

Thermally stable 2D YMnO₃ enabling blue visible camouflage with mid-infrared transparency

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

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The manuscript entitled “Thermally stable 2D YMnO₃ enabling blue visible camouflage with mid-infrared transparency” addresses an important challenge, namely achieving visible coloration together with mid-infrared (MIR) transparency under high-temperature conditions. The proposed non-equilibrium strategy to decouple visible and phonon-mediated optical responses is conceptually interesting, and the experimental results are generally convincing. Overall, the work is timely and relevant, and I would recommend publication after minor revision.

The analysis of visible coloration would benefit from a more complete optical treatment. While CIE coordinates are derived from reflectance spectra, no optical constants or angle-dependent measurements are provided. The perceived color may change with viewing angle due to interference and substrate effects, which could represent a limitation for practical applications. In addition, it would be beneficial to include the dependence of color on material thickness, as this can also influence the observed appearance. Providing optical constants would allow calculation of the color for arbitrary angles within the CIE framework, or at least these limitations should be discussed.

The comparison with prior work is generally appropriate; however, it would benefit from a broader context. The current discussion focuses mainly on pigments and oxide systems (Table S1), while other relevant material classes are not considered. In particular, ultrathin transparent conductive or plasmonic metallic films, which can also exhibit combined visible and infrared optical functionality, are not discussed. Including a comparison with recent work on ultrathin high-quality gold films (e.g., Nano Letters, DOI: 10.1021/acs.nanolett.4c04311) would help to better position the present results within the wider field.

I also note that the wavelength–color mapping in Fig. 4b,c appears slightly incorrect. Given that the manuscript already employs the CIE framework for color analysis, it would be more appropriate to ensure that the color mapping is consistent with CIE standards. In addition, the manuscript would benefit from minor language corrections to improve clarity and readability.

In summary, this is a strong and interesting manuscript that would benefit from additional clarifications. With these revisions, the work would be suitable for publication.

Reviewer #2

(Remarks to the Author)

In the manuscript “Thermally stable 2D YMnO₃ enabling blue visible camouflage with mid-infrared transparency” Zhu et al. explore the realization of a 2D hexagonal YMnO₃ system through microwave-shock synthesis, featuring high thermal robustness, a blue visible appearance, and MIR transparency, enabling the decoupling of visible and infrared optical constraints in thermally stable oxides.

The work is well presented, with a very extensive study of the material properties, and it can be of great interest for the scientific community. However, I believe that with some integration, the manuscript can be made clearer. Therefore, I recommend publication in the journal, provided that the authors make the further integrations outlined below.

- First, no discussion is provided regarding the environmental stability of the materials. Is material degradation possible

under certain conditions? Could the authors clarify this point in more detail?

- Do the authors have any statistics regarding the characteristics of the films synthesized under the same experimental conditions?

- All the measurements reported in this work were obtained using a reflection configuration. However, I believe that measuring the transmittance of the material would also provide valuable support for the significance of the results.

- The mechanism behind the suppression of the LO–TO splitting is not clear; can the authors quantify this effect to support this claim?

- Minor Remarks: The figures are generally a bit confusing. It might be helpful to correct a few typos (e.g., “spectrally stable, spectrally stable” or “Thata” in Fig. 2) and to improve the overall writing style. Additionally, adding more details about the synthesis process could strengthen the manuscript.

Reviewer #3

(Remarks to the Author)

This manuscript presents a hexagonal 2D YMnO₃ system and argues that, through microwave-shock-enabled non-equilibrium synthesis, the material can simultaneously exhibit a blue visible appearance, mid-infrared transparency, and high-temperature robustness.

The main point of the work is that dimensional confinement and retained rigid Mn–O coordination provide a structure-based route to mitigate the long-standing conflict between visible coloration and MIR optical constraints, and most of the manuscript is organized around structural validation and cross-spectral optical/thermal characterization in support of this claim.

The topic is interesting and potentially important for high-temperature multispectral camouflage. However, before the work can be considered for publication, several key points should be clarified.

- The XAFS, FTIR, and Raman results support that the local Mn–O coordination is retained and are consistent with the authors' short-range bonding picture. However, these data alone do not seem sufficient to fully support the broader claim that the conflict between visible coloration and MIR optical constraints is resolved, since the long-range polarization contribution and the LO–TO splitting, which are central to the authors' explanation, are not directly shown here.

- One important point that should be clarified is whether the manuscript demonstrates MIR transparency of 2D YMnO₃ itself, or rather that the coating does not significantly disturb the low-emissivity radiative boundary of the underlying metallic substrate. As presented, much of the optical and thermographic evidence seems to support the latter interpretation, since the results are mainly discussed in terms of the coating/substrate composite and the metallic background. The authors should therefore clarify whether “MIR transparency” is being used as an intrinsic material property or as an effective behavior in the present coating geometry.

- The manuscript makes repeated use of apparent temperature from infrared thermography to support the optical performance. Since much of the performance argument relies on this thermographic contrast, it would strengthen this part if the authors more explicitly relate the observed temperature differences to the measured MIR reflectance/emittance. In particular, it would be helpful to clarify how the effective emissivity of the coating/substrate system is expected to translate into the reported apparent-temperature differences under the present measurement conditions.

Reviewer #4

(Remarks to the Author)

Manuscript # NCOMMS-26-019705

Title: Thermally stable 2D YMnO₃ enabling blue visible camouflage with mid-infrared transparency

General Evaluation:

The manuscript presents an interesting and potentially impactful approach to achieving blue coloration with MIR transparency in thermally stable oxides. The experimental work is comprehensive and relevant for high-temperature camouflage applications. However, the central mechanism is largely supported by indirect evidence, with insufficient validation of phonon-related claims. Additional quantitative analysis and stronger justification are needed.

The manuscript presents interesting results and is generally well-structured. After addressing the highlighted concerns and improving the clarity of the mechanistic explanations, the work will be suitable for publication in Nature Communications following revision.

1. The manuscript claims decoupling of visible coloration and MIR response—how does this fundamentally differ from prior phonon-engineering approaches in polar oxides?

2. Can the authors quantitatively compare their strategy with existing blue pigments or MIR-transparent coatings to clearly establish novelty?

3. Is the suppression of LO–TO splitting experimentally verified or only inferred from indirect observations?

4. The claim of decoupling visible coloration and MIR response is central; however, no direct measurement of LO–TO splitting is presented. This should be experimentally validated.

5. Can the reduction in long-range polarization be directly evidenced (e.g., dielectric function or Born effective charge analysis)?

6. The manuscript attributes MIR transparency to suppressed Reststrahlen band expansion, but dielectric function(ϵ_1 , ϵ_2) data are not provided. This is a critical missing link.
7. The absence of DFT or phonon dispersion calculations makes the mechanistic explanation largely speculative. Can the authors provide theoretical validation?
8. How reproducible is the microwave-shock synthesis in terms of phase purity and nanosheet thickness?
9. What is the scalability and batch-to-batch consistency of the non-equilibrium synthesis method?
10. The reported thickness (~0.7 nm) suggests a monolayer structure, but no direct crystallographic or layer-number confirmation (e.g., cross-sectional TEM or AFM statistics) is provided.
11. The proposed mechanism relies on reduced *Born effective charge (Z^*), yet no quantitative or computational evidence is shown—this weakens the core argument.
12. Can the authors clarify the role of glucose-derived carbon and whether residual carbon affects optical properties?
13. The high-temperature stability test (8 h at 100 °C, 900 °C annealing) is limited—long-term cycling and oxidation resistance are not sufficiently explored.
14. The ethanol flame test introduces carbon contamination, which the authors acknowledge—this raises concerns about the reliability of post-test optical data.
15. The coating performance is demonstrated on controlled substrates, but real aerospace conditions (mechanical stress, erosion, oxidation) are not simulated or discussed.
16. The authors are encouraged to include relevant recent literature in the Introduction to strengthen the scientific background and highlight the current significance of the study.

