing the cavity frequency (see for example Fig. S18). Therefore, closing the cavity effectively corresponds to cooling down the sample that is thus driven to the insulating state (blue curve in Fig. 3B). The effect is reversed when, starting from the insulating state, the cavity frequency is decreased (red curve). Closing and opening the cavity can then be interpreted as an effective temperature scan, therefore leading to the hysteretic behaviour observed in Fig. 3B.”

Minor points:

1) In the description of the material’s low-temperature commensurate CDW phase, I suggest replacing the term “Mott insulator” simply with “insulator.” This is to account for the unsettled debate about the nature of this state (i.e., spin-frustrated Mott vs. spin-singlet band insulator). See for example Y. D. Wang et al., Nat. Commun. 11, 4215 and many other recent papers. For the same reason, when the authors mention that the ground state of bulk 1T-TaS₂ is driven by the interplay among the Coulomb repulsion, lattice strain, and Fermi surface nesting, they should also include the interlayer hopping.

We thank the Referee for this input.

We have made the suggested changes in the manuscript and subsequently added a citation to Y. D. Wang et al., Nat. Commun. 11, 4215 (a paper later cited in the submitted manuscript) at the end of the sentence pointed out by the Referee.

2) I suggest that the insets of Figure 3A also display the transition temperatures of the free-space material as a reference.

We agree that adding the transition temperature of the free-space material in the insets of Fig. 3A may help to emphasize the temperature renormalization that the cavity environment prompts. In fact, in the submitted version of the manuscript, the purpose of the insets was to show the comparison of the measured critical temperatures of the sample as a function of the cavity alignment, thereby highlighting the huge modifications introduced by the cavity alignment.

However, as the dependencies plotted in the previous insets of Fig. 3A can be directly grasped from the main panel of Fig. 3A, we have preferred to replace the former insets with a panel displaying the raw time-domain THz traces

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acquired by fixing the temperature (154 K) and tilting the cavity mirrors. In our opinion, this better emphasizes how the cavity alignment control the radiation heat load onto the sample and drives the sample from a dielectric to a metallic state.

We have moved the former insets of Fig. 3A in the supplementary materials (Fig. S33) and, as suggested by the Referee, we included the free-space reference.

3) The axes of the right panels in Figure 2 are incorrect. The values should span the range 11-15 if the transmission is expressed in %. The same applies to the vertical axes of the two panels in Figure 3 and in Figure 4C.

We apologize for not clearly stating in the manuscript how the hysteretic curves in Fig. 2 were worked out in the submitted version of the manuscript. This may have generated confusion in understanding the range of the horizontal axis of the right panels in Fig. 2 and the vertical axes in Fig. 3 and 4C.

In Fig. 2 (right panels), we showed the low-frequency transmission integrated in the range $0.2 \text{ THz} < \omega < 1.5 \text{ THz}$. The confusion may arise from the fact that we plotted, on the same panel, the curves obtained both in free space and within the 11.5 GHz cavity. By looking at the color scales of the corresponding maps in the left panels of Fig. 2, one may notice that the transmission (T) ranges are very different in the two configurations: while T ranges from -2% to 11% in the free-space case, it ranges from -0.02% to 0.09% when the sample is enclosed in the cavity. This is reasonable, as the amount of light transmitted by the sample within the cavity is expected to be less with respect to the free-space configuration in which the cavity mirror are removed. The integration performed at low-frequency will therefore yield very different values in free space and within the cavity. In order to facilitate the comparison of the hysteresis measured in the two configurations (right panel of Fig.2), we have rescaled the free-space hysteretic curves in order to fall in the same transmission range as the 11.5 GHz cavity ones. We stress that this is just a translation along the horizontal axis, which does not affect the measured transition temperature by any means.

In the same fashion, the vertical axes in Fig. 3 and 4C are correct because they refer to the transmission of the sample measured within the cavity, which is responsible for a substantial attenuation of the transmitted light.

We apologize again for not mentioning in the caption of Fig. 2 that the free-space hysteretic curves were translated for display needs. We now included a comment on this in the revised caption.

Furthermore, we take this opportunity to inform the Referee that we slightly change the arrangement of Fig. 2 in the revised manuscript. In order to make clearer the comparison between the free-space and the cavity data, we plotted the four transmission maps on the same row (each one with its corresponding color scale) and we plotted the four hysteretic curves on the same plot (current Fig. 2C).

