nature portfolio

Peer Review File

Cavity-altered superconductivity

Corresponding Author: Dr Itai Keren

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

Version 0:

Reviewer comments:

Referee #1

(Remarks to the Author)
Review of “Cavity-Altered Superconductivity”

Reviewer

July 27, 2025

The manuscript presents an exceptionally innovative investigation into the control of superconducting properties through resonant coupling with hyperbolic phonon polaritons in van der Waals heterostructures. By ingeniously combining hexagonal boron nitride (hBN) with the organic superconductor κ-(BEDT-TTF)2Cu[N(CN)2]Br (κ-ET), the authors provide compelling evidence for cavity-mediated suppression of superfluid density driven by vacuum fluctuations. This represents a paradigm shift from traditional optical excitation approaches.
The experimental methodology, employing magnetic force microscopy (MFM) and scattering-type scanning near-field optical microscopy (s-SNOM), is complemented by rigorous theoretical modeling. The demonstration of electromagnetic environment engineering as a tool to modify superconductivity opens exciting possibilities in quantum material design. The observed resonant coupling between hBN hyperbolic modes and the C=C stretching vibration in κ-ET establishes a fundamentally new mechanism for controlling quantum phases.

Major Strengths
• The experimental approach is outstanding, particularly the use of MFM for local superfluid density mapping and s-SNOM for polariton dispersion visualization. These techniques provide mutually reinforcing evidence for the cavity effect.
• The control experiments with non-resonant materials (RuCl3 and BSCCO) are carefully designed and effectively eliminate alternative interpretations, highlighting the specificity of the observed phenomenon.
• The theoretical framework, including first-principles molecular Langevin dynamics simulations, provides strong support for the proposed interaction mechanism between zero-point fluctuations and the C=C vibrational mode.
• The suggestion that cavity engineering could potentially enhance superconductivity, possibly increasing Tc, represents a visionary direction for future research that could have transformative implications.
• The manuscript is exceptionally well-organized, with clear logical progression from experimental results to theoretical interpretation. The figures

are carefully prepared and effectively communicate the key findings, particularly the avoided crossings in polariton dispersion and the suppression of Meissner response.

Suggestions for Improvement

1. While the MFM data convincingly demonstrate the cavity effect, the depth profile of this influence could be further characterized. Depth-resolved spectroscopic measurements might provide valuable additional insights into the spatial extent of the modification.
2. The connection between polariton coupling and suppressed superconductivity would benefit from more detailed theoretical treatment. Clarifying whether this occurs through modified electron-phonon interactions or alternative pathways would strengthen the mechanistic interpretation.
3. The temperature dependence near Tc is well-documented, but additional intermediate-temperature data could provide valuable information about the transition dynamics between the normal and superconducting states.
4. The exciting possibilities offered by hyperbolic cavities would be more compelling with additional examples from different material systems to support the claim of broad applicability.
5. While the authors have thoughtfully considered potential interfacial effects, a more systematic discussion of alternative explanations (such as charge transfer processes) would make the conclusions even more robust.

Recommended Revisions

• The MFM discussion would benefit from referencing recent advances in this technique for studying novel superconductors and superconducting devices, such as [1, 2, 3, 4, 5]. Including examples of superconducting property modification under various conditions [6] would provide valuable context for interpreting the dF/dz signals.
• Adding examples of interface superconductivity, such as those discussed in [7], would strengthen the discussion of heterostructure effects.
• Including additional temperature-dependent data would help clarify the suppression dynamics across the full temperature range.
• Expanding the discussion on potential cavity-induced enhancement of superconductivity, particularly regarding possible mechanisms for Tc increase, would highlight promising future directions.
• A more detailed consideration of interfacial effects would make the argument for cavity-mediated phenomena even more compelling.
• For greater impact, consider moving some of the supplementary data (particularly figure S14 panels a/d or b/e) to the main text, as they provide strong evidence for hBN influence.

Overall Assessment

This work represents a landmark advance in the emerging field of cavity quantum materials, demonstrating that vacuum fluctuations in engineered electromagnetic environments can profoundly modify superconducting ground states.
The experimental evidence is comprehensive and supported by sophisticated theoretical modeling. While some mechanistic details warrant further exploration, the study establishes a robust foundation for future investigations into cavity-mediated control of quantum phases.
The implications extend far beyond superconductivity, potentially influencing diverse areas including correlated electron systems, polaritonic devices, and quantum information science. The manuscript is exceptionally well-executed and makes a compelling case for this new approach to quantum material engineering.
After addressing the minor points raised above, this outstanding work will be suitable for publication in Nature. The scientific rigor, novelty, and potential impact fully meet the journal's high standards.

References

[1] Vasily S Stolyarov, Ivan S Veshchunov, Sergey Yu Grebenchuk, Denis S Baranov, Igor A Golovchanskiy, Andrey G Shishkin, Nan Zhou, Zhixiang Shi, Xiaofeng Xu, Sunseng Pyon, et al. Domain meissner state and spontaneous vortex-antivortex generation in the ferromagnetic superconductor eufe2 (as0. 79p0. 21) 2. Science advances, 4(7):eaat1061, 2018.
[2] Vasily S Stolyarov, Vsevolod Ruzhitskiy, Razmik A Hovhannisyan, Sergey Grebenchuk, Andrey G Shishkin, Olga V Skryabina, Igor A Golovchanskiy, Alexander A Golubov, Nikolay V Klenov, Igor I Soloviev, et al. Revealing josephson vortex dynamics in proximity junctions below critical current. Nano letters, 22(14):5715–5722, 2022.

[3] Razmik A Hovhannisyan, Sergey Yu Grebenchuk, Semen A Larionov, Andrey G Shishkin, Artem K Grebenko, Nadezhda E Kupchinskaya, Ekaterina A Dobrovolskaya, Olga V Skryabina, Alexey Yu Aladyshkin, Vyacheslav V Dremov, et al. Scanning vortex microscopy reveals thicknessdependent pinning nano-network in superconducting niobium films. Communications Materials, 6(1):42, 2025.
[4] SA Larionov, AG Shishkin, D Roditchev, D Yu Vodolazov, and VS Stolyarov. Peculiarities of the vortex dynamics in a granular niobium superconducting bridge. Physical Review B, 111(21):214511, 2025.
[5] A Yu Aladyshkin, RA Hovhannisyan, S Yu Grebenchuk, SA Larionov, AG Shishkin, OV Skryabina, AV Samokhvalov, AS Mel'nikov, D Roditchev, and VS Stolyarov. Magnetic force microscopy versus scanning quantumvortex microscopy: Probing pinning landscape in granular niobium films. arXiv preprint arXiv:2507.05172, 2025.
[6] S Yu Grebenchuk, Zh A Devizorova, IA Golovchanskiy, IV Shchetinin, G-H Cao, AI Buzdin, D Roditchev, and VS Stolyarov. Crossover from ferromagnetic superconductor to superconducting ferromagnet in p-doped eufe 2 (as 1- x p x) 2. Physical Review B, 102(14):144501, 2020.
[7] Andrei Kudriashov, Ian Babich, Razmik A Hovhannisyan, Andrey G Shishkin, Sergei N Kozlov, Alexander Fedorov, Denis V Vyalikh, Ekaterina Khestanova, Mikhail Yu Kupriyanov, and Vasily S Stolyarov. Revealing intrinsic superconductivity of the nb/bisbte2se interface. Advanced Functional Materials, 32(49):2209853, 2022.

Referee #2

(Remarks to the Author)
Keren et al. report the observation of a strong suppression of the superfluidity density in the molecular superconductor k-ET at the interface with a layer of hexagonal boron nitride (hBN). The authors perform magnetic force microscopy measurements – which are sensitive to the Meissner effect – to quantify the superfluid density and observe a strong suppression of the superconducting density. The claim of the paper is that this suppression is related to the coupling of the hyperbolic mode of hBN which is close in frequency to the infrared active C=C stretching mode in the ET molecules. This mode is known to be involved in the superconductivity of the compound, as also external drive by resonant mid- infrared pulses seem to suggest (Phys. Rev. Lett. 127, 197002).

The resonant coupling condition between the cavity and the C=C mode is investigated by s-SNOM measurements and by transfer-matrix calculations. The authors also fabricate other “control” devices terminated by RuCl3 – in which optical phonons lie at much lower frequency than C=C mode. The main observation reported is that only resonant hBN cavities contribute to the quench
of the superconducting behavior in k-ET.

The experimental finding is remarkable and commendable is the careful and detailed analysis of the effect reported in the supplementary material. The observation reported is the first convincing experimental demonstration that superconductivity can be influenced at equilibrium through the tailored engineering of the electromagnetic environment. If confirmed, this would constitute a major advancement in the development of strategies to control material properties via cavity electrodynamics. Given its broad implications, the study is highly relevant to multiple research communities and aligns well with the wide-ranging interests of Nature's readership.

However, I find the main conclusion of the paper which attributes the observed superfluid suppression to the resonant coupling of hBN to the C=C mode weak and not entirely supported by the data presented, which makes it difficult to conclusively and univocally link the observation to strong coupling of the cavity with a phonon mode. Underneath a detailed list of major and minor points which the authors should consider prior to publication.

1. The effect measured on the hBN-terminated device is surprisingly large. However, a small suppression is detectable also in the device terminated by the RuCl3 layer. This is particularly evident in the cyan curve in Fig. 3d, for example. The authors seem to dismiss this effect as negligible, but its amplitude is definitely above the noise level and is clearly distinguishable at low temperature. The presence of this suppression seems at odds with the authors' interpretation which attributes the observed quench of superfluid density to the coupling to the C=C mode and seems to suggest instead that a quench of superconductivity – even if smaller – is achieved also by stacking what the authors called a “non-resonant cavity”. The authors should discuss this evidence in more detail.