<https://doi.org/10.1016/j.ceramint.2025.09.340>
<https://doi.org/10.1016/j.jallcom.2025.180821>
<https://doi.org/10.1103/PhysRevMaterials.9.024408>

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

I thank the authors for their detailed and constructive responses to my comments. The revisions satisfactorily address the main concerns raised in my review.

Overall, I am satisfied with the authors' revisions and recommend acceptance of the manuscript after routine editorial checks.

Reviewer #2

(Remarks to the Author)

The authors have provided clear and detailed responses, and all the revisions/integrations from the previous round have been adequately addressed. The manuscript now presents improved quality and clarity. Therefore, in my opinion, it can be considered for publication.

Reviewer #3

(Remarks to the Author)

The authors have revised the manuscript well and addressed the reviewer comments satisfactorily. The manuscript entitled "Thermally Stable 2D YMnO₃ Enabling Blue Visible Camouflage with Mid-Infrared Transparency" is suitable for publication and can be accepted.

Reviewer #4

(Remarks to the Author)

I have carefully evaluated the revised manuscript and am satisfied with the authors' revisions. In my opinion, the concerns raised during the review process have been adequately addressed. I am pleased to recommend acceptance of the manuscript for publication.

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Response and List of Changes to Referees' Comments

We sincerely thank you for the careful and thorough evaluation of our manuscript entitled “*Thermally stable 2D YMnO₃ enabling blue visible camouflage with mid-infrared transparency*”. We are grateful to the Editor for providing us with the opportunity to revise and improve this work, and to all reviewers for their constructive, thoughtful, and valuable comments. We have carefully considered all the comments and suggestions and have revised the manuscript extensively accordingly. In the revised manuscript, we have clarified the central scientific claims, strengthened the supporting analyses and discussions, improved the overall presentation, and corrected the issues identified during peer review. For clarity, all changes have been highlighted in the *Revised Manuscript*, and our detailed point-by-point responses to each comment are provided below.

Response to Referees' Comments

◆ Reviewer #1:

General Comment: The manuscript entitled “Thermally stable 2D YMnO₃ enabling blue visible camouflage with mid-infrared transparency” addresses an important challenge, namely achieving visible coloration together with mid-infrared (MIR) transparency under high-temperature conditions. The proposed non-equilibrium strategy to decouple visible and phonon-mediated optical responses is conceptually interesting, and the experimental results are generally convincing. Overall, the work is timely and relevant, and I would recommend publication after minor revision.

Response: We are grateful to the reviewer for the positive overall assessment of our manuscript. We appreciate the reviewer’s recognition of the significance of addressing visible coloration together with MIR transparency under high-temperature conditions, as well as the conceptual interest and experimental validity of the proposed strategy. We have carefully considered the reviewer’s comments and revised the manuscript accordingly. The detailed point-by-point responses are provided below.

Comment 1-1: The analysis of visible coloration would benefit from a more complete optical treatment. While CIE coordinates are derived from reflectance spectra, no optical constants or angle-dependent measurements are provided. The perceived color may change with viewing angle due to interference and substrate effects, which could represent a limitation for practical applications. In addition, it would be beneficial to include the dependence of color on material thickness, as this can also influence the observed appearance. Providing optical constants would allow calculation of the color for arbitrary angles within the

CIE framework, or at least these limitations should be discussed.

Response: We thank the reviewer for this insightful comment. We agree that a more complete optical treatment of visible coloration would ideally include wavelength-dependent optical constants, such as $n(\lambda)$ and $k(\lambda)$, and angle-dependent optical measurements. Such information would allow the reflectance spectra of the coating/substrate system to be modeled under different incident or viewing geometries and then converted into CIE chromaticity coordinates. Therefore, we agree that optical constants and angle-dependent measurements would be valuable for predicting arbitrary-angle color within the CIE framework.

In response to this point, we have revised the manuscript to more clearly define the applicable scope of the CIE analysis. Specifically, the following statement has been added after the CIE chromaticity analysis in the revised manuscript: “These CIE coordinates describe the visible appearance under the present coating/substrate configuration and measurement geometry; however, without wavelength-dependent optical constants or angle-resolved measurements, possible viewing-angle, interference, and substrate effects should be considered under different application conditions.” This revision clarifies that the reported CIE coordinates should be interpreted as colorimetric descriptors under the present measurement configuration, rather than arbitrary-angle color predictions. It also explicitly addresses the possible influence of viewing angle, interference effects, and substrate contributions, as suggested by the reviewer.

To further address the reviewer’s concern that material thickness can influence the observed appearance, we added thickness-dependent visible reflectance spectra of fixed-composition pure YMnO_3 coatings with thicknesses of 5, 10, 15, 25, and 35 μm , as shown in **Fig. R1**. The corresponding CIE chromaticity coordinates and lightness L^* values are summarized in **Table R1**. The CIE chromaticity coordinates and lightness L^* were calculated from the measured reflectance spectra in the 400-750 nm range using the CIE 1931 2° standard observer and the D65 standard illuminant. The tristimulus values were obtained by integrating the spectral reflectance with the D65 illuminant and the CIE color-matching functions, followed by conversion to x , y , and L^* according to the standard CIE LAB definitions.

The added data show that coating thickness has a clear effect on the apparent brightness and color intensity. As the coating thickness increases from 5 to 35 μm , the visible reflectance intensity increases, and L^* increases from 32.00 to 45.47, indicating enhanced lightness with increasing coating thickness. Meanwhile, the CIE coordinates remain within a relatively narrow range, with $x=0.287\text{-}0.303$ and $y=0.377\text{-}0.383$, and the overall spectral profiles are retained. These results indicate that coating thickness mainly modulates the apparent lightness and color intensity, rather than causing a substantial shift in the characteristic visible color response of YMnO_3 within the tested thickness range.

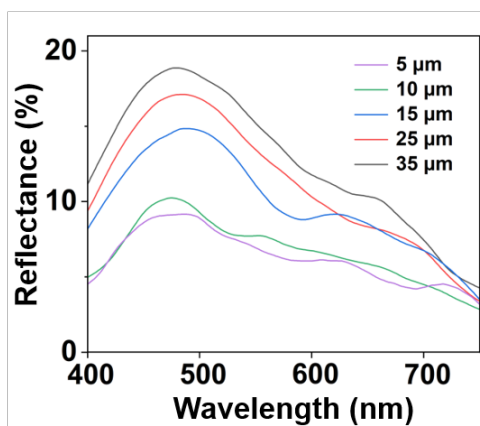


Fig. R1. Thickness-dependent visible reflectance spectra of pure YMnO₃ coatings.

Table R1. CIE chromaticity coordinates and lightness of pure YMnO₃ coatings with different thicknesses.