4) The authors did not cite relevant literature on THz and sub-THz cavities embedding materials with macroscopic quantum effects [e.g., F. Appugliese et al., *Science* **375**, 1030 (2022)].

We apologize for not citing the work pointed out by the Referee and other related papers. We have appropriately incorporated the relevant literature in the revised manuscript.

Specifically, we included the following references: Appugliese et al. ([10.1126/science.abl5818](https://doi.org/10.1126/science.abl5818)), Paravicini-Bagliani et al. (<https://doi.org/10.1038/s41567-018-0346-y>), Garcia-Vidal et al. ([10.1126/science.abd0336](https://doi.org/10.1126/science.abd0336)), Canaguier-Durand (<https://doi.org/10.1002/anie.201301861>).

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5) All the error bars reported in the figures should be defined in the corresponding legends. I could not find any description of how the error bars were estimated in Figures 3 and 4. The authors should also specify the temperature stability range of their cryostat during a measurement at a fixed temperature. I assume this value to be negligible. However, if it turned out to be significant, all data should show an error bar along the temperature axes.

We thank the Referee for pointing this out. The estimation of the error bars was mentioned in the supplementary materials, precisely in the section devoted to explaining how the critical temperatures were estimated starting from the hysteresis curves. In particular, for each data point, we usually acquire three independent scans; the error bars are the standard deviation associated to the critical temperature calculated over three consecutive scans.

Following the Referee's suggestion, we now added a similar description in the caption of Fig. 4A and Fig. S33 (which now contains the former insets of Fig. 3A).

Regarding the temperature stability of the cryostat, we confirm that the temperature deviation is less than 1K during a measurement at a fixed temperature. Therefore, we did not include error bars along the temperature axes in any figure.

We have now specified the temperature stability range of the cryostat in the description of the experimental setup (supplementary materials).

We thank the Referee again for the careful assessment of the manuscript and the insightful inputs.

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Referee #3 (Remarks to the Author):

This study reports the control of a phase transition in the dichalcogenide 1T-TaS₂ through the coupling to a GHz Fabry-Perot cavity. The bare material shows a hysteresis curve between 140 and 180 K. In the current experiment, this transition is traced via the change of the low frequency transmission of a terahertz pulse, which traces the presence of free carriers. As shown in Fig. 2, regardless of whether the material is in the metallic or the insulating phase, the insertion of the material into the GHz cavity does not noticeably change the optical response of the system in this frequency regime, which lies substantially above any GHz cavity resonance. Remarkably, however, by tuning the fundamental cavity resonance (i.e. by moving the Fabry-Perot mirrors relative to the sample), the authors raise or lower the temperature range of the hysteresis curve by an astonishing amount – between 110-135 K (at a cavity frequency of around 15 GHz) to 185-210 K (for a cavity frequency above 100 GHz). At any cavity frequency, the temperature range shrinks substantially from 38 K in the bare material to 23-33 K in the cavity-coupled complex.

This cavity-induced shift of the hysteresis behaviour, and in particular its incredible magnitude, is entirely unexpected and I cannot think of any previous work that would even hint at such an amazing impact of low-frequency cavities on correlated materials. The experimental setup is conceptually very simple and the authors carefully rule out alternative explanations of these measurements. It is – together with the experiments by the group of Jerome Faist (*Science* [375, 6584 \(2022\)](#)) – the only ‘clean’ demonstration to date of a cavity altering the macroscopic electronic properties of a solid state system. Therefore, I anticipate that this study will be a major breakthrough for the cavity control of quantum materials, and will have a tremendous impact on the future development of the field. However, as I detail below, the authors should provide additional data at specific points, if possible, and clarify their conclusions, to further substantiate their conclusions:

We thank the Referee for reviewing the manuscript and acknowledging the impact of our study in the field of the cavity control of materials.

Before addressing the concerns that the Referee raises, we would like to mention that the main modifications that we made to the current version of the manuscript is the inclusion of additional measurements and simulations. In particular, in order to discriminate between the two theoretical scenarios that we proposed, we carried out:

- Temperature measurements of the actual temperature of the sample by sealing a micrometric thermocouple in thermal contact with the sample. This allowed us to run an independent set of measurements in which the sample’s temperature is monitored in the same cavity configurations that we studied with THz spectroscopy.
- Finite elements simulations of a membrane enclosed in a tunable cavity to quantify the effects of incoherent thermal heating as function of the cavity length and alignment.