2. Following to the comment in point one, in the supplementary material the authors show that inhomogeneity of the hBN/k-ET interface plays a crucial role in the amplitude of the superfluid suppression (Supp. Note 9 and measurements in SI). I wonder if the difference between hBN- and RuCl3-terminated structures could be due to the different qualities of the two interfaces. The presence of only one structure terminated by RuCl3 (device 1) also makes difficult to assess the reproducibility of this result.

3. As the authors discuss, the hBN hyperbolic dispersion results in a higher photonic density in the mid-Infrared. This frequency is in the region where the effects associated to the cavity-mediated radiative coupling between the external

photonic bath and the sample are expected to give sizeable
thermodynamical effects (Phys. Rev. B 111, 165425 and Nat Chem 17(3):439-447). A possibly relevant observation in this sense is that the optical phonons of $RuCl_3$ – lying at lower energy - would be less sensitive to this effect, which could provide a mechanism for the cavity suppression of the
superfluid density which do not require strong coupling. Could the author comment on this?

4. The authors reports also measurements on hBN/BSCCO structures. The lack of superfluid suppression is used as an argument to confirm the contribution of the resonant coupling with the C=C mode. While in principle these measurements are interesting, I don't find them particularly relevant to substantiate their claim regarding the relevance of strong coupling with C=C modes. The emergence of superconductivity in BSCCO occurs for different (still unclear) reasons. There are no resonances close to the hBN hyperbolic modes that are thought to be implicated in the superconducting transition in BSCCO. As such, showing that hBN does not prompt any superfluid change in BSCCO has no implications in the interpretation of the results in k-ET.
A side point related to these measurements. In the inset of Fig. 1b, it seems that the MFM measurements on bare BSCCO (grey) are much noisier than the ones performed on hBN/BSCCO (blue). Can the authors comment on that?

5. I encourage the authors to include in Fig. 3 the dF curves as a function of the tip eight for different temperature in the hBN/k-ET device (basically, a copy of Fig. 3a but for the cavity). A comparison of this data with the one measured in bare k-ET would be important to understand the trend of the superfluid suppression within the cavity as a function of temperature. Moreover, it would be interesting to compare the same temperature-dependent curves for the $RuCl_3$-terminated structure. Is the trend the same?

6. The data presented in Fig. 1 have been taken for a k-ET sample 200 um thick. Does the thickness of the k-ET have any impact on the amplitude of the suppression measured after hBN encapsulation? Did the authors measure also hBN cavities in which the k-ET samples have different
thicknesses?

7. Did the authors check the quality of the structures over time? Did they detect any sign of degradation? While the manuscript is generally well written, the following input could improve
the readability of the manuscript.

8. In the x-axis label of Fig. 4, the authors use the word "cavity thickness", which I find a bit misleading as it might hint to a change in the cavity resonance.

9. I personally find the structure of the paper suboptimal at the moment. The reading flow is somehow interrupted between Fig. 1 and Fig. 2. I would rather swap Fig. 2 and 3 to present first all the Meissner data and discuss then the phonon-polariton dispersion.

10.It would be helpful for the reader to mention in the caption of Fig. 1a,b the temperature at which the measurements have been performed.

11.The authors might want to include the citation to a recent relevant contributions in the field DOI: 10.1126/science.abl5818, which i think is missing.

Overall, even though I find that the current version of the paper needs some revision, the experimental evidence reported is really remarkable and definitely worth to be considered for publication in Nature Journal.

Referee #3

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports.

Version 1:

Reviewer comments:

Referee #1

(Remarks to the Author)
I have reviewed the authors' responses to the reviewers' comments on the manuscript "Cavity-altered superconductivity".

The authors have adequately addressed the points raised in my initial review. The provided clarifications and additional data in the supplement are satisfactory.

Based on the revisions, I consider the manuscript suitable for publication in Nature.

Referee #2

(Remarks to the Author)
We have carefully reviewed the authors' response and examined the revisions made to both the main manuscript and the Supplementary Information. Importantly, the authors have fabricated and characterized an additional control heterostructure, $RuCl_3$/k-ET (device 5), in which a small suppression of the superfluid density is again observed, as shown in the newly added Fig. 3d.
The explanations provided by the authors—(i) the higher deposition temperature of $RuCl_3$ and (ii) the better interface quality in $RuCl_3$/k-ET compared to hBN/k-ET—appear reasonable and support the interpretation that the stronger suppression seen in the hBN-terminated structure cannot be solely attributed to interface or growth-related effects. The role played by strong coupling to vacuum fluctuations in suppressing superfluid density is a plausible explanation.
We believe that the reported experimental evidence is a milestone for the field and will certainly stimulate a vivid debate on the mechanism underlying the observation and, more importantly, will serve as a platform for defining the design criteria for cavity control of quantum materials.
Overall, we find that the authors have satisfactorily addressed all previous concerns and made a commendable effort to include the additional measurements requested. The revised manuscript meets the high standards in term of both scientific quality and novelty for publication in Nature.

Referee #3

(Remarks to the Author)
I co-reviewed this manuscript with one of the reviewers who provided the listed reports.

Referee #4

(Remarks to the Author)
I have carefully read the revised manuscript, supplementary material file, and resubmission letter. I am very satisfied with the replies by the authors to the issues that were raised. I therefore strongly recommend the Editors to publish the revised version of "Cavity-altered superconductivity" in its present form and without further delay.

Response to the reviewers

Dear Dr. Editor

We would like to express our gratitude for your attention to our manuscript entitled "Cavity-altered superconductivity" and for selecting four expert reviewers who are deeply knowledgeable on the topic of our work. We wish to thank the reviewers for their insightful comments. In their responses, all four reviewers recognize the significance of our achievement. Terms such as "a landmark advance," "remarkable" and "a key step towards the holy grail" are used by the reviewers. The reviewers accept our interpretation of the data as evidence of cavity-mediated suppression of superfluid density, while inviting us to consider additional interfacial effects. The reviewers agree that the data are complete and request only auxiliary results. The reviewers agree that the paper is easy to read; we do follow a suggestion by Reviewers 2&3 to rearrange the figures.
Overall, all reviewers suggest that, taking their comments into account, the paper is suitable to be published in Nature. Reviewer 4 suggests that "it needs to be accepted and published with the uttermost urgence."

In the revised version of the manuscript, we address all critical remarks by the reviewers. We have completed additional experiments requested by the reviewers and present new data in Fig. 3 as well as in several sections in the SI. We have added additional theoretical analysis requested by Reviewer 1 (SI sections 3 and 13). All these changes have allowed us to further improve the manuscript. We believe the revised version meets the high bar for publication in Nature.

Below, we will address the reviewers' comments one by one:

Reviewer 1:

The manuscript presents an exceptionally innovative investigation into the control of superconducting properties through resonant coupling with hyperbolic phonon polaritons in van der Waals heterostructures. By ingeniously combining hexagonal boron nitride (hBN) with the organic superconductor κ-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]Br$ (κ-ET), the authors provide compelling evidence for cavity-mediated suppression of superfluid density driven by vacuum fluctuations. This represents a paradigm shift from traditional optical excitation approaches.
The experimental methodology, employing magnetic force microscopy (MFM) and scattering-type scanning near-field optical microscopy (s-SNOM), is complemented by rigorous theoretical modeling. The demonstration of electromagnetic environment engineering as a tool to modify superconductivity opens exciting possibilities in quantum material design. The observed resonant coupling between hBN hyperbolic modes and the C=C stretching vibration in κ-ET establishes a fundamentally new mechanism for controlling quantum phases.

Major Strengths

• The experimental approach is outstanding, particularly the use of MFM for local superfluid density mapping and s-SNOM for polariton dispersion visualization. These techniques provide mutually reinforcing evidence for the cavity effect.

• The control experiments with non-resonant materials ($RuCl_3$ and BSCCO) are carefully designed and effectively eliminate alternative interpretations, highlighting the specificity of the observed phenomenon.

• The theoretical framework, including first-principles molecular Langevin dynamics simulations, provides strong support for the proposed interaction mechanism between zero-point fluctuations and the C=C vibrational mode.

• The suggestion that cavity engineering could potentially enhance superconductivity, possibly increasing T_c, represents a visionary direction for future research that could have transformative implications.

• The manuscript is exceptionally well-organized, with clear logical progression from experimental results to theoretical interpretation. The figures are carefully prepared and effectively communicate the key findings, particularly the avoided crossings in polariton dispersion and the suppression of Meissner response.

We thank the reviewer for their encouraging and thoughtful assessment. We are grateful for the recognition of our combined MFM and s-SNOM approach, the rigor of our control experiments, and the strength of our theoretical modeling. We appreciate the reviewer's remarks on the broader implications for cavity engineering and quantum material design, which reinforce the significance and potential impact of our work.

Suggestions for Improvement

1. While the MFM data convincingly demonstrate the cavity effect, the depth profile of this influence could be further characterized. Depth-resolved spectroscopic measurements might provide valuable additional insights into the spatial extent of the modification.

Response 1.1. We thank the reviewer for this suggestion and agree that the depth profile of the effect would be a useful way to better understand how hBN is modifying the electronic ground state of κ-ET. However, depth-resolved spectroscopic measurements in our case are physically impossible. Currently available spectroscopic techniques such as ellipsometry require large sample areas: at least mm^2, whereas typical dimensions of our structures are on the order of μm^2.