Thickness (μm)	CIE x	CIE y	L*
5	0.301	0.382	32.00
10	0.303	0.383	33.34
15	0.292	0.380	40.20
25	0.287	0.383	43.09
35	0.290	0.377	45.47

Comment 1-2: The comparison with prior work is generally appropriate; however, it would benefit from a broader context. The current discussion focuses mainly on pigments and oxide systems (Table S1), while other relevant material classes are not considered. In particular, ultrathin transparent conductive or plasmonic metallic films, which can also exhibit combined visible and infrared optical functionality, are not discussed. Including a comparison with recent work on ultrathin high-quality gold films (e.g., Nano Letters, DOI: 10.1021/acs.nanolett.4c04311) would help to better position the present results within the wider field.

Response: We appreciate the reviewer for this valuable suggestion. We agree that the original benchmarking was somewhat limited, as it mainly focused on blue inorganic pigments and MIR-transparent oxides.

In response, we revised Supplementary Table 1 by adding a representative ultrathin Au film from a recent Nano Letters study (Nano Lett. 2024) as a metallic-film benchmark. This addition broadens the scope of comparison beyond the original pigment- and oxide-centered framework and helps position the present work within the wider class of materials that exhibit combined visible and infrared optical functionalities. Because a directly comparable thermal limit was not explicitly reported in that study, we did not assign a speculative value and instead marked the

corresponding entry as “Not reported”.

To ensure that the revised table is applicable across different material classes, we also updated the table title and replaced the original categories “Crystal type” and “Blue origin” with the more general descriptors “Material type” and “Visible optical origin”. These revisions provide a more unified basis for comparing blue pigments, MIR-transparent oxides, and ultrathin metallic films.

Overall, the revised table offers a broader and more balanced benchmarking context for evaluating the present material system. For clarity, the revised version of Supplementary Table 1 is provided below as **Table R2** in this response letter, and the corresponding revisions in the Supplementary Information have been highlighted in red.

Table R2. Benchmark comparison of representative blue inorganic materials, MIR-transparent oxides, and selected ultrathin metallic films for dual-band camouflage applications.

Material	Material type	Visible optical origin	MIR emissivity	Max T	Ref
Prussian Blue	Coordination framework	Intervalence charge transfer	~0.1	300 °C	Nat. Sustain. 2022, 5, 339-347
CoAl ₂ O ₄	Spinel oxide	Co ²⁺ d-d transition	>0.9	1400 °C	RSC Adv. 2015, 5, 87932-87939.
Y ₂ O ₃	Cubic oxide	—	0.15	1100 °C	J. Mater. Chem. A. 2025, 13, 39330
Ultrathin Au film	Metallic film	Thickness-enabled transparency	Low emissivity	—	Nano Lett. 2024, 24, 16270-16275
2D YMnO ₃ (this work)	2D hexagonal oxide	Mn-O electronic modulation	0.13	900 °C	—

Comment 1-3: I also note that the wavelength-color mapping in Fig. 4b, c appears slightly incorrect. Given that the manuscript already employs the CIE framework for color analysis, it would be more appropriate to ensure that the color mapping is consistent with CIE standards. In addition, the manuscript would benefit from minor language corrections to improve clarity and readability.

Response: We sincerely thank the reviewer for this careful and constructive comment. We agree that, since the manuscript employs the CIE framework as the formal basis for color analysis, the wavelength-color bars originally shown beneath Fig. 4b and 4c were not sufficiently rigorous and could introduce inconsistency in the visual presentation. These color bars were initially included only as schematic guides to the visible wavelength range, rather than as strict CIE-compliant color mappings. However, we fully recognize the reviewer’s concern that their appearance may be

misleading in the context of a CIE-based discussion.

To address this issue, we have revised Fig. 4 by removing the non-standard wavelength-color bars beneath Fig. 4b and 4c in the updated version. The visible optical response is now conveyed directly by the reflectance spectra and Kubelka-Munk analysis, while the color characteristics of the samples are evaluated in a standardized manner through the CIE 1931 chromaticity diagram in Fig. 4d. In this way, the revised figure now presents the spectral data and the color analysis in a more rigorous and internally consistent form. For the reviewer's convenience, the revised Fig. 4b and Fig. 4c are provided below as **Fig. R2**.

In addition, in response to the reviewer's suggestion regarding clarity and readability, we made several minor language and typographical corrections in the relevant figure captions and associated text. Specifically, in the caption of Fig. 4, panel c was revised from "Kubelka-Munk transformed absorption derived from diffuse reflectance" to "Kubelka-Munk function (K/S) derived from the diffuse reflectance spectra", because K/S is more appropriately described as the Kubelka-Munk function rather than as a direct absorbance term. Panel d was revised from "CIE 1931 chromaticity coordinates calculated from the visible spectra with a magnified view" to "CIE 1931 chromaticity coordinates calculated from the reflectance spectra, with the corresponding enlarged view", so that the wording is more precise and fully consistent with the reflectance-based CIE analysis used in the manuscript. These revisions were made to improve both terminological accuracy and internal consistency with the color-analysis framework adopted in Fig. 4.

We also corrected several minor language and typographical issues elsewhere in the revised manuscript. In the caption of Fig. 1, "Blue coloration MIR transparency trade off" was revised to "Trade-off between blue coloration and MIR transparency" to provide a grammatically complete and clearer expression of the physical relationship discussed in the schematic. In the same caption, "enhanced of the Born effective charge" was corrected to "enhancement of the Born effective charge" to resolve the grammatical error and improve scientific readability. In the Results text, " Mn_3O_4 in XRD" was corrected to " Mn_3O_4 in XRD" by inserting the missing space. In the opening paragraph of the Fig. 5 section, the missing space in "14,29,33 The central premise" was corrected, and "dual band camouflage" was standardized as "dual-band camouflage" to ensure proper compound-adjective usage and consistency of technical expression. All changes are highlighted in red in the *Revised Manuscript*.

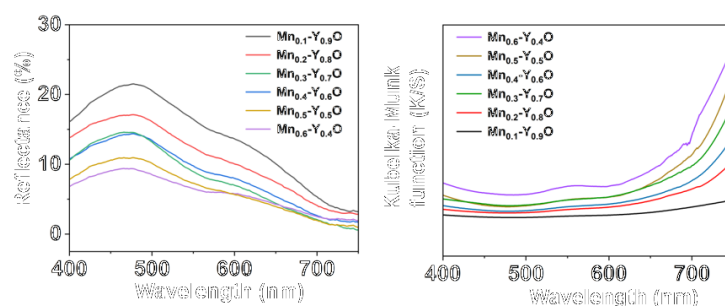


Fig. R2. Revised reflectance spectra and Kubelka-Munk function (K/S) curves.

◆ Reviewer #2:

General Comment: In the manuscript “Thermally stable 2D YMnO₃ enabling blue visible camouflage with mid-infrared transparency” Zhu et al. explore the realization of a 2D hexagonal YMnO₃ system through microwave-shock synthesis, featuring high thermal robustness, a blue visible appearance, and MIR transparency, enabling the decoupling of visible and infrared optical constraints in thermally stable oxides. The work is well presented, with a very extensive study of the material properties, and it can be of great interest for the scientific community. However, I believe that with some integration, the manuscript can be made clearer. Therefore, I recommend publication in the journal, provided that the authors make the further integrations outlined below.

Response: We are grateful for the reviewer’s positive evaluation and constructive suggestions. The manuscript has been revised to improve its clarity, completeness, and internal consistency. In particular, additional discussion and supporting analysis have been incorporated regarding the environmental stability of the YMnO₃ coating, the statistical reproducibility of films prepared under identical conditions, the optical characterization, and the quantitative interpretation of the LO-TO splitting suppression associated with visible-MIR decoupling. Detailed responses to the individual comments are provided below.