The main outcomes of these additional measurements is that the Purcell-like scenario is the more plausible one. In particular, the temperature measurements qualitatively confirmed all the trends that we already observed via THz spectroscopy, while the simulations further ruled out the possibility that the effects can be explained by an incoherent thermal heating of the sample.

We included in the supplementary materials these additional data and discussed in the main text the major results. In the following, we will refer also to these data to address the Referee’s questions.

- In particular, in Fig. 3a, the authors explore the transition from the cavity-controlled to the free space regime

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through the gradual misalignment of the cavity mirrors. Two important questions on this transition should be addressed:

1) It is shown that the misalignment moves the hysteresis curve towards the free space limit. But even at an angle of almost 10 degrees, there is still an enormous deviation from the free space hysteresis curve of around 20 K. Are there technical reasons that restrict the authors from going to larger values angles and explore the vanishing of the remaining discrepancy?

In this regard, the authors only provide measurements of the cavity resonance change in the supplementary material. It would be helpful if they could provide measurements of the cavity Q-factor as a function of the misalignment and demonstrate that it is indeed the vanishing of the cavity resonance which is responsible for recovering the free space limit.

We thank the Referee for this comment. We agree that the free-space limit should be eventually recovered by misaligning the cavity, as the trend that we reported in Fig. 3A suggests. However, the deviation from the free-space configuration that we still observe in the cavity misaligned by almost 10 degrees indicates that, even in this configuration, a non-null light-matter coupling is still present. Unfortunately, this is the largest misalignment that can be obtained in the current design due to technical constraints of the piezo-controlled cavity.

In order to clarify the connection between the mirrors' alignment and the cavity quality factor Q, we estimated Q of the misaligned cavity as a function of the misalignment angle (Fig. R5). This estimate is based on the THz fields transmitted by the cavity that, as highlighted in the dashed box in Fig. S20A, display the reflection associated to the cavity round trip. The reduction in the intensity of the cavity peak upon misaligning the cavity mirrors can be attributed to the reduced photon lifetime and hence to the reduced quality factor of the cavity. The quality factors have been extrapolated from Fig. S20A by approximating the exponential decay with a linear fit.

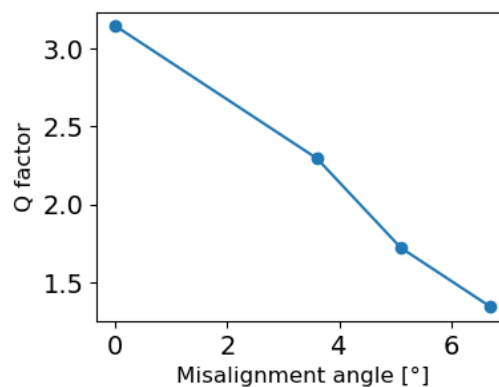


Figure R5: Cavity quality factor as function of the misalignment of the cavity mirrors.

Furthermore, we stress that the change in the measured critical temperature as a function of the cavity alignment is a genuine cavity-mediated effect and cannot be accounted by incoherent radiative cooling from the samples. In fact, by performing finite elements simulations in which the membrane is enclosed in two mirrors with tunable alignment, we demonstrated that the local temperature at the sample's position is not affected by the misaligning angle (Fig. S21). This rules out the possibility that trivial geometrical arguments can be used to explain the renormalization of the measured critical temperature as function of the cavity alignment, and indicates instead at a cavity-mediated process due to the cavity electro-dynamics.

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We included the estimation of the quality factors (Fig. R4) in the supplementary materials (current Fig. S20B) and changed the text accordingly. We also added the abovementioned simulations of the membrane's thermal profile as a function of the alignment of the cavity mirrors (Fig. S21).

2) There should be a second route to recover the free space limit - by increasing the distance between the mirrors and sending the resonance frequency to zero (as also sketched in Fig. S11). Therefore, one would expect that, in Fig. 4a, the critical temperatures should approach the free space limit at small cavity frequencies. Yet this is the regime where the authors observe the largest shifts in these critical temperatures. This to me is perhaps the most perplexing of the results presented in this manuscript and it should be discussed carefully. At large frequencies the critical temperatures appear to arrive at some kind of saturation regime, but there is no such limit visible at small frequencies.