We note that although MFM cannot directly measure the depth profile, an estimate of "the spatial extent of the modification" can nevertheless be inferred from modeling, as we report in SI sections 5.5 and 9 of the revised manuscript.

2. The connection between polariton coupling and suppressed superconductivity would benefit from more detailed theoretical treatment. Clarifying whether this occurs through modified electron-phonon interactions or alternative pathways would strengthen the mechanistic interpretation.

Response 1.2. We appreciate the relevance of this comment. In this context, we remark that our work reports the discovery of an entirely new effect, and we do not assert any definitive interpretation of the superconducting mechanism at this stage. Determining the possible roles that hyperbolic modes (HMs) may play in affecting superconductivity remains an open and compelling question, and we hope our findings will stimulate further investigation.

Following the reviewer's suggestion, we have added section 13 to the SI in which we discuss possible pathways for cavity-altered superconductivity in hBN/κ-ET structures. A proper theory of the cavity alteration of superconductivity needs to consider (a) the dynamical strong coupling between electrons, (b) the modification of the electron-phonon coupling and (c) the polariton formation where hBN HMs with high photonic density of states couple to the C=C mode. The effects reported in our work cannot simply be explained by a static modification of the Hubbard U. It is therefore likely that the dynamic aspect is relevant; i.e., the renormalization of pairing interactions at finite frequency (especially in the vicinity of HMs) might play a role. At this stage, we do not have a full microscopic theory, but what our study demonstrates is that the amplitude of the C=C stretching mode is quenched via coupling to the high-momentum HMs of hBN. It is important to note that this specific C=C mode in κ-ET plays a central role in controlling its superconducting transition temperature by modifying the electronic dynamical screening and, consequently, the e-e pairing [Buzzi, M., et al. "Photomolecular high-temperature superconductivity." *Physical Review X* 10.3 (2020): 031028] (Ref. 29 in the manuscript). Our analysis of the spatial extent of field fluctuations outside the hBN surface further supports the long-range influence of the HMs of hBN, as we report in the new SI section 3.

Through extensive DFT and Langevin simulations, we found that only the out-of-plane electric field of the HM can perturb the C=C stretching mode, which is an IR-active vibration with an electric dipole oriented primarily along the out-of-plane direction. In a standard Fabry–Perot-type cavities, the out-of-plane coupling is vanishingly weak since the electric field is parallel to the plane. Because the frequency range of the HM dispersion branches overlaps with that of the C=C mode, the hybridization effect occurs, as shown in Fig. 4a,e of the main text.

In summary, in our revised submission, we present multiple theoretical analyses that: (1) address the nature of the HM-to-C=C mode coupling (Fig. 4), (2) support the long-range nature of such coupling (SI section 3) and (3) discuss possible microscopic mechanisms for cavity-altered superconductivity (SI section 13).

3. The temperature dependence near Tc is well-documented, but additional intermediate-temperature data could provide valuable information about the transition dynamics between the normal and superconducting states.

Response 1.3. We thank the reviewer for the suggestion and agree that additional data at intermediate temperatures are valuable. We report these data in the revised version of the SI (Fig. S20; reproduced below as Fig. R1). These data attest to gradual changes at intermediate temperatures, thus validating all the inferences in our manuscript. At the moment, we are reluctant to embark on a more detailed analysis of the functional form of the temperature dependence, for a number of reasons: first, our cryogenic atomic force microscope operates in a helium exchange gas. It is therefore imperative for any quantitative analysis of the MFM data to evaluate the evolution of the mechanical properties of the cantilever as a function of temperature and exchange gas pressure near the superfluid-liquid-gas phase transition. Second, empirically, we observe the presence of temperature-dependent background forces influenced by multiple experimental factors, including the pressure of the helium exchange gas. Unfortunately, as the Meissner force decreases in strength near T_c, the background forces dominate, making it impossible to resolve with confidence a power law behavior in the superfluid density. Quantitative analysis of the temperature dependence therefore requires technical innovations including the construction of an ultra-high vacuum MFM instrument operating at temperatures down to 2K. Commercial systems with these specifications do not exist and the development of the required home-built apparatus is in progress. In summary, the intermediate temperature data validate the expected gradual evolution of the effects we observe, but the precise analysis of the transition dynamics must be postponed until the construction of the new apparatus is complete (~12-18 months).

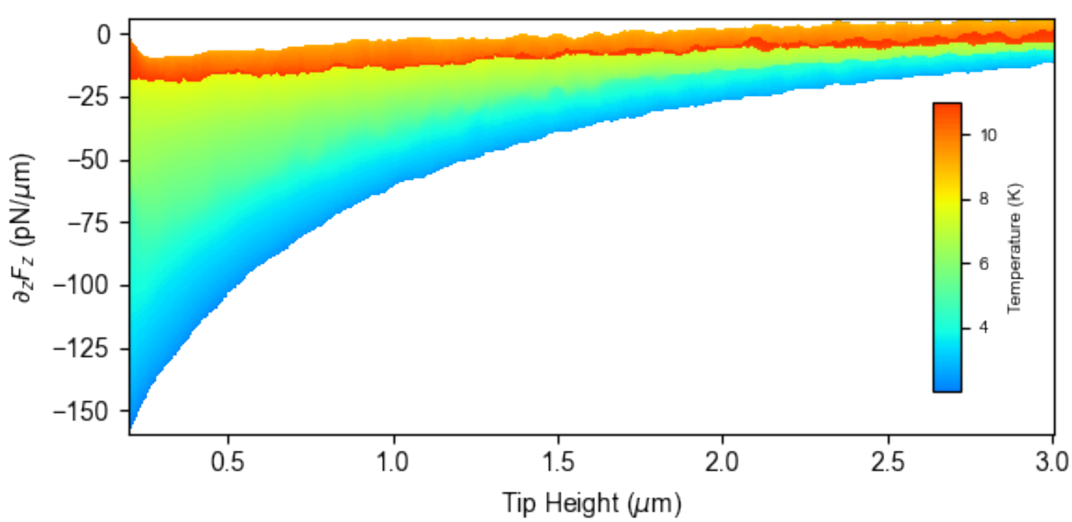

Figure R1 (also Fig. S20): ***Temperature-dependent MFM signal plotted in the form of $\partial_z F_z$ as a function of tip height above an hBN/κ-ET interface:*** *At low temperatures and short tip-sample separations, Meissner repulsion is evident and decays gradually with increasing temperature. Near T_c (11.5 K) and at high tip-sample separations, background forces contribute significantly to the shape of the data.*

4. The exciting possibilities offered by hyperbolic cavities would be more

compelling with additional examples from different material systems to support the claim of broad applicability.

Response 1.4. We thank the reviewer for making this excellent suggestion. Hyperbolic materials span a large range of frequencies. We have revised the last sentence of the manuscript to clarify the variety of systems that host hyperbolic modes.

5. While the authors have thoughtfully considered potential interfacial effects, a more systematic discussion of alternative explanations (such as charge transfer processes) would make the conclusions even more robust.

Response 1.5. We thank the reviewer for the opportunity to enrich our discussion. Charge transfer processes may be expected when dealing with $RuCl_3$, in view of the large work function of this material. Charge transfer at the hBN/κ-ET interface should be much smaller and cannot account for the large suppression of superfluid density observed. Here is why:

A subset of the co-authors have systematically investigated charge transfer at the interface of $RuCl_3$ with various (semi)conductors and insulators [Sternbach, Aaron J., et al. "Quenched excitons in WSe_2/α-$RuCl_3$ heterostructures revealed by multimessenger nanoscopy." *Nano Letters* 23.11 (2023): 5070-5075, Vitalone, Rocco A., et al. "Charge transfer plasmonics in bespoke graphene/α-RuCl3 cavities." *ACS nano* 18.43 (2024): 29648-29657]. These experiments and their first-principles DFT analysis show that any electronic effects at the $RuCl_3$/κ-ET interface (or κ-ET/hBN interface), including charge transfer or interface dipole formation [Rizzo, Daniel J., et al. "Polaritonic probe of an emergent 2D dipole interface." *Nano Letters* 23.18 (2023): 8426-8435], are purely local. If present, the interfacial effects predominantly impact the topmost of layer of κ-ET or only a few layers of κ-ET. Therefore, these interfacial effects cannot account for the suppression of superfluid density over an extended range in the interior of κ-ET. Surface-only modification of the superfluid density in κ-ET is essentially undetectable.

We have added a short discussion of charge-transfer effects in the last paragraph of the "Meissner effect at resonant and non-resonant interfaces" section and a more thorough discussion in a new section of the supplementary material (SI section 14).

Recommended Revisions

• The MFM discussion would benefit from referencing recent advances in this technique for studying novel superconductors and superconducting devices, such as [1, 2, 3, 4, 5]. Including examples of superconducting property modification under various conditions [6] would provide valuable context for interpreting the dF/dz signals.

Response 1.6. We thank the reviewer for alerting us to these works. Although all of the suggested references are indeed relevant, due to the citation limitation, we are unable to cite all six papers. We have added [6], as a study that used MFM to characterize modifications in superconductivity under different conditions. We would be happy to add additional references and are requesting permission from the editor to exceed the reference limit to include references [1-5] suggested by the reviewer.

• Adding examples of interface superconductivity, such as those discussed in [7], would strengthen the discussion of heterostructure effects.