Comment 2-1: First, no discussion is provided regarding the environmental stability of the materials. Is material degradation possible under certain conditions? Could the authors clarify this point in more detail?

Response: We sincerely thank the reviewer for raising this important point. We agree that the original manuscript did not discuss the environmental stability of the YMnO₃ coating in sufficient detail. In the revised manuscript, we have clarified this issue by distinguishing the thermal robustness directly supported by the present experiments from the broader question of long-term environmental stability under complex service conditions.

The results in this work mainly demonstrate that the YMnO₃ coating retains its infrared-regulation function under the thermal conditions experimentally examined. Specifically, the 25 μm YMnO₃ film maintained a stable apparent temperature during prolonged exposure at 100 °C. The coating also preserved a low apparent radiation temperature after annealing at 900 °C for up to 8 h. In addition, short-duration flame exposure did not cause an obvious loss of the infrared-regulation function. These observations indicate that, within the tested thermal conditions, there is no obvious evidence of rapid intrinsic degradation of the YMnO₃ coating.

At the same time, we acknowledge that the apparent infrared response of the coating may be affected under more severe exposure conditions. In particular, after prolonged flame exposure, slight carbon deposition was observed on the coating surface. Such surface contamination may introduce additional extrinsic mid-infrared

loss channels and thereby alter the measured infrared response. Therefore, the response variation observed after prolonged flame exposure should be interpreted as a surface-contamination-related effect under an extreme exposure condition, rather than as direct evidence of rapid intrinsic degradation of the YMnO_3 phase itself.

To define the scope of the stability conclusion more precisely, we have expanded the discussion associated with Fig. 3 by adding the following statement: “These results indicate that the YMnO_3 coating retains its infrared-regulation function under the thermal conditions examined in this work, with no obvious evidence of rapid intrinsic degradation.” This added discussion defines the stability regime directly supported by the present thermal-response data and avoids overextending the conclusion to untested long-term environmental conditions.

To further avoid misinterpreting the prolonged-flame-exposure result as intrinsic material degradation, we have expanded the discussion associated with Fig. 5 by adding the following statement: “This observation suggests that the changes observed after prolonged flame exposure are more plausibly attributed to surface carbon deposition and the associated extrinsic MIR losses, rather than to rapid intrinsic degradation of the YMnO_3 phase itself.” This added discussion clarifies that the performance variation observed under prolonged flame exposure is more likely associated with an extrinsic surface-contamination effect, rather than intrinsic phase degradation. All changes are highlighted in red in the *Revised Manuscript*.

Comment 2-2: Do the authors have any statistics regarding the characteristics of the films synthesized under the same experimental conditions?

Response: We appreciate the reviewer’s valuable suggestion regarding the statistical reproducibility of the film characteristics under identical experimental conditions. To address this point, we carried out an independent reproducibility assessment by preparing three separate 2D YMnO_3 films using the same synthesis and coating procedures, followed by repeated morphological and broadband optical characterizations. The corresponding results are provided in **Fig. R3** and **Fig. R4** in this response.

The SEM images obtained from the three independently prepared films show comparable nanosheet-assembled surface morphology and continuous film coverage. This consistency indicates that the characteristic film morphology is not an isolated result from a single specimen, but can be reproducibly obtained under the same experimental conditions. Representative regions were selected for each film to enable a reliable comparison of the overall morphological features.

In addition, the UV-vis-NIR and MIR spectra collected from the three independently prepared films exhibit highly consistent spectral profiles. The repeated UV-vis-NIR spectra retain the characteristic visible reflection response associated with the blue appearance of the films, while the MIR spectra show consistent infrared optical behavior across the independently prepared samples. These results confirm that the key broadband optical characteristics of the 2D YMnO_3 films are reproducible under the same preparation conditions.

Taken together, the repeated SEM observations and broadband optical measurements demonstrate that both the nanosheet-based film morphology and the optical response discussed in the manuscript are reproducible across independently prepared films, rather than being derived from a single representative sample.

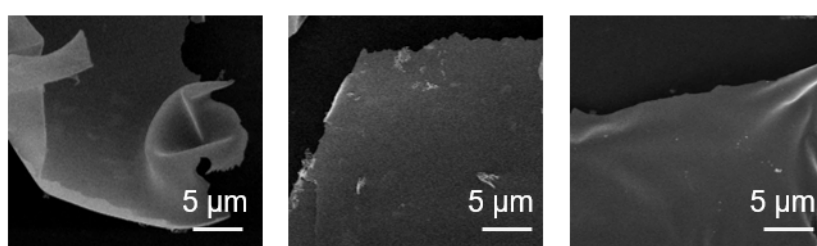


Fig. R3. Morphological reproducibility of independently prepared 2D YMnO₃ films.

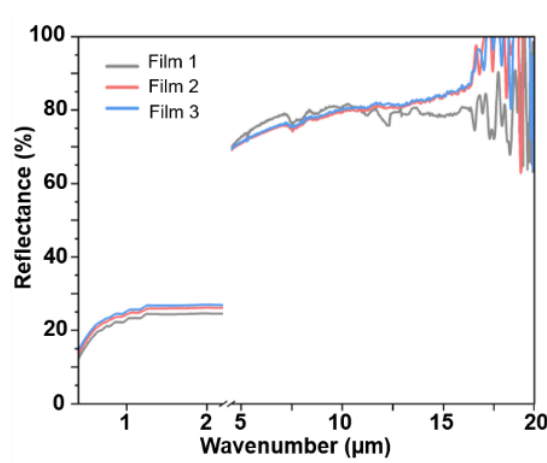


Fig. R4. Reproducibility of broadband optical response for independently prepared 2D YMnO₃ films.

Comment 2-3: All the measurements reported in this work were obtained using a reflection configuration. However, I believe that measuring the transmittance of the material would also provide valuable support for the significance of the results.

Response: We appreciate the reviewer's insightful suggestion regarding the value of transmission-geometry measurements. We agree that a direct transmission-geometry measurement would provide an intuitive assessment of whether MIR radiation is attenuated during propagation through the YMnO₃ layer. Although the optical measurements in the present work were performed in a reflection configuration, the reviewer's concern is fundamentally related to whether the YMnO₃ material itself introduces appreciable absorption loss in the MIR region. To address this issue from a material-level optical perspective, we have added the wavelength-dependent optical constants, including the refractive index n and extinction coefficient k , of the YMnO₃ layer.

The extinction coefficient k is directly relevant to MIR propagation loss because it quantifies the absorptive component of optical attenuation. Specifically, the absorption coefficient is related to k by $\alpha = \frac{4\pi k}{\lambda}$. Under a single-pass, absorption-only approximation, the absorption-induced propagation factor through a film with thickness d can be estimated as $T_{abs} \approx \exp(-\alpha d)$. This expression is not intended to replace a measured total transmittance spectrum, which can also include reflection, interference, and scattering contributions. Instead, it provides a direct way to evaluate whether the YMnO_3 layer itself causes significant MIR absorption loss.