Would it be possible to increase the data range of Fig. 4a to even smaller frequencies? How do the authors explain this startling phenomenon?

We thank the Referee for raising this point, which has indeed a key importance. As the Referee correctly states (and as it was also pointed out by Referee#1 question#3), it would be reasonable that the low-frequency cavity data points should be smoothly connected to the free space datasets. What we observed instead is a large deviation of the critical temperatures measured in large cavities from the free-space reference. Depending on the scenario used to interpret the experiment (free energy vs Purcell effect), the results of Fig. 4A suggest that either the critical temperature is lower in large cavities than in free space or the cavity-mediated sample's temperature is increased when the sample is embedded in large cavities.

In this regard, the additional temperature measurements and the finite elements simulations that we now included in the manuscript helped us clarify this point.

We simulated a bare membrane enclosed in a tunable cavity (Fig. S16C) and calculated how much the temperature in the middle of the membrane (T_{membrane}) deviates from the temperature at the edge, which is kept fixed and equal to the cold finger's temperature (T_{ext}). As it would be expected, the temperature deviation for longer cavities smoothly approaches the free-space reference. This trend has been directly measured in our setup by sealing a Cr-Al junction within the bare membranes. The results in Fig. S11B show that the intuitive picture in which the free space limit is recovered by increasing the cavity length is confirmed in the membrane.

However, by placing the micrometric thermocouple in thermal contact with the sample and repeating the experiment, we observed that the data acquired in the lowest-frequency cavity deviate from this trend (Fig. S11A). In particular, we observed a non-monotonic behavior: the sample is cooler than free space in smaller cavities, but warmer in larger cavities, in which the sample's temperature overcomes the free-space reference. We included these new results in the main manuscript in Fig. 4B. The trend that we measured confirms the evidence already collected via THz spectroscopy and reinforces the robustness of the observation, especially because the temperature measurements were performed in a sample belonging to a different batch.

We agree with the Referee that for even longer cavity we would expect the transition temperature to approach the free space and it is reasonable to assume so. Unfortunately, we could not carry out measurements with cavities larger than 11.5 GHz due to geometric constraints of the vacuum chamber, which (as also mentioned to Referee#2 in question#2) was initially designed to study the strong coupling in higher frequency micrometric cavities. A complete redesign of the experimental chamber would be needed to perform measurements with longer cavities.

Finally, it is worth highlighting that, while still unexplained, the anomalous behavior that we observed when the sample is embedded in the cavity is genuine and cannot be ascribed to a possible experimental artifact because it was not observed in the cavity without the sample. Moreover, it has to be a consequence of the cavity electrostatics within the cavity since a simple incoherent thermal effect (as the simulations and measurements demonstrated) would not reproduce the experimental evidence.

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We carried out measurements of both the membrane's and the sample's local temperature for different cavity lengths. We included these results in Fig. S11 in the supplementary materials and Fig. 4B in the main manuscript, showing that the non-monotonic trend observed via THz spectroscopy is a sample-dependent effect which is reproducible also in other batches.

- Two possible theoretical explanations are provided for the experimental observations - one based on a hybridization scenario, where the cavity couples to domain wall fluctuations in the metallic phase to form new eigenstates and thus change the free energy, and one based on the Purcell effect, which effectively decouples said domain wall fluctuations from the electromagnetic environment when the photonic density of states is reduced at their respective resonance frequency. Moreover, in Section S9 of the supplementary material, the authors argue convincingly that the first scenario is insufficient to explain the observations, as the phase space volume that can be affected by the direct cavity coupling is small compared to the full system. They conclude that "this puts more emphasise on the second mechanism". Thus, they rule out a strong coupling scenario and instead favour a weak coupling scenario. But it is not apparent why the same phase space argument should not apply to this Purcell scenario, too. If strongly coupled cavity modes cannot affect the free energy, why should weakly coupled modes be able to achieve this?

We thank the Referee for this very profound comment. We will justify here our argument and discuss it more carefully in the manuscript.

As the Referee correctly highlights, the main argument that we raised against the free-energy picture is that it is unlikely that a single cavity mode can perturb the material in the thermodynamic limit. The phase space volume needed to induce significant changes in the free energy landscape would require unreasonably large couplings. The origin of this limit is that we considered a closed system (material + cavity) that has no exchanges with the external environment.