Response 1.7. We thank the reviewer for this suggestion and agree that the interface-engineering of superconductivity is a vibrant field with a rich history that provides relevant context for our work. We have added this citation (citation number 48 in the revised manuscript).

• Including additional temperature-dependent data would help clarify the suppression dynamics across the full temperature range.

Response 1.8. We agree that data across the full temperature range are valuable. As noted in Response 1.3, we have added intermediate-temperature measurements to the SI (Fig. S20 and Fig. R1), which confirm the gradual evolution of the observed effects. A detailed quantitative analysis of the suppressed superfluid dynamics requires the construction of a new home-built instrument.

• Expanding the discussion on potential cavity-induced enhancement of superconductivity, particularly regarding possible mechanisms for Tc increase, would highlight promising future directions.

Response 1.9. We thank the reviewer for this suggestion for improvement. Our work is primarily an experimental one, and as is often the case in emerging fields, a number of theoretical scenarios could potentially be used to explain experimental results that are undeniable. To stimulate further theoretical and experimental exploration, we have attempted in this manuscript to avoid a single full theoretical description of the hBN/κ-ET system. Instead, we describe what we can confidently say about mode coupling in the system and outline multiple theoretical approaches that may explain these results and the potential routes for how this could impact superconductivity (SI section 13).

We have added SI section 13 which addresses an important parameter in cavity-coupled systems, the detuning between the cavity modes and resonances in the target system δ. Detuned cavity-embedded systems have been discussed in the context of atomic, molecular and optical physics [Kelly, Shane P., et al. "Resonant light enhances phase coherence in a cavity QED simulator of fermionic superfluidity." *Physical Review Research* 4.4 (2022): L042032]. In SI section 13, we consider two models for detuned HM modes coupled with a phonon that mediates superconductivity. These lead to both increased and suppressed superfluid density, when considering linear electron-phonon coupling and quadratic coupling, respectively.

Since superconductivity in κ-ET is thought to be unconventional [Buzzi, M., et al. "Photomolecular high-temperature superconductivity." *Physical Review X* 10.3 (2020): 031028] (Ref. 29 in the manuscript), we cannot directly evaluate the effect of the cavity-coupled phonon on superconductivity. Instead, we compare the two models for different types of electron-phonon couplings.

SI section 13 contains Fig. R2 (Fig. S21), where the (T=0) relative change of the superconducting gap $\frac{\Delta-\Delta_0}{\Delta_0}$ is plotted as a function of δ/ω_D, where ω_D is the Debye frequency. Here Δ is the superconducting gap and Δ_0 is the superconducting gap at $\delta = 0$. We calculate the gap for the case of linear electron-phonon coupling (displayed on the left-hand side of Fig. R2) and quadratic electron-phonon coupling (displayed on the right-hand side of Fig. R2).

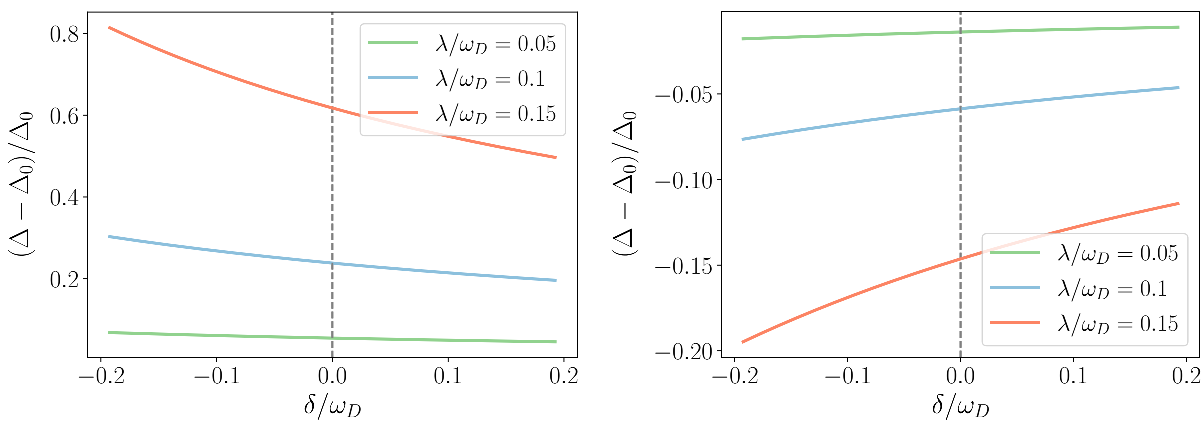

Figure R2 (also Fig. S21): ***Modification of the BCS superconducting gap due to coupling with hyperbolic modes.*** *The relative change of the BCS superconducting gap, $(\Delta - \Delta_0)/\Delta_0$, as a function of the phonon-HM detuning δ, for a couple of different phonon-HM couplings λ. The left panel shows the full result for the case of linear coupling, while the right panel shows the full result for the case of quadratic coupling.*

We find that in the case of linear electron-phonon coupling, the superconducting gap is increased by coupling to the HMs, while in the case of quadratic coupling the superconducting gap is decreased.

• A more detailed consideration of interfacial effects would make the argument for cavity-mediated phenomena even more compelling.

Response 1.10. We thank the reviewer for this suggestion. We have added text expanding possible interfacial effects. We thank the reviewer for raising this point. Although $RuCl_3$'s large work function suggests possible charge transfer, prior studies (see Response 1.5) show that such interfacial effects are highly localized and can impact only the top layers of κ-ET. They cannot explain the extended suppression of superfluid density that has been observed.

• For greater impact, consider moving some of the supplementary data (particularly figure S14 panels a/d or b/e) to the main text, as they provide strong evidence for hBN influence.

<u>Response 1.11</u>. We thank the referee for suggesting this. When originally writing the paper, we decided that the level of detail needed to interpret the data in Fig. S14 (now Fig. S12) might not be appreciated by the non-expert reader. We are worried that at first glance, these data may be interpreted as if the effect depends on the interface quality between hBN and/κ-ET. However, our interpretation of these data is that the effect depends on the orientation between the hBN and κ-ET planes. Our interpretation is supported by the observation that the effect is diminished near sharp wrinkles in hBN, but is not hampered near smooth bubbles (Fig. S10). This distinction is key, and would need to be discussed. Given the word limit, we have decided to leave the figure in the SI.

Overall Assessment

This work represents a landmark advance in the emerging field of cavity quantum materials, demonstrating that vacuum fluctuations in engineered electromagnetic environments can profoundly modify superconducting ground states.
The experimental evidence is comprehensive and supported by sophisticated theoretical modeling. While some mechanistic details warrant further exploration, the study establishes a robust foundation for future investigations into cavity-mediated control of quantum phases.
The implications extend far beyond superconductivity, potentially influencing diverse areas including correlated electron systems, polaritonic devices, and quantum information science. The manuscript is exceptionally well-executed and makes a compelling case for this new approach to quantum material engineering. After addressing the minor points raised above, this outstanding work will be suitable for publication in Nature. The scientific rigor, novelty, and potential impact fully meet the journal's high standards.

<u>Response 1.12</u>. We deeply appreciate this very positive assessment of our work. The reviewer seems to have accepted our interpretation of cavity electrodynamics as responsible for the profound modification of superfluid density. Furthermore, the reviewer acknowledges the generality of our work. We are grateful for the recommendation to publish in *Nature*.

References

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Grebenchuk, Andrey G Shishkin, Olga V Skryabina, Igor A Golovchanskiy, Alexander A Golubov, Nikolay V Klenov, Igor I Soloviev, et al. Revealing josephson vortex dynamics in proximity junctions below critical current. Nano letters, 22(14):5715–5722, 2022.
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Reviewers 2&3

Keren et al. report the observation of a strong suppression of the superfluidity density in the molecular superconductor k-ET at the interface with a layer of hexagonal boron nitride (hBN). The authors perform magnetic force microscopy measurements – which are sensitive to the Meissner effect – to quantify the superfluid density and observe a strong suppression of the superconducting density. The claim of the paper is that this suppression is related to the coupling of the hyperbolic mode of hBN which is close in frequency to the infrared active C=C stretching mode in the ET molecules. This mode is known to be involved in the superconductivity of the compound, as also external drive by resonant mid- infrared pulses seem to suggest (Phys. Rev. Lett. 127, 197002).

The resonant coupling condition between the cavity and the C=C mode is investigated by s-SNOM measurements and by transfer-matrix calculations. The authors also fabricate other “control” devices terminated by RuCl3 – in which optical phonons lie at much lower frequency than C=C mode. The main observation reported is that only resonant hBN cavities contribute to the quench of the superconducting behavior in k-ET.

The experimental finding is remarkable and commendable is the careful and detailed analysis of the effect reported in the supplementary material. The observation reported is the first convincing experimental demonstration that superconductivity can be influenced at equilibrium through the tailored engineering of the electromagnetic environment. If confirmed, this would constitute a major advancement in the development of strategies to control material properties via cavity electrodynamics. Given its broad implications, the study is highly relevant to multiple research communities and aligns well with the wide-ranging interests of Nature's readership.

We thank the reviewers for their positive reviews. We appreciate the reviewers' acceptance of our interpretation of altered superconductivity at equilibrium due to the electromagnetic environment, and their recognition of the relevance of our work to the readers of *Nature*.

However, I find the main conclusion of the paper which attributes the observed superfluid suppression to the resonant coupling of hBN to the C=C mode weak and not entirely supported by the data presented, which makes it difficult to conclusively and univocally link the observation to strong coupling of the cavity with a phonon mode. Underneath a detailed list of major and minor points which the authors should consider prior to publication.