As shown in **Fig. R5**, the extracted refractive index n remains relatively stable at approximately 2.1 within the analyzed infrared spectral range, indicating that no pronounced anomalous dispersion is observed in this region. More importantly, the extinction coefficient k decreases rapidly from the short-wavelength side and remains close to zero in the MIR region. Since k directly determines the absorption coefficient through $\alpha = \frac{4\pi k}{\lambda}$, these near-zero k values indicate that the absorption-induced propagation loss in the YMnO_3 layer is weak. Therefore, although a direct transmission measurement would be useful for evaluating the total transmission response of a specific film geometry, the added optical-constant analysis provides material-level evidence that YMnO_3 does not introduce appreciable intrinsic MIR absorption loss within the examined spectral range. This result supports the low-loss MIR character of YMnO_3 and is consistent with the reflection-based broadband optical response and infrared thermographic behavior reported in the manuscript.

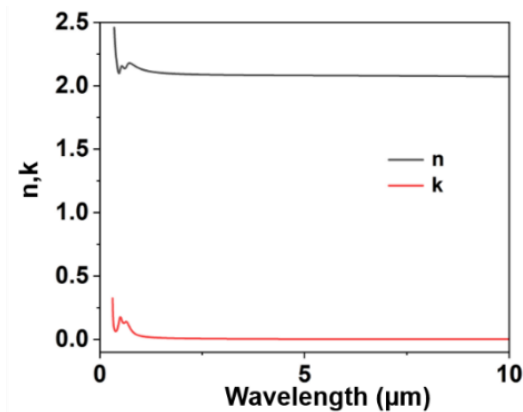


Fig. R5. Wavelength-dependent optical constants of the YMnO_3 layer extracted from optical analysis.

Comment 2-4: The mechanism behind the suppression of the LO–TO splitting is not clear; can the authors quantify this effect to support this claim?

Response: We sincerely appreciate the reviewer’s insightful comment, which points to the need to clarify both the physical origin and the quantitative magnitude of the suppressed LO-TO splitting. In the revised manuscript, we have addressed this issue

by adding polarized infrared reflectance measurements and factorized dielectric-function fitting for both bulk and 2D YMnO₃.

In polar crystals, LO-TO splitting originates from the macroscopic electric field generated by long-wavelength polar lattice vibrations. Its magnitude is therefore closely related to the long-range Coulomb contribution to the polar phonon response. When the dynamic polar response is weakened, the non-analytic long-range Coulomb contribution is reduced, leading to a smaller separation between the TO and LO phonon frequencies. For 2D YMnO₃, we interpret the suppressed LO-TO splitting as a consequence of weakened long-range polarization in the dimensionally confined structure, while the local Mn-O coordination framework is largely retained.

To address this important point, we have added polarized infrared reflectance measurements and factorized dielectric-function fitting for both bulk and 2D YMnO₃. The measured and fitted polarized infrared reflectance spectra are shown in **Fig. R6**. In the revised analysis, the TO and LO phonon frequencies were not assigned from apparent reflectance maxima or minima. Instead, they were extracted from the fitted phonon dielectric response using the factorized dielectric-function model:

$$\varepsilon(\omega) = \varepsilon_{\infty} \prod_j \frac{\omega_{\text{LO},j}^2 - \omega^2 - i\gamma_{\text{LO},j}\omega}{\omega_{\text{TO},j}^2 - \omega^2 - i\gamma_{\text{TO},j}\omega}$$

where $\omega_{\text{TO},j}$ and $\omega_{\text{LO},j}$ are the transverse and longitudinal optical phonon frequencies of the j -th infrared-active polar mode, respectively, and $\gamma_{\text{TO},j}$ and $\gamma_{\text{LO},j}$ are the corresponding damping constants.

The LO-TO splitting was then calculated as:

$$\Delta\omega_{\text{LO-TO},j} = \omega_{\text{LO},j} - \omega_{\text{TO},j}$$

The representative fitting results are summarized in **Table R3**. The extracted data show that 2D YMnO₃ exhibits reduced LO-TO splitting compared with bulk YMnO₃ for all selected infrared-active polar modes. Specifically, the LO-TO splitting decreases from 19.78 to 16.31 cm⁻¹ for mode 1, from 14.94 to 12.69 cm⁻¹ for mode 2, from 94.16 to 51.76 cm⁻¹ for mode 3, and from 368.62 to 239.86 cm⁻¹ for mode 4. These correspond to reductions of 17.54%, 15.06%, 45.03%, and 34.93%, respectively. Since LO-TO splitting originates from the macroscopic electric field associated with long-wavelength polar lattice vibrations, the consistent decrease in $\Delta\omega_{\text{LO-TO}}$ indicates that the long-range Coulomb contribution to the polar phonon response is weakened in 2D YMnO₃.

In addition to the LO-TO splitting analysis, we extracted the real and imaginary parts of the dielectric function, ε_1 and ε_2 , from the same fitting procedure, as shown in **Fig. R7**. The fitted spectra reproduce the main phonon-response features of the measured polarized infrared reflectance, with RMSE values of 4.4197% for bulk YMnO₃ and 3.1683% for 2D YMnO₃, supporting the reliability of the extracted dielectric-function parameters.

The dielectric-function spectra provide more direct evidence for the suppression of Reststrahlen-band expansion. In polar dielectrics, the spectral region where ε_1 becomes negative corresponds to a Reststrahlen-type response, where electromagnetic

propagation is strongly suppressed and MIR phonon-related optical loss is enhanced. Bulk YMnO₃ exhibits a broad negative- ϵ_1 interval from 509.94 to 914.12 cm⁻¹, with a width of 404.18 cm⁻¹. In contrast, the negative- ϵ_1 interval of 2D YMnO₃ is compressed to 586.64-778.02 cm⁻¹, with a width of 191.38 cm⁻¹. This corresponds to a narrowing of 212.80 cm⁻¹, or approximately 52.65%. The upward shift of the low-frequency boundary and the downward shift of the high-frequency boundary indicate that the Reststrahlen-type polar response is compressed rather than expanded in 2D YMnO₃. This result directly supports reduced MIR phonon-polariton loss and suppressed Reststrahlen-band expansion.

We further estimated the mean effective charges from the infrared phonon oscillator strengths extracted from the dielectric-function fitting. As summarized in **Table R4**, the estimated mean cationic effective charge decreases from 4.92 in bulk YMnO₃ to 4.55 in 2D YMnO₃, corresponding to a reduction of approximately 7.52%. The magnitude of the oxygen effective charge also decreases from 3.28 to 3.03, corresponding to a reduction of approximately 7.62%. Although this optical estimate is not intended to replace a full tensorial Born-effective-charge calculation, the consistent decrease in the oscillator-strength-derived effective charges further supports a weakened dynamic polar response in 2D YMnO₃.

Taken together, these added measurements and analyses provide quantitative support for the suppressed LO-TO splitting and clarify its connection with the weakened long-range polar response in 2D YMnO₃. The corresponding discussion, reflectance-fitting results, extracted phonon parameters, and effective-charge estimation have been incorporated into the *Revised Manuscript* and highlighted in red.