However, this is not the case for the second description that we proposed. In fact, in the Purcell-like scenario, we considered an open system in which the material embedded in the cavity is in thermal contact with the external photon bath. Therefore, the second scenario is essentially a non-equilibrium stationary state approach in which thermal exchanges between the sample, the cold finger and the photon bath are allowed. As a result, the system will take away energy from one mode and redistribute it on the others, on a very fast timescale which is set by a microscopic scattering between the modes due to anharmonic interactions. In this context, the argument that a single cavity mode cannot affect the system in the thermodynamic limits is no longer a restriction.

In synthesis, the main difference between the two theoretical descriptions lies in the fact that the first scenario considers a closed system, while the second one is built on the hypothesis that the system is open. In this regard, a simple yet effective example that clarifies how the phase space argument no longer applies for open systems is our daily experience with the sunbeams: sunlight is always absorbed at $q=0$, but this does not prevent objects from getting warm.

Moreover, the temperature measurements that we now included in the manuscript confirmed that the sample's emission spectrum (and hence its local temperature) is coherently affected by the cavity settings, such as its length and alignment. This further confirms that a Purcell-like scenario seems to be the dominant theoretical explanation of the experimental observations.

We added the following sentence in the main text, right before the conclusion.

"In contrast to the first scenario, the phase space restriction is no longer valid in the second mechanism (Fig. 1A), in which an open system is considered and thermal exchanges between the material, the cold finger and the photon bath are allowed. This, together with the experimental evidence that the cavity can coherently heat up or cool

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down the sample as a function of the frequency (Fig. 4B) suggests that the second mechanism in Fig. 1A is the dominant effect in the observation reported.”

We warmly thank again all Referees for the constructive revision of the manuscript and the detailed and profound comments given.

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Reviewer Reports on the First Revision:

Referees' comments:

Referee #1 (Remarks to the Author):

I have studied the authors' response to all three referees and reviewed the updated manuscript and supplemental materials. I appreciate the comprehensive answers, in particular the new temperature calibration experiments using the minuscule Cr-Al thermal couple. The additional experiments and analyses reveal the following points:

(a) The actual sample temperature (T_{int}) is indeed very different from the cold head temperature (T_{ext}), and the actual sample temperature changes with the cavity mirror configuration in a way that is quasi-quantitatively consistent with the perceived change of the insulator-metal transition temperature.

(b) As I suspected before, incoherent thermal radiation due to the close proximity of the cavity mirrors has a sizeable effect on the actual sample temperature, but this effect alone cannot explain all aspects of the data (for example, as shown in Figure S15, the apparent T_c increases as mirrors get closer regardless of whether the mirrors are cryogenic or at room temperature).

(c) The authors therefore invoke cavity QED to account for the effects that cannot be otherwise explained, which is the central conclusion of the manuscript.

I find the central conclusion in point (c) is not well supported because of two reasons.

Firstly, as the authors acknowledge, there are crucial observations that are inconsistent with the cavity QED interpretation, notably the non-monotonic trend of the effective T_c as a function of cavity frequency (Figure 4A and Figure 4B). Both I and Referee #3 raised this question in the previous round and the authors offered a lengthy answer. However, as far as I understand, the authors cannot explain this anomaly and they wrote in the response letter, "The evidence collected so far does not allow to fully understand the origin of this anomalous behavior of the sample." Following the logic of exclusion in point (b) above, one can conclude that there must be other mechanisms at play beyond cavity QED and incoherent radiation effect.

Secondly, neither the free-energy renormalization scenario nor the Purcell-like scenario can quantitatively explain such a large change of the sample temperature due to cavity QED. I appreciate that both scenarios offer a semi-qualitative agreement (except the non-monotonic trend mentioned above), but this qualitative agreement also depends on an artificial choice of parameters. For example, the authors chose 15 GHz as the central frequency of some collective modes in the sample to model the renormalization of the free energy, so that as the cavity frequency increases within the probed range, $T_{\text{int}}/T_{\text{ext}}$ decreases (Figure S7C). The choice of 15 GHz seems arbitrary and the authors could not identify its microscopic origin. If instead the authors used 50 GHz, then $T_{\text{int}}/T_{\text{ext}}$ will first increase and decrease within the experimentally probed cavity frequency range,

therefore breaking the qualitative agreement. Hence, the “qualitative agreement” seems to be a circular argument. More importantly, considering the extremely low Q factor around 3 to 4 (Figure S2), the quantitative agreement is too far off. The authors are well aware of this inconsistency, and they stated, “...extremely large couplings would be needed for the free energy changes to explain the observed shifts...”, and “a quantitative agreement of the [Purcell-like] effect could not be reached...”