1. The effect measured on the hBN-terminated device is surprisingly large. However, a small suppression is detectable also in the device terminated by the RuCl3 layer. This is particularly evident in the cyan curve in Fig. 3d, for example. The authors seem to dismiss this effect as negligible, but its amplitude is definitely above the noise level and is clearly distinguishable
at low temperature. The presence of this suppression seems at odds with the authors' interpretation which attributes the observed quench of superfluid density to the coupling to the C=C mode and seems to suggest instead that a quench of superconductivity – even if smaller – is achieved also by stacking what the authors called a "non-resonant cavity". The authors should discuss this evidence in more detail.

Response 2-3.1. We thank the reviewers for their insightful comments and for inviting us to elaborate on the effects induced by interfacing $RuCl_3$ and hBN with κ-ET.

The reviewers are correct in pointing out a mild suppression of the superfluid density at the $RuCl_3$/κ-ET interface. There are many effects that could potentially suppress superconductivity at the interface. Strain and sample damage during fabrication are just a couple. Specifically, $RuCl_3$ was transferred at a higher temperature than hBN (110 C and 60 C, respectively). Therefore, we expect any sample damage to be greater near the $RuCl_3$ crystal. In a newly studied device (Device 5), $RuCl_3$ was transferred at an even greater temperature (130 C), with minimal impact on superfluid density in κ-ET (Response 2-3.2). To address the possibility of charge transfer at the interfaces, we have added section 14 to the SI and references to it in the main text (refer also to Response 1.5).

Regarding negligibility, as is evident in Fig. 1, the suppression of superfluid density under $RuCl_3$ is present but is much weaker. To strengthen this point, we have amended Fig. 3 (now Fig. 2) to include a revised panel d. We present this alteration in Fig. R3. The histogram in panel d shows a clear distinction between the observed effects over hBN and values measured over $RuCl_3$.

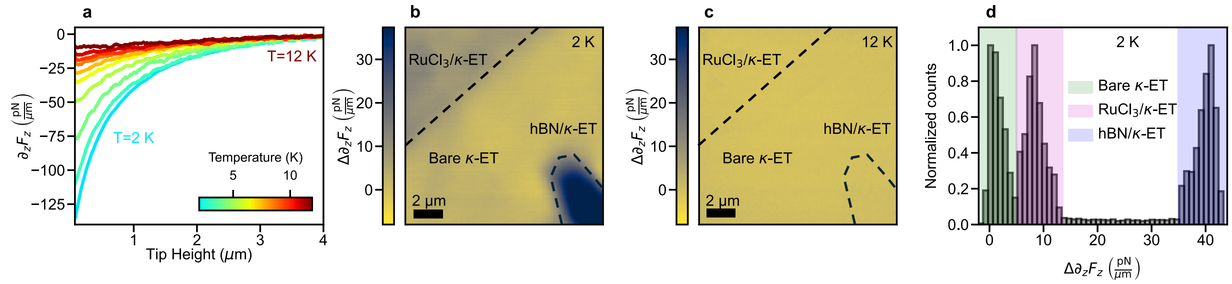

Figure R3: ***The amended Fig. 3 (now Fig. 2):*** *The previous panel d was replaced with a normalized histogram of the values measured in b. The histogram, taken within the field of view of panel b, shows a clear separation between values measured over the area of κ-ET coupled to hBN (Blue shading), over $RuCl_3$/κ-ET (magenta shading) and over unmodified bare κ-ET (green shading).*

2. Following to the comment in point one, in the supplementary material the authors show that inhomogeneity of the hBN/k-ET interface plays a crucial role in the amplitude of the superfluid suppression (Supp. Note 9 and measurements in SI). I wonder if the difference between hBN- and RuCl3-terminated structures could be due to the different qualities of the two interfaces. The presence of only one structure terminated by RuCl3 (device 1) also makes difficult to assess the reproducibility of this result.

<u>Response 2-3.2</u>. We thank the reviewers for bringing up this significant question.

We first address the reviewers' concern regarding the origin of the difference between the two interfaces. Below, we describe arguments that make us believe it is highly unlikely that different interface qualities could explain the difference between hBN/κ-ET and $RuCl_3$/κ-ET devices:

(1) The fabrication steps for depositing the hBN and $RuCl_3$ microcrystals on the κ-ET surface are identical, except for the slight difference in release temperatures. However, if there were any surface damage during the release, the higher release temperature for $RuCl_3$ (60 C for hBN and 110 C for $RuCl_3$, Methods) would suggest that the damage should be worse under $RuCl_3$, when in fact we see a larger effect under hBN. Moreover, the newly studied Device 5 continues to show a minimal effect on superfluid density with even higher 130 C release temperature.

(2) Our evidence suggests that the spatial inhomogeneity in the suppression of superfluid density under hBN is not due to any arbitrary type of variation in the interface but instead arises from specific conditions. For convenience, here we refer to the areas of hBN as being "effective" where the superfluid density is strongly suppressed and "ineffective" where the superfluid density is minimally impacted by hBN. Bubbles form between hBN and κ-ET that produce hBN-κ-ET separations near 10 nm (Fig. S10). However, these smooth deformations constitute "effective" areas of hBN. From this observation, we conclude that nanoscale separation between κ-ET and hBN does not reduce

effectiveness. By contrast, sharp wrinkles in hBN produce “ineffective” areas. These may arise from modifications to the HM dispersion near the wrinkles or altered orientation between the hBN and κ-ET planes. Because the $RuCl_3$ is sitting flat on the κ-ET surface, and we do not observe any of the types of ineffective interface morphologies that we observe in hBN, there is no evidence for poor interface quality in the $RuCl_3$/κ-ET area. The data instead supports good interface quality.

Second, we address the reviewers’ concern of reproducibility. The $RuCl_3$ flake used in Device 1 is of large area (a few 10s of μm^2). Using hBN samples with similar areas, we observed unquestionable suppression of superfluid density. In addition, we have fabricated and measured the additional $RuCl_3$/κ-ET device (Device 5). In Device 5, we once again observed that $RuCl_3$ had only a weak effect on the superfluid density in κ-ET. We present large-area MFM images for Devices 1 and 5, along with their corresponding topography images, in Fig. R4 (Fig. S13).

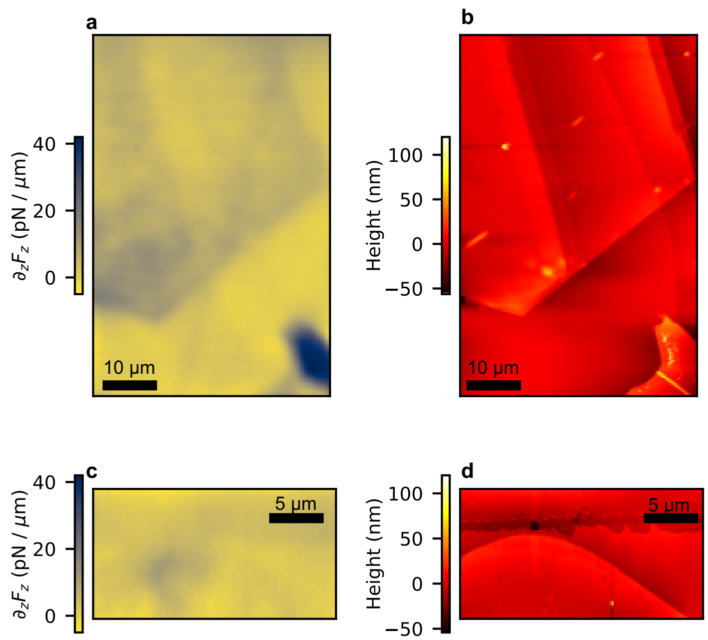

*Figure R4 (also Fig. S13)**: Large-scale imaging of RuCl₃**. a, Constant-height MFM. The dominant signal originates from the hBN/κ-ET interface. RuCl₃ shows significantly weaker suppression over a large scale. Notably, hBN crystals over similar length scales have shown significant suppression of superfluid density. b, Topography image corresponding to the MFM image. c, Constant-height MFM (lift=300 nm) above an additional RuCl₃/κ-ET device. The color scale is shared with a. d, Topography corresponding to c.*

To further stress the weak effect of $RuCl_3$, we performed measurements of $\partial_z F_z$ as a function of tip height (as in Fig. 1) above the $RuCl_3$/κ-ET interface and above bare κ-ET in the new Device

5. We find nearly indistinguishable traces (Fig. R5), reproducing the results in the manuscript and highlighting the weakness of the suppression under $RuCl_3$. The fitting procedure outlined in SI section 5 results in a suppression of superfluid density of roughly 4%. This result is included in SI section 15 and the resulting suppression of superfluid density is included as a new data point in Fig. 3.

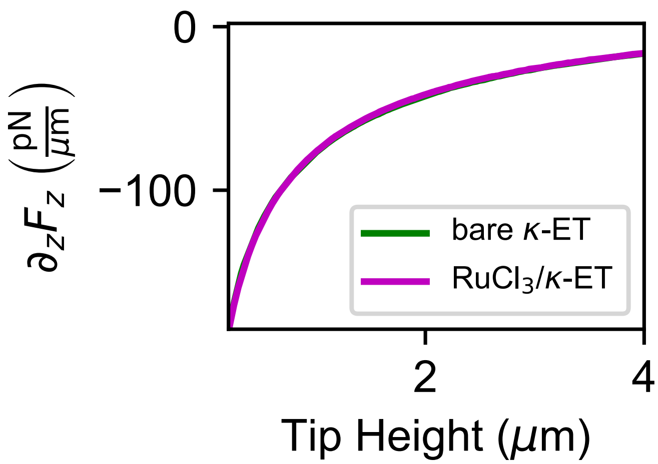

Figure R5 (also Fig. S22): ***$\partial_z F_z$ as a function of tip height above the $RuCl_3$/κ-ET interface in Device 5.*** *Data collected on the new Device 5. The representative traces above bare κ-ET and above $RuCl_3$/κ-ET are nearly indistinguishable. Each measurement was taken with a tip bias chosen to minimize work function disparities.*

To summarize, in response to the reviewer's comments, we have included Figs. R4 and R5 in the SI sections 8 and 15 with complementary text and added the data point for Device 5 in Fig. 3.