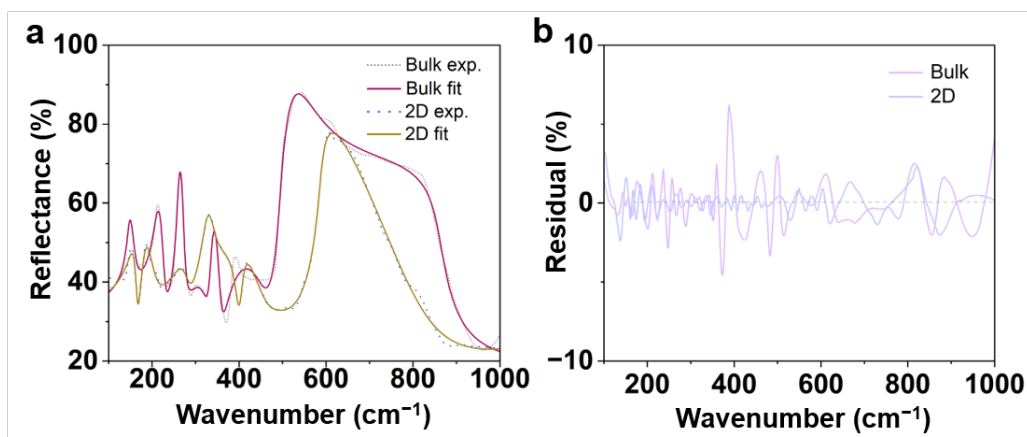


Fig. R6. Polarized infrared reflectance fitting and residual analysis of bulk and 2D YMnO₃. a. Experimental and FPSQ-fitted polarized infrared reflectance spectra of bulk and 2D YMnO₃. b. Corresponding fitting residuals, defined as $\Delta R = R_{\text{exp}} - R_{\text{fit}}$.

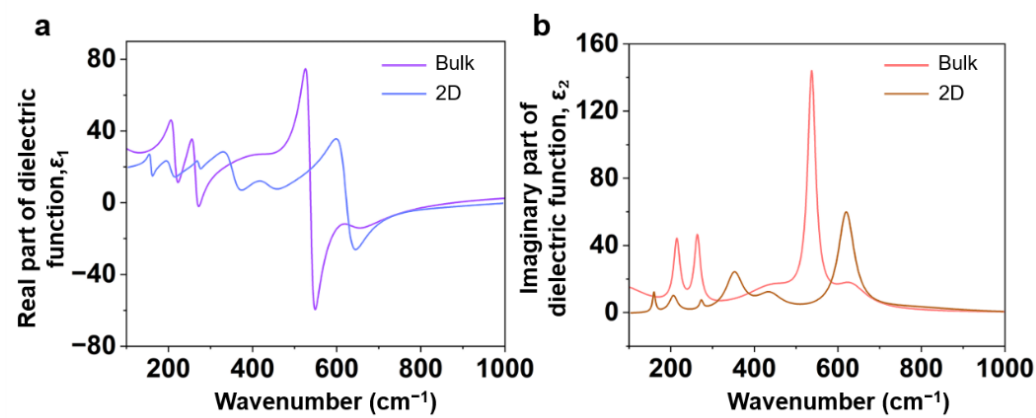


Fig. R7. Extracted dielectric functions of bulk and 2D YMnO₃. a, Real part, ϵ_1 ; narrowed and weakened negative- ϵ_1 Reststrahlen response in 2D YMnO₃. b, Imaginary part, ϵ_2 ; suppressed polar-phonon dielectric loss in 2D YMnO₃.

Table R3. Representative LO-TO splitting values extracted from effective FPSQ fitting of polarized infrared reflectance spectra.

Mode	Sample	$\omega_{TO} / \text{cm}^{-1}$	$\omega_{LO} / \text{cm}^{-1}$	$\Delta\omega_{LO-TO} / \text{cm}^{-1}$	Reduction of $\Delta\omega_{LO-TO}$ relative to bulk
1	Bulk	208.01	227.79	19.78	—
1	2D	184.90	201.21	16.31	17.54%
2	Bulk	258.25	273.19	14.94	—
2	2D	272.65	285.33	12.69	15.06%
3	Bulk	369.00	463.15	94.16	—
3	2D	407.65	459.41	51.76	45.03%
4	Bulk	493.92	862.53	368.62	—
4	2D	587.73	827.59	239.86	34.93%

Table R4. Mean dynamic effective charges estimated from infrared phonon oscillator strengths for bulk and 2D YMnO₃.

Sample	Mean $Z_{Y/Mn}^*$ estimated from IR oscillator strength	Mean Z_O^* estimated from charge neutrality
Bulk	4.92	-3.28
2D	4.55	-3.03

Comment 2-5: Minor Remarks: The figures are generally a bit confusing. It might be helpful to correct a few typos (e.g., “spectrally stable, spectrally stable” or “Thata” in Fig. 2) and to improve the overall writing style. Additionally, adding more details about the synthesis process could strengthen the manuscript.

Response: We appreciate the reviewer’s careful reading and constructive suggestions. We have carefully checked the figures, in-figure labels, and corresponding descriptions, and revised the relevant parts to improve the clarity, readability, and reproducibility of the manuscript. The revisions were made at three levels, as detailed below.

At the proofreading level, the typographical errors pointed out by the reviewer have been corrected. The duplicated expression “spectrally stable, spectrally stable” was

revised to “spectrally stable, low-emissivity”, and the typo “Thata” in Fig. 2 was corrected to the proper theta notation. The corresponding results are provided below as **Fig. R8** for clarity. These corrections remove unintended wording and labeling errors that may have affected the readability of the figure-related descriptions.

At the textual-clarity level, the manuscript has been polished to improve grammar, sentence flow, abbreviation formatting, chemical notation, and terminology consistency. For example, the original sentence: “Collectively, these results indicate that the rapid thermal history associated with microwave processing enables fast crystallization of the hexagonal phase YMnO_3 with within XRD detection limit.” was revised to: “Collectively, these results indicate that the rapid thermal history associated with microwave processing promotes the crystallization of hexagonal YMnO_3 , with no obvious crystalline secondary phase detectable by XRD.” This revision corrects the grammatical error in the original sentence and makes the XRD-based statement more precise by specifying that no obvious crystalline secondary phase is detectable by XRD. As another example, the original sentence: “For more stringent hot-side conditions, establish a thermal-endurance boundary via annealing at 900 °C and evaluate the retention of the low-radiance appearance after thermal history by benchmarking 2D YMnO_3 against Prussian blue (PB).” was revised to: “For more stringent hot-side conditions, a thermal-endurance boundary was established by annealing at 900 °C, and the retention of the low-radiance appearance after thermal treatment was evaluated by benchmarking 2D YMnO_3 against Prussian blue (PB).” This revision changes the original instruction-like wording into a formal Results-style description with a clear grammatical subject. The abbreviation “FTIR” was also introduced in the standard form as “Fourier-transform infrared spectroscopy (FTIR)” to improve formatting consistency.

At the methodological-description level, the Methods section has been expanded in response to the reviewer’s suggestion to add more details about the synthesis process. The original synthesis description was brief and mainly stated the precursor amounts, grinding time, and microwave treatment condition. In the revised manuscript, this part now provides a more complete stepwise description, including the precursor composition and amounts, grinding procedure, formation of a homogeneous precursor mixture, transfer into a clean and dry crucible, microwave irradiation at 650 W for 10 min in ambient air, representative temperature monitoring using a Raytek M13 infrared detector, the maximum temperature of approximately 850 °C under the present reaction conditions, natural cooling in air, product collection, and gentle grinding before subsequent structural, spectroscopic, and coating fabrication experiments.

Overall, these revisions correct typographical and formatting issues, improve the clarity of the relevant descriptions, and provide a more reproducible description of the microwave-shock synthesis route. All changes are highlighted in red in the *Revised Manuscript*.