Given the above concerns, I feel very split about my recommendation. On one hand, the experimental technique and innovations (including the added temperature measurements) are trailblazing, and the detected effect of sample temperature renormalization is amazing. The authors have also performed extensive checks and reproducibility measurements to exclude common experimental artefacts. From this point of view, the results deserve to be published in some form. On the other hand, the interpretation of any cavity QED effect is not substantiated and in fact contradicted by key experimental data, so publishing the paper as a milestone of cavity control of quantum materials is misleading. In this regard, a lack of understanding of what effect can consistently explain the dataset calls for additional work before publication is granted.

I will therefore leave the decision to the editor to judge whether it is appropriate to publish the very interesting set of results even though their origin is not accounted for. If the publication route is chosen, I suggest the manuscript undergo a major revision in terms of presentation (including the title) because of the tenuous experimental evidence for cavity QED. The paper may be more appropriately titled “Anomalous renormalization of sample temperature in a Fabry-Pérot cavity” because, as the authors carefully showed, the dominant effect is a mismatch of the sample temperature and the cold head temperature. Currently, the manuscript is written as if cavity QED can control the insulator-metal phase transition, but the physics of the phase transition is not relevant because the sample temperature is not what the cold head sensor says it is.

I once again thank the authors for their hard work in addressing all of the three referees’ concerns and congratulate them on observing this peculiar effect. Even though the origin is unknown and the link to cavity QED is unconvincing, the experimental part is solid and deserves to be known in the community in some form so that other groups can try to follow up and investigate the underlying cause.

Referee #2 (Remarks to the Author):

After carefully reviewing the remarks provided by all referees and thoroughly considering the responses offered by the authors, I am pleased to state that the authors have delivered a convincing and satisfactory rebuttal. The revised paper exhibits significant improvements compared to the previous version. While the suggested microwave experiments could not be performed, the measurements conducted with the micrometric Cr-Al junctions have confirmed the results obtained with time-domain terahertz spectroscopy and likely clarified the Purcell-like scenario as the dominant effect at play. Furthermore, these measurements have enabled the authors to decouple their discussion from any potential strong-coupling scenarios associated with domain wall

fluctuations (which were largely speculative). In my opinion, the final article represents an exceptional piece of work and therefore merits publication in Nature.

Before proceeding with publication, I would like to offer some final remarks to the authors.

Since the new data supports the weak coupling limit, which corresponds to a nonequilibrium stationary state, the sentence “the use of resonant optical cavities has emerged as a suggestive possibility for controlling material properties at equilibrium and without driving” might be misinterpreted within the scope of this paper. While I appreciate the authors' intention, I recommend tuning the language to encompass both equilibrium and nonequilibrium phenomena when discussing this topic.

Second, it would be beneficial for the authors to enhance the clarity of the experimental configuration by including a schematic panel in Figure 1. Specifically, including the two measured temperatures, T_{int} and T_{ext} , in the diagram would provide a more straightforward visualization of the measurement procedure for a broader readership. The current version of the paper might be challenging to comprehend without a careful review of the Supplementary Information.

Referee #3 (Remarks to the Author):

The authors have clarified the issues raised in the first round of reports. The paper is now acceptable for publication.

Author Rebuttals to First Revision:

Referee #1 (Remarks to the Author):

I have studied the authors' response to all three referees and reviewed the updated manuscript and supplemental materials. I appreciate the comprehensive answers, in particular the new temperature calibration experiments using the minuscule Cr-Al thermal couple. The additional experiments and analyses reveal the following points:

- (a) The actual sample temperature (T_{int}) is indeed very different from the cold head temperature (T_{ext}), and the actual sample temperature changes with the cavity mirror configuration in a way that is quasi-quantitatively consistent with the perceived change of the insulator-metal transition temperature.
- (b) As I suspected before, incoherent thermal radiation due to the close proximity of the cavity mirrors has a sizeable effect on the actual sample temperature, but this effect alone cannot explain all aspects of the data (for example, as shown in Figure S15, the apparent T_c increases as mirrors get closer regardless of whether the mirrors are cryogenic or at room temperature).
- (c) The authors therefore invoke cavity QED to account for the effects that cannot be otherwise explained, which is the central conclusion of the manuscript.