3. As the authors discuss, the hBN hyperbolic dispersion results in a higher photonic density in the mid-Infrared. This frequency is in the region where the effects associated to the cavity-mediated radiative coupling between the external photonic bath and the sample are expected to give sizeable thermodynamical effects (Phys. Rev. B 111, 165425 and Nat Chem 17(3):439-447). A possibly relevant observation in this sense is that the optical phonons of RuCl3 – lying at lower energy - would be less sensitive to this effect, which could provide a mechanism for the cavity suppression of the superfluid density which do not require strong coupling. Could the author comment on this?

Response 2-3.3. We thank the referee for this astute interpretation of our experiments. The referee raised an interesting point concerning the heat exchange between the cavity and the external reservoir, which can alter the nominal temperature of the material inside the cavity.

Following the reviewer's comment, we have added a citation in the revised manuscript (citation 13).

4. The authors reports also measurements on hBN/BSCCO structures. The lack of superfluid suppression is used as an argument to confirm the contribution of the resonant coupling with the C=C mode. While in principle these measurements are interesting, I don't find them particularly relevant to substantiate their claim regarding the relevance of strong coupling with C=C modes. The emergence of superconductivity in BSCCO occurs for different (still unclear) reasons. There are no resonances close to the hBN hyperbolic modes that are thought to be implicated in the superconducting transition in BSCCO. As such, showing that hBN does not prompt any superfluid change in BSCCO has no implications in the interpretation of the results in k-ET.
A side point related to these measurements. In the inset of Fig. 1b, it seems that the MFM measurements on bare BSCCO (grey) are much noisier than the ones performed on hBN/BSCCO (blue). Can the authors comment on that?

Response 2-3.4. We thank the reviewers for sharing their perspective. We believe our MFM data for hBN/BSCCO are valuable in several respects. First, MFM studies of superconductivity residing under hidden interfaces are novel. It is therefore imperative to document our ability to reliably probe such hidden interfaces by means of MFM. BSSCO is a canonical layered superconductor and thus serves as a natural choice for documenting across-interfaces MFM data. Second, BSSCO does not host "*resonances close to the hBN hyperbolic modes that are thought to be implicated in the superconducting transition*," as pointed out by the reviewers. In the manuscript, we merely report on these facts and let the reader draw their own conclusions.

Regarding the referee's comment on noise: The bare BSCCO curve is plotted using a wider line for visual clarity. The noise in the two curves is comparable. We have added a clarifying statement in the caption to Fig. 1 regarding the line width.

5. I encourage the authors to include in Fig. 3 the dF curves as a function of the tip eight for different temperature in the hBN/k-ET device (basically, a copy of Fig. 3a but for the cavity). A comparison of this data with the one measured in bare k-ET would be important to understand the trend of the superfluid suppression within the cavity as a function of temperature. Moreover, it would be interesting to compare the same temperature-dependent curves for the RuCl3-terminated structure. Is the trend the same?

Response 2-3.5. We thank the reviewer for this insightful comment. We report the intermediate-temperature data (for hBN/κ-ET) in the revised SI (Fig. S20, reproduced in Response 1.3 as Fig. R1). A detailed analysis is not feasible because background forces—arising from multiple experimental factors, including the helium exchange gas—vary strongly with temperature and dominate as the Meissner force weakens near T_c. This background thus obscures the temperature-dependence of superfluid density near T_c. Resolving this limitation will require a custom UHV system. For now, the data confirm the expected gradual evolution across intermediate temperatures. $RuCl_3$/κ-ET traces at 2 K are nearly indistinguishable from data for unobscured κ-ET. For that reason, and the inability to undertake a detailed quantitative analysis, we have not measured the full temperature dependence above the $RuCl_3$ terminated structures.

6. The data presented in Fig. 1 have been taken for a k-ET sample 200 um thick. Does the thickness of the k-ET have any impact on the amplitude of the suppression measured after hBN encapsulation? Did the authors measure also hBN cavities in which the k-ET samples have different thicknesses?

Response 2-3.6. We thank the reviewer for this excellent idea. In practice, thinning down κ-ET to the few-hundred-nm limit in a controlled manner remains extremely challenging. We have explored exfoliation techniques, and while thin fractured κ-ET crystals are occasionally obtained (with extremely low yield, e.g., Device 2 is one example), we have found these samples to be technically incompatible with MFM measurements. In general, κ-ET is not readily amenable to exfoliation and/or other processing methods. We anticipate that technical innovations, through, for example, focused ion beam (FIB) lithography, will make this possible to address in the future. However, the development of FIB protocols is definitely outside the scope of the present work.

7. Did the authors check the quality of the structures over time? Did they detect any sign of degradation? While the manuscript is generally well written, the following input could improve the readability of the manuscript.

Response 2-3.7. We thank the reviewer for their comment. MFM measurements required months of data acquisition for each device over multiple sessions. No degradation was detected. In many cases, the samples were thermally cycled multiple times, and no degradation was found. Furthermore, we used both freshly cleaved and as-grown κ-ET surfaces, and both showed comparable effects. This lack of degradation is in agreement with literature reporting the stability of the κ-ET surfaces [Rohr Eur. Phys. J. B 69, 167 (2009)]. We have added these comments in the Methods section on device fabrication.

8. In the x-axis label of Fig. 4, the authors use the word "cavity thickness", which I find a bit misleading as it might hint to a change in the cavity resonance.

Response 2-3.8. We thank the reviewers for pointing out this possible source of confusion. We have switched the label to "hBN/$RuCl_3$ thickness."

9. I personally find the structure of the paper suboptimal at the moment. The reading flow is somehow interrupted between Fig. 1 and Fig. 2. I would rather swap Fig. 2 and 3 to present first all the Meissner data and discuss then the phonon-polariton dispersion.

Response 2-3.9. We appreciate this comment from the reviewer. We agree with the spirit of reviewer's suggestion and have reordered diagrams and the text. The changes that have been made are:
Fig. 1 -> Fig. 1
Fig. 2 -> Fig. 4
Fig. 3 -> Fig. 2
Fig. 4 -> Fig. 3

10. It would be helpful for the reader to mention in the caption of Fig. 1a,b the temperature at which the measurements have been performed.

Response 2-3.10. We thank the reviewers for pointing this out. We have added the temperature in the caption.

11.The authors might want to include the citation to a recent relevant contributions in the field DOI: 10.1126/science.abl5818, which i think is missing.

Response 2-3.11. We thank the reviewer for noticing this omission. We have introduced this citation in the main text (citation 16 in the revised version).

Overall, even though I find that the current version of the paper needs some revision, the experimental evidence reported is really remarkable and definitely worth to be considered for publication in Nature Journal.

Response 2-3.12. We thank the reviewers for finding our evidence remarkable, and worthy of consideration for publication in *Nature*. We have addressed key points made by the reviewers and added additional auxiliary data to both the supplemental information and the main text to address the reviewers' concerns.

--- Report by Reviewer #4 ---

Please find below my review on manuscript 2025-05-13674, entitled "Cavity-altered superconductivity", by I. Keren et al.

The authors present exciting and, importantly, convincing results on the modification of the superfluid density of a molecular superconductor via the phonon polariton modes of a resonant hyperbolic medium (specifically, hexagonal Boron Nitride, hBN). Using magnetic force microscopy and scattering-type near-field optical spectroscopy, they carry out a detailed, muti-faceted analysis of a series of devices in which a slab of hBN (of thickness on the order of 20-100 nm) is placed on top of a thick slab of a specific molecular superconductor (κ-ET). The key idea of this work is that the phonon mode which is responsible for superconductivity in κ-ET falls in the range of the hyperbolic phonon polariton modes of the hBN slabs. This leads to mode hybridization between κ-ET and hBN, which in turns leads to a transfer of spectral weight and the subsequent modification of the superfluid density of the bulk superconductor. Let me state upfront that I am truly impressed by this manuscript for three essential reasons:

i) The authors have reported a wealth of results obtained in control experiments where the above mentioned resonance condition is not met;

ii) The main finding of the authors (suppression of the superfluid density) does manifest in a variety of devices where hBN slabs have a thickness that differs by a factor that can be as large as 5;

iii) Experimental data are corroborated by a theoretical analysis of mode coupling.

Also, the manuscript by Keren et al. is well written and the results seem convincing. This is a key, milestone result in a field (cavity materials engineering) which, so far, has been essentially (and unfortunately) driven by theory. Boosting the critical temperature of a superconductor via the polaritonic modes of a passive (i.e. dark) cavity is THE holy grail of the field. In this manuscript, the authors show that polaritons can suppress the superfluid density of an ordinary BCS superconductor. I therefore consider these results as a key step towards the holy grail.