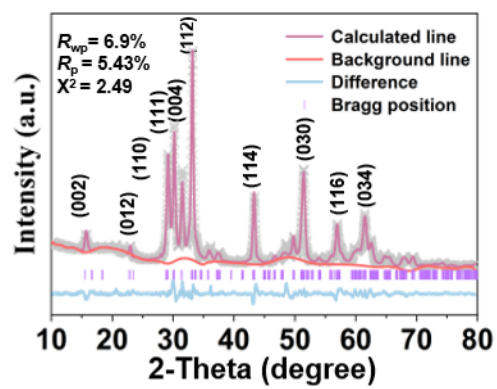


Fig. R8. Rietveld-refined XRD pattern of YMnO_3 .

◆ Reviewer #3:

General Comment: This manuscript presents a hexagonal 2D YMnO₃ system and argues that, through microwave-shock-enabled non-equilibrium synthesis, the material can simultaneously exhibit a blue visible appearance, mid-infrared transparency, and high-temperature robustness. The main point of the work is that dimensional confinement and retained rigid Mn-O coordination provide a structure-based route to mitigate the long-standing conflict between visible coloration and MIR optical constraints, and most of the manuscript is organized around structural validation and cross-spectral optical/thermal characterization in support of this claim. The topic is interesting and potentially important for high-temperature multispectral camouflage. However, before the work can be considered for publication, several key points should be clarified.

Response: We are grateful to the reviewer for the careful assessment of our manuscript and for recognizing the scientific value and potential significance of the hexagonal 2D YMnO₃ system for high-temperature multispectral camouflage. We also agree that the mechanistic interpretation and the optical and thermal-radiative performance discussion should be presented with greater precision and clearer experimental grounding. In the revised manuscript, we have carefully reconsidered the wording and scope of the main claims and made corresponding revisions to improve the clarity, rigor, and balance of the discussion. The specific comments raised by the reviewer are addressed point by point below.

Comment 3-1: The XAFS, FTIR, and Raman results support that the local Mn–O coordination is retained and are consistent with the authors’ short-range bonding picture. However, these data alone do not seem sufficient to fully support the broader claim that the conflict between visible coloration and MIR optical constraints is resolved, since the long-range polarization contribution and the LO-TO splitting, which are central to the authors’ explanation, are not directly shown here.

Response: We are grateful for the reviewer’s careful assessment, which correctly distinguishes the local-structure evidence from the broader visible-MIR decoupling mechanism. We agree that the XAFS, FTIR, and Raman results mainly support the retention of local Mn-O coordination and the short-range bonding framework in 2D YMnO₃. These measurements alone do not directly quantify the macroscopic long-range polar response or the LO-TO splitting. Therefore, they are not sufficient by themselves to fully establish the suppression of Reststrahlen-band expansion or the resolution of the visible-coloration/MIR-optical constraint.

To address this important point, we have added polarized infrared reflectance measurements and factorized dielectric-function fitting for both bulk and 2D YMnO₃. The measured and fitted polarized infrared reflectance spectra are shown in **Fig. R9**. In the revised analysis, the TO and LO phonon frequencies were not assigned from

apparent reflectance maxima or minima. Instead, they were extracted from the fitted phonon dielectric response using the factorized dielectric-function model:

$$\varepsilon(\omega) = \varepsilon_{\infty} \prod_j \frac{\omega_{\text{LO},j}^2 - \omega^2 - i\gamma_{\text{LO},j}\omega}{\omega_{\text{TO},j}^2 - \omega^2 - i\gamma_{\text{TO},j}\omega}$$

where $\omega_{\text{TO},j}$ and $\omega_{\text{LO},j}$ are the transverse and longitudinal optical phonon frequencies of the j -th infrared-active polar mode, respectively, and $\gamma_{\text{TO},j}$ and $\gamma_{\text{LO},j}$ are the corresponding damping constants.

The LO-TO splitting was then calculated as:

$$\Delta\omega_{\text{LO-TO},j} = \omega_{\text{LO},j} - \omega_{\text{TO},j}$$

The representative fitting results are summarized in **Table R5**. The extracted data show that 2D YMnO₃ exhibits reduced LO-TO splitting compared with bulk YMnO₃ for all selected infrared-active polar modes. Specifically, the LO-TO splitting decreases from 19.78 to 16.31 cm⁻¹ for mode 1, from 14.94 to 12.69 cm⁻¹ for mode 2, from 94.16 to 51.76 cm⁻¹ for mode 3, and from 368.62 to 239.86 cm⁻¹ for mode 4. These correspond to reductions of 17.54%, 15.06%, 45.03%, and 34.93%, respectively. Since LO-TO splitting originates from the macroscopic electric field associated with long-wavelength polar lattice vibrations, the consistent decrease in $\Delta\omega_{\text{LO-TO}}$ indicates that the long-range Coulomb contribution to the polar phonon response is weakened in 2D YMnO₃.

In addition to the LO-TO splitting analysis, we extracted the real and imaginary parts of the dielectric function, ε_1 and ε_2 , from the same fitting procedure, as shown in **Fig. R10**. The fitted spectra reproduce the main phonon-response features of the measured polarized infrared reflectance, with RMSE values of 4.4197% for bulk YMnO₃ and 3.1683% for 2D YMnO₃, supporting the reliability of the extracted dielectric-function parameters.

The dielectric-function spectra provide more direct evidence for the suppression of Reststrahlen-band expansion. In polar dielectrics, the spectral region where ε_1 becomes negative corresponds to a Reststrahlen-type response, where electromagnetic propagation is strongly suppressed and MIR phonon-related optical loss is enhanced. Bulk YMnO₃ exhibits a broad negative- ε_1 interval from 509.94 to 914.12 cm⁻¹, with a width of 404.18 cm⁻¹. In contrast, the negative- ε_1 interval of 2D YMnO₃ is compressed to 586.64-778.02 cm⁻¹, with a width of 191.38 cm⁻¹. This corresponds to a narrowing of 212.80 cm⁻¹, or approximately 52.65%. The upward shift of the low-frequency boundary and the downward shift of the high-frequency boundary indicate that the Reststrahlen-type polar response is compressed rather than expanded in 2D YMnO₃. This result directly supports reduced MIR phonon-polariton loss and suppressed Reststrahlen-band expansion.

We further estimated the mean effective charges from the infrared phonon oscillator strengths extracted from the dielectric-function fitting. As summarized in **Table R6**, the estimated mean cationic effective charge decreases from 4.92 in bulk YMnO₃ to 4.55 in 2D YMnO₃, corresponding to a reduction of approximately 7.52%. The magnitude of the oxygen effective charge also decreases from 3.28 to 3.03, corresponding to a reduction of approximately 7.62%. Although this optical estimate

is not intended to replace a full tensorial Born-effective-charge calculation, the consistent decrease in the oscillator-strength-derived effective charges further supports a weakened dynamic polar response in 2D YMnO₃.

Taken together, these analyses provide a clearer and more complete evidence chain. XAFS, FTIR, and Raman support the retention of local Mn-O coordination and short-range structural rigidity, which is important for preserving the visible optical response. In contrast, polarized infrared reflectance, factorized dielectric-function fitting, LO-TO splitting analysis, ϵ_1/ϵ_2 spectra, negative- ϵ_1 Reststrahlen-region analysis, and oscillator-strength-derived effective-charge estimates provide direct evidence for the weakened long-range polar response in 2D YMnO₃. Therefore, the revised manuscript no longer relies solely on local-structure characterization to support the visible–MIR decoupling mechanism. Instead, it shows that the conflict between visible coloration and MIR optical constraints is mitigated through the combined effect of retained short-range Mn-O bonding and suppressed long-range polarization-induced Reststrahlen-band expansion. The relevant fitting curves, dielectric-function spectra, and quantitative analyses are provided below for clarity.