I find the central conclusion in point (c) is not well supported because of two reasons.

Firstly, as the authors acknowledge, there are crucial observations that are inconsistent with the cavity QED interpretation, notably the non-monotonic trend of the effective T_c as a function of cavity frequency (Figure 4A and Figure 4B). Both I and Referee #3 raised this question in the previous round and the authors offered a lengthy answer. However, as far as I understand, the authors cannot explain this anomaly and they wrote in the response letter, "The evidence collected so far does not allow to fully understand the origin of this anomalous behavior of the sample." Following the logic of exclusion in point (b) above, one can conclude that there must be other mechanisms at play beyond cavity QED and incoherent radiation effect.

Secondly, neither the free-energy renormalization scenario nor the Purcell-like scenario can quantitatively explain such a large change of the sample temperature due to cavity QED. I appreciate that both scenarios offer a semi-qualitative agreement (except the non-monotonic trend mentioned above), but this qualitative agreement also depends on an artificial choice of parameters. For example, the authors chose 15 GHz as the central frequency of some collective modes in the sample to model the renormalization of the free energy, so that as the cavity frequency increases within the probed range, $T_{\text{int}}/T_{\text{ext}}$ decreases (Figure S7C). The choice of 15 GHz seems arbitrary and the authors could not identify its microscopic origin. If instead the authors used 50 GHz, then $T_{\text{int}}/T_{\text{ext}}$ will first increase and decrease within the experimentally probed cavity frequency range, therefore breaking the qualitative agreement. Hence, the "qualitative agreement" seems to be a circular argument. More importantly, considering the extremely low Q factor around 3 to 4 (Figure S2), the quantitative agreement is too far off. The authors are well aware of this inconsistency, and they stated, "...extremely large couplings would be needed for the free energy changes to explain the observed shifts...", and "a quantitative agreement of the [Purcell-like] effect could not be reached..."

Given the above concerns, I feel very split about my recommendation. On one hand, the experimental

technique and innovations (including the added temperature measurements) are trailblazing, and the detected effect of sample temperature renormalization is amazing. The authors have also performed extensive checks and reproducibility measurements to exclude common experimental artefacts. From this point of view, the results deserve to be published in some form. On the other hand, the interpretation of any cavity QED effect is not substantiated and in fact contradicted by key experimental data, so publishing the paper as a milestone of cavity control of quantum materials is misleading. In this regard, a lack of understanding of what effect can consistently explain the dataset calls for additional work before publication is granted.

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I once again thank the authors for their hard work in addressing all of the three referees’ concerns and congratulate them on observing this peculiar effect. Even though the origin is unknown and the link to cavity QED is unconvincing, the experimental part is solid and deserves to be known in the community in some form so that other groups can try to follow up and investigate the underlying cause.

We thank the Referee for the insightful comments now as well as in the previous report.

We would like to start by clarifying a slight misunderstanding. The referee believes that cavity electrodynamics alone cannot explain our observation and that we acknowledged this in our previous response. In particular, the referee#1 states that the non-monotonous dependence of the temperature change with cavity fundamental frequency cannot be accounted for by either of the mechanisms proposed, quoting us in: “*The evidence collected so far does not allow to fully understand the origin of this anomalous behavior of the sample.*”

It is important to clarify that our statement refers to the fact that while the conclusion of the paper is that the cavity controlled Purcell-like change of the heat load of the sample is probably the dominant effect, we cannot exclude that a cavity controlled renormalization of the free energy is at play. The statement did not refer to the non-monotonous behavior of the temperature with cavity frequency, which indeed is consistent with the Purcell-like effect. In this respect, the Purcell effect predicts the *enhancement and reduction* of radiative transitions rates under different resonance conditions and thus can also explain the observation that low frequency cavities may coherently heat up the sample with respect to the free space while high frequency ones cool it down. This non monotounous behavior can be seen in Fig. S7 ($\Omega < \omega$).

The misunderstanding may come partly from the fact that our model focuses on comparing the response of the sample at different cavity frequencies which are mostly above the material resonance (Fig. S7).