I therefore strongly recommend this work for publication, with absolutely no hesitation. I am 100% sure that this work will stimulate many more experimental results. Inspired by this work, indeed, we will see soon direct measurements of the superfluid stiffness of 2D superconductors as modified by resonant dielectric engineering. Hopefully in all-van-der-Waals stacks.

We thank the reviewer for these extraordinarily positive comments. We are delighted with the reviewer's assessment of our accomplishment as a "key step towards the holy grail" of the cavity quantum materials field. The reviewer suggests that our work will stimulate many more experiments and recommends our work for publication in *Nature* with "absolutely no hesitation."

I have only two important remarks for the authors, which should be considered before publication:

1) Reading the manuscript, I found a little bit of disconnect between the ultra-brief

description of the theoretical figure Fig. 2(a) and the experimental data shown in Fig. 2(c). Could the authors explain better the link between the kink in the fifth fringe, which is present in the experimental data, and the clearly visible hybridization between the weakly dispersive κ-ET phonon mode and the highly dispersive high-momentum hBN phonon polariton modes? Also, why does the kink appear only in the fifth fringe? For a broad audience, it would be probably beneficial to be reminded about SNOM fringes in bare hBN too, but I do understand that space is an issue here.

Response 4.1. We thank the reviewer for these comments. In the revised manuscript, we carefully addressed the reviewer's concerns in SI section 10.2.

The interference fringes represent a standing wave pattern produced by propagating hyperbolic phonon-polaritons (HPhPs), the manifestation of the HMs when the metallized tip is externally illuminated. These fringes are affected by the hybridization between the HPhPs and the C=C stretching mode, which causes a change in the HPhP wavelength near the frequency resonant with the C=C mode. This variation of the wavelength manifests as a kink in the real-space experiments, as the HPhP interference fringes shift at the resonant frequency. Each fringe is impacted by the interaction between the HPhPs and the κ-ET molecular resonance, but the effect is cumulative and thus easier to observe for polaritons that travel longer distances [Hillenbrand, Rainer, et al. "Visible-to-THz near-field nanoscopy." *Nature Reviews Materials* 10.4 (2025): 285-310]. Indeed, assuming a wavelength change of $\delta\lambda$, after n oscillation periods, the total travel length difference is n$\delta\lambda$. Therefore, fringes associated with larger propagation distances are more strongly impacted by the anomalies in the (ω,q) dispersion. For this reason, in Fig. 4c we mark the dashed line at the fifth peak of the propagation profile. We note that this strategy is quite common in the near-field studies of subtle changes in polaritonic profiles (see for example an article on the subtle changes in plasmon-polariton wavelength caused by Fizeau drag: Fig. 2c-d in Dong, Yinan, et al. "Fizeau drag in graphene plasmonics." *Nature* 594.7864 (2021): 513-516).

To fully address the reviewer's comment, we have added Fig. R6 (Fig. S17). In this figure, we plot the phonon polariton dispersion of bare hBN (panel a) for comparison with the coupled case (panel b, which is also Fig. 4a). Using the dispersions in panels a and b we simulate the real space s-SNOM profiles (panels c and d). We trace individual fringes to show that (1) real-space kinks do not appear in the hBN/SiO_2 case and (2) the kink in the hBN/κ-ET case is cumulative and most apparent in the higher-order fringes. We have added text describing these principles in the SI section 10, accompanying Fig. S17.

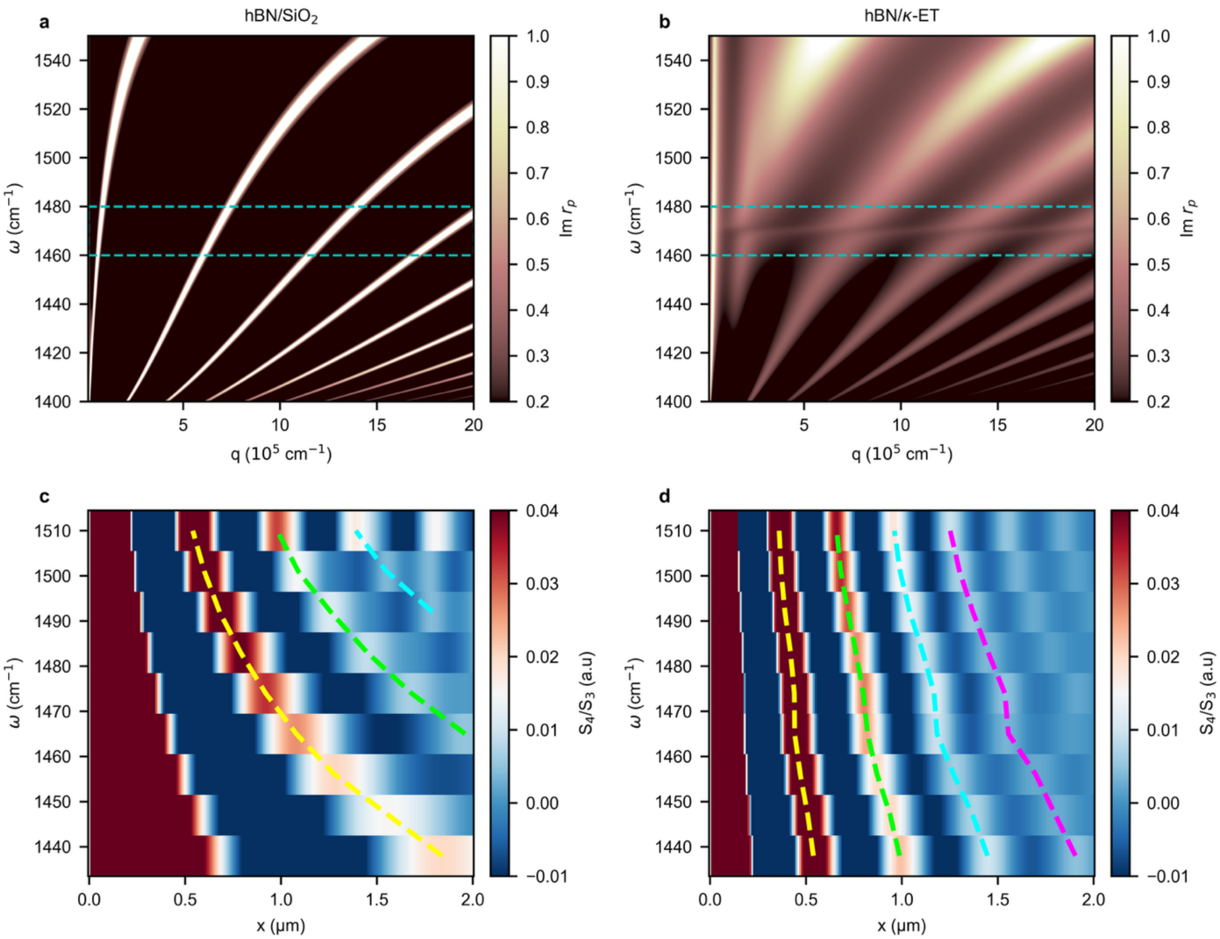

Figure R6 (also Fig. S17): ***Polaritonic dispersion and mode hybridization in hBN/κ-ET structures.*** *Panels a,b: dispersions for the hBN/SiO_2 and hBN/κ-ET cases. The details of these calculations are described in SI section 10.2. Panels c,d: real-space s-SNOM experiment simulations corresponding to the dispersion plots in panels a,b.*

2) When I write papers about "cavity quantum materials", I typically get complaints from people (perhaps from people in quantum optics community?) asking me "Where is the cavity effect?" I do perfectly understand what the authors mean by "cavity" with reference to hBN slabs. Don't get me wrong. But, for a broad audience, I think that a bit of an explanation is needed here. For example, a real microscopic theory of the effect should explain the little dependence of the superfluid density suppression on the hBN thickness seen in Fig. 4. Any comment on this? I do strongly suspect that the findings of the authors would theoretically persist in the limit of large hBN thickness, which is in perfect agreement with the subwavelength theory discussed in Ref. 28 and in the following paper https://arxiv.org/abs/2403.20067. I do understand that picking up a signal with MFM in the limit of "bulk" hBN is impossible, but the conceptual (theoretical) question remains here. I believe that data in Fig. 4 (thickness

dependence) are telling us something important. In my humble opinion, they are clearly indicating that the scalar (electric field) potential coupling is playing a key role here. Am I wrong? If, on the other hand, the vector potential coupling would play a dominant role, the effect would rapidly disappear with increasing hBN thickness (see for example Fig. 3 of https://doi.org/10.1103/PhysRevLett.130.216901). Could the authors clearly comment on this in their work? The field is mature enough (from the theoretical point of view) and therefore we do need to extract fundamental information from experiments at this point, either pointing towards the importance of the scalar potential coupling or against it (and therefore pointing towards the importance of the vector potential coupling). Physics is of course gauge invariant, but the "natural" description of subwavelength (e.g. hBN) polaritonic cavities is in terms of the scalar potential coupling, which is precisely defined provided that one picks up the generalized Coulomb gauge suggested for example in Ref. 28 and references therein to earlier work. I would appreciate an answer on this point. In this respect, the authors could consider adding a citation to https://arxiv.org/abs/2403.20067 (to appear, PNAS), where a discussion of this point for the case of the amplitude of the SdH oscillations is reported.

Response 4.2. We thank the referee for the great points raised here.

To expand on the notion of hBN slabs acting as cavities for a broad audience, we have added an SI section (new SI section 3). In this section, we address the cavity aspects of hBN from two complementary perspectives: (1) In SI section 3.1, we address the frequency-selective enhancement of the photonic density of states and electric field fluctuations, and (2) In SI section 3.2, we address the mode response of the hBN cavity to oscillating dipolar fields. Below, we describe both in more detail.

In SI section 3.1, we present the dispersion of photonic modes in hyperbolic hBN alongside the results for a canonical Fabry-Pérot (FP) cavity. Fundamentally, all cavities modify the dispersion of light from free space (panel b) towards the more complex behavior exemplified in panels e and h. Altered dispersion implies a frequency-selective enhancement in the photonic partial local density of states (PLDOS), as shown in panels c, f, and i. The fundamental frequency of the FP cavity ω_0 is determined by the dimensions of the resonator d; the fundamental frequency of the hyperbolic cavity we employ is dictated by materials resonance. The hBN slab achieves far larger PLDOS enhancements, and these enhancements extend outside the slab surface (panel i, displaying PLDOS at various distances z away from the hBN surface).

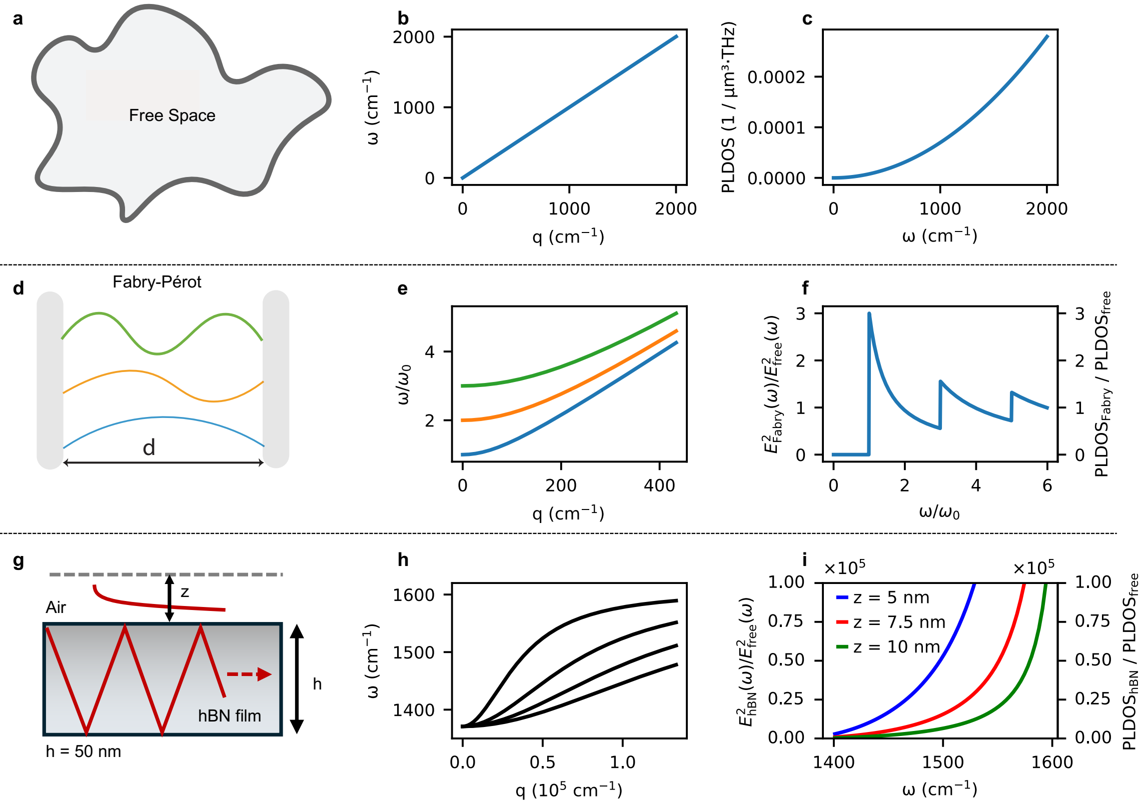

Figure R7 (also Fig. S3): ***Electromagnetic cavities: hBN and Fabry-Pérot resonators****. (a-c) dispersion (b) and PDOS (c) of photons in free space. (d-f) dispersion (e) and electric field fluctuation relative to the free-space fluctuations (f) in a Fabry-Pérot setting. The colored curves in e correspond to the cavity modes sketched in d. The second y-axis in (f) displays the PDOS relative to the free-space PDOS. (g-i) dispersion (h) and electric field fluctuation relative to the free-space fluctuations (i) in an hBN slab setting. The second y-axis in (i) displays the PDOS relative to the free-space PDOS. The curves are calculated at various heights above the hBN surface.*

In SI section 3.2, we model the real-space patterns associated with confined modes in an hBN slab. We calculate the electric field induced by a nearby oscillating dipole. When the dipole oscillates with a frequency within the range of hyperbolicity, HPhP modes are confined to the hBN slab. We believe this is a highly intuitive illustration of the cavity behavior of hBN that will be easily accessible to readers.

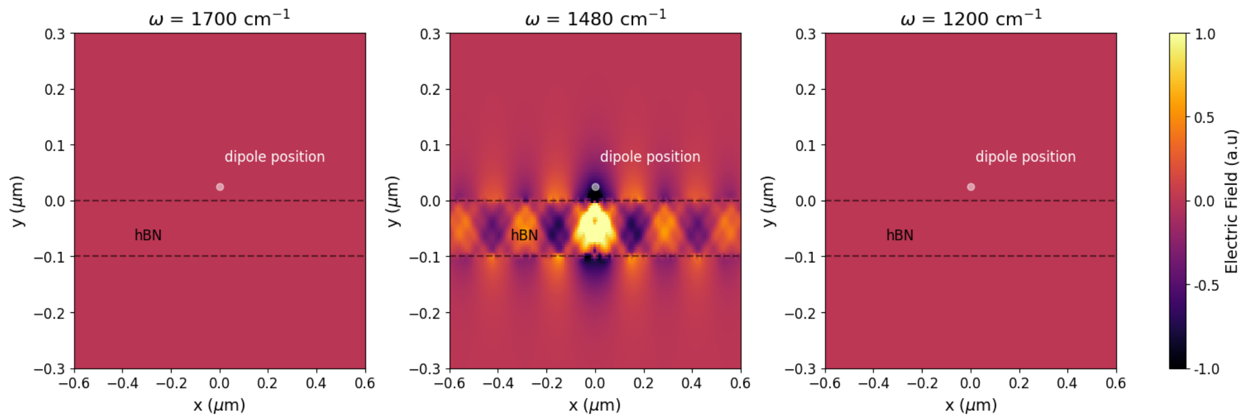

Figure R8 (also Fig. S4): ***Simulation of the electric field of a dipole near an hBN slab.*** *Notably, at ω=1480 cm^{-1} (within the hyperbolic range) confined modes appear within the hBN slab (black dotted lines). This figure was added to SI section 3.*

Let us clarify several points that are important when addressing the central question raised by the referee.

First, we are discussing the interaction of the HMs of the hBN cavity, which contain both in-plane and out-of-plane evanescent electric field components. The out-of-plane (E_z) component is particularly relevant here, as it mediates the coupling between the different HM bands of hBN and the C=C stretching mode of κ-ET. Since the κ-ET is not metallic along the out-of-plane direction, the fluctuating E_z field of the HMs should penetrate into the bulk of the κ-ET, enabling direct coupling to the IR-active C=C stretching mode (supported by Figs. R7, R8). This coupling arises predominantly from the scalar potential component of the electric field, as the referee correctly pointed out.

The mechanism we propose requires well-defined hyperbolic phonon-polariton branches, rather than Fabry–Pérot–geometric type cavity confinement. In hBN, such modes persist over a broad range of thicknesses. In our case, the long-range E_z field of the HMs enables direct polaritonic coupling to molecular vibrations.

Second, at large distances—or in non-subwavelength cavities—the transverse component of the field makes a significant contribution, unlike in subwavelength cavities, where the transverse component is typically overshadowed by the dominant longitudinal component.

Regarding the choice of gauge: Our simulations of the reduction of the C=C amplitude utilize the physical dipolar electric fields and hence are gauge agnostic. Therefore, we do not believe that our modeling has any bearing on the gauge choice.

Following the reviewer's comment, we have added SI section 3 in the revised manuscript, as well as a citation to the PNAS paper suggested by the reviewer (citation 14 in the manuscript).

In summary, and for the reasons stated above, I very strongly believe that the manuscript by Dr. Keren and co-workers represents a key, outstanding result in the field of cavity quantum materials and that it needs to be accepted and published with the uttermost urgence, provided that the two comments above have been taken into account by the authors.

We thank the reviewer for the very highly positive assessment of our work.

Summary

We have addressed every concern raised by the reviewers and added multiple figures to the SI.

Key concerns addressed:

- Readability: We have restructured the paper. In the revised paper, Figs.1-3 concern MFM measurements and Fig. 4 concerns optical measurements and theory. We have revised panel d in Fig. 3.
- Reproducibility of the weak signal observed over $RuCl_3$ signal: We have fabricated and measured an additional $RuCl_3$/κ-ET device (Device 5), which has reproduced the weak effect. The additional data point was added to Fig. 3.
- Theoretical suggestions for enhancing superconductivity: We have provided an additional model in SI. The model predicts either enhancement or suppression of the superfluid density directly via selective hybridization between the HM and C=C mode. These are discussed in the context of possible pathways to cavity-enhanced superconductivity.
- Temperature dependence of the hBN/κ-ET interface: We have provided the requested data in the the SI.

We are happy to submit the revised version of our manuscript for publication in *Nature*.