This choice was made for consistency with the experiments to illustrate that in the frequency range used increasing the cavity frequency effectively cools down the sample. This naturally gives monotonous

behavior. However, it should be made clear that tuning the cavity at lower frequencies can in principle explain the non-monotonous behavior and a comparison with low cavity frequencies eventually tending to 0 frequency (free space) is not reported in the manuscript.

In any case, it is important to stress that the role of the theoretical model in the manuscript is to give an interpretative framework of the observation and a full theoretical study of this mechanism is beyond the scope of this paper.

The measurements have shown that the phase transition control within the cavity is associated with a strong thermal renormalization, suggesting a Purcell-like mechanism to be the dominant effect at play. As extensively pointed out in the previous responses and in the manuscript, all the experimental evidence point towards a cavity-mediated effect:

- the sensitivity of the phase transition to the cavity geometry and the fact that, regardless the mirrors temperature,

- the effective critical temperature is pushed up upon increasing the frequency of the cavity fundamental mode.

- as proven by the complementary temperature measurements together with the finite elements simulation these evidences are in stark contrast with an incoherent heating scenario and point towards an effect mediated by the cavity geometry

- the observed effects are due to the presence of the sample, since in absence of sample only tiny changes are observed. This indicates that the effects observed are related to both the low energy physics of the sample and its coupling to the cavity modes.

We improved the discussion and abstract, following the Referee request, clarifying that the control of the phase transition is due (in large part) to cavity driven temperature changes.

Reviewer#1 suggests that also the title should be revised from: “Cavity control of the metal-to-insulator transition in 1T-TaS₂” to “Anomalous renormalization of sample temperature in a Fabry-Pérot cavity”. We revise the title, but choose the title: “Thermal control of a metal-to-insulator transition in a quantum material in a Terahertz Fabry-Pérot cavity”.

We opt for the title because, while we agree with reviewer#1 that it is important to include in the title a reference to the thermal mechanism, we also think that it is important to retain the message about the control of the phase transition to provide an adequate description of the focus of the work. We think this title is more representative of the content of the paper and accounts for both aspects.

Finally, it is important to note that while the conclusion of the paper is that the cavity controlled Purcell-like change of the heat load of the sample is probably the dominant effect, we cannot exclude that a cavity controlled renormalization of the free energy is at play. We therefore believe that the revised discussion including both scenarios will benefit the readers and will possibly influence the community to look at both aspects of cavity electrodynamics.

We sincerely thank once again the Referee#1 for their insightful and constructive feedback to the manuscript provided.

Referee #2 (Remarks to the Author):

After carefully reviewing the remarks provided by all referees and thoroughly considering the responses offered by the authors, I am pleased to state that the authors have delivered a convincing and satisfactory rebuttal. The revised paper exhibits significant improvements compared to the previous version. While the suggested microwave experiments could not be performed, the measurements conducted with the micrometric Cr-Al junctions have confirmed the results obtained with time-domain terahertz spectroscopy and likely clarified the Purcell-like scenario as the dominant effect at play. Furthermore, these measurements have enabled the authors to decouple their discussion from any potential strong-coupling scenarios associated with domain wall fluctuations (which were largely speculative). In my opinion, the final article represents an exceptional piece of work and therefore merits publication in Nature.

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We sincerely appreciate the insightful assessment of the revised version of our manuscript and are glad of the positive evaluation given.

To follow the Referee comment, we have clarified that the observed effect hints towards a non-equilibrium Purcell-like effect and that the control of the phase transition is associated with a strong thermal renormalization induced by the cavity environment. Additionally, since we showed that the dominant effect at play is a non-equilibrium mechanism mediated by the cavity electromagnetic environment which does not require strong coupling, we reduced the emphasis given to polaritonic effects and introduced a more balanced description of weak and strong coupling both at the abstract level and within the manuscript.

Following the suggestion of the Referee, we have also included in Fig.1A the notation used for the external temperature (T_{ext}) and the temperature of the sample (T_{int}) which can be modified by the cavity geometry.

We thank the Referee once more for the careful assessment of the manuscript and the profound input given which we believe have significantly benefited the quality of the manuscript.

Referee #3 (Remarks to the Author):

The authors have clarified the issues raised in the first round of reports. The paper is now acceptable for publication.

We warmly thank Reviewer#3 for the insightful comments given and for supporting the publication.