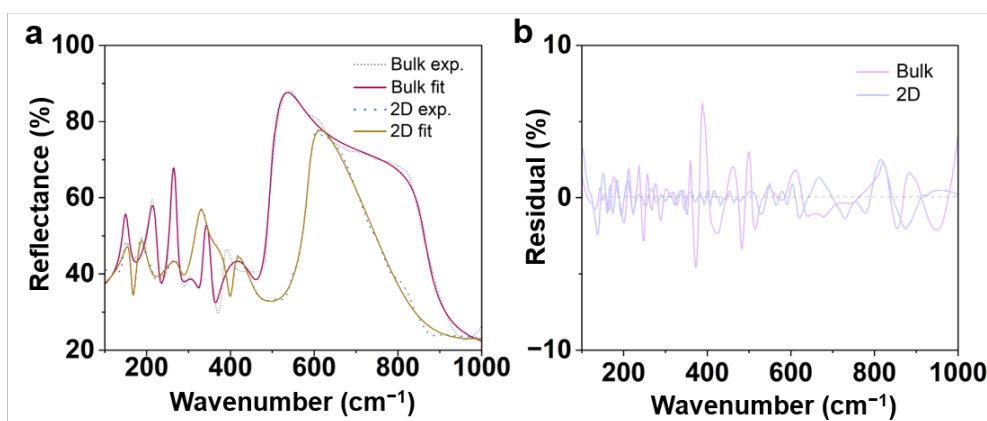


Fig. R9. Polarized infrared reflectance fitting and residual analysis of bulk and 2D YMnO₃. a. Experimental and FPSQ-fitted polarized infrared reflectance spectra of bulk and 2D YMnO₃. b. Corresponding fitting residuals, defined as $\Delta R = R_{\text{exp}} - R_{\text{fit}}$.

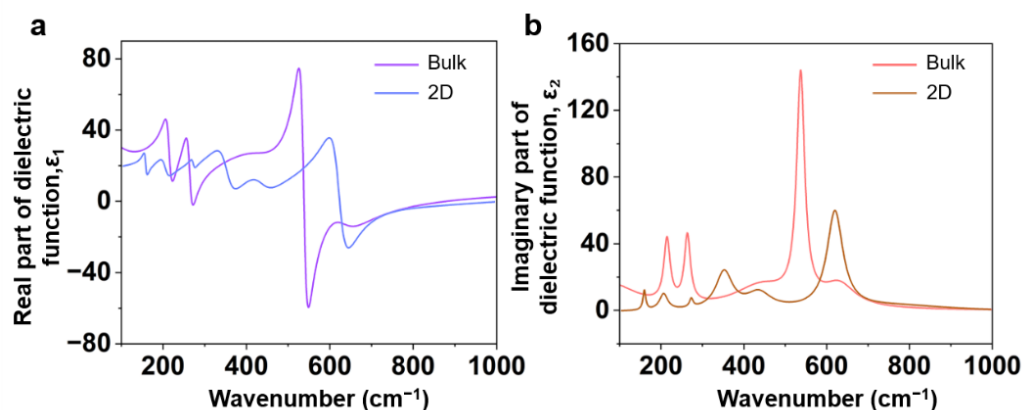


Fig. R10. Extracted dielectric functions of bulk and 2D YMnO₃. a, Real part, ϵ_1 ;

narrowed and weakened negative- ε_1 Reststrahlen response in 2D YMnO₃. b, Imaginary part, ε_2 ; suppressed polar-phonon dielectric loss in 2D YMnO₃.

Table R5. Representative LO-TO splitting values extracted from effective FPSQ fitting of polarized infrared reflectance spectra.

Mode	Sample	$\omega_{\text{TO}} / \text{cm}^{-1}$	$\omega_{\text{LO}} / \text{cm}^{-1}$	$\Delta\omega_{\text{LO-TO}} / \text{cm}^{-1}$	Reduction of $\Delta\omega_{\text{LO-TO}}$ relative to bulk
1	Bulk	208.01	227.79	19.78	–
1	2D	184.90	201.21	16.31	17.54%
2	Bulk	258.25	273.19	14.94	–
2	2D	272.65	285.33	12.69	15.06%
3	Bulk	369.00	463.15	94.16	–
3	2D	407.65	459.41	51.76	45.03%
4	Bulk	493.92	862.53	368.62	–
4	2D	587.73	827.59	239.86	34.93%

Table R6. Mean dynamic effective charges estimated from infrared phonon oscillator strengths for bulk and 2D YMnO₃.

Sample	Mean $Z_{\text{Y/Mn}}^*$ estimated from IR oscillator strength	Mean Z_{O}^* estimated from charge neutrality
Bulk	4.92	-3.28
2D	4.55	-3.03

Comment 3-2: One important point that should be clarified is whether the manuscript demonstrates MIR transparency of 2D YMnO₃ itself, or rather that the coating does not significantly disturb the low-emissivity radiative boundary of the underlying metallic substrate. As presented, much of the optical and thermographic evidence seems to support the latter interpretation, since the results are mainly discussed in terms of the coating/substrate composite and the metallic background. The authors should therefore clarify whether “MIR transparency” is being used as an intrinsic material property or as an effective behavior in the present coating geometry.

Response: We thank the reviewer for raising this important and conceptually precise point. We agree that the relationship between the MIR transparency of 2D YMnO₃ itself and the effective radiative behavior of the coating/substrate configuration should be clarified more explicitly.

In this work, “MIR transparency” is used to describe the low-loss MIR response of the YMnO₃ layer, which allows the underlying low-emissivity metallic boundary to remain radiatively effective in the coating/substrate geometry, rather than an apparent optical effect generated by the metallic substrate itself. This layer-level low-loss response is associated with the 2D-structure-enabled suppression of long-range polarization contributions and the retention of rigid local Mn-O bonding, which together reduce phonon-mediated MIR attenuation and suppress the expansion of the Reststrahlen response. Accordingly, preservation of the low-emissivity metallic boundary is not the origin of the MIR transparency, but rather the device-level consequence of the low-loss MIR character of the YMnO₃ layer in the practical

coating/substrate configuration.

To further distinguish the layer-level MIR response from the effective radiative behavior of the coating/substrate configuration, we have added optical-constant analysis of the 2D YMnO₃ layer. As shown in **Fig. R11**, the refractive index remains nearly stable at approximately 2.1, whereas the extinction coefficient decreases rapidly and remains very low over the measured near-infrared to mid-infrared range up to 10 μm . Because k is directly associated with absorptive optical loss, the near-zero extinction coefficient provides direct evidence that the 2D YMnO₃ layer itself is weakly absorbing in this spectral region. This result complements the MIR reflectance and thermographic measurements by showing that the coating does not merely inherit the radiative behavior of the metallic substrate, but instead possesses a low-loss optical character that allows the underlying low-emissivity boundary to remain radiatively effective. Accordingly, the coating/substrate configuration demonstrates the device-level manifestation of the low-loss MIR response of the 2D YMnO₃ layer.

This interpretation is supported by several sets of results in the manuscript. At the structural level, FTIR, Raman, XPS, EPR, and XAFS analyses show that the 2D YMnO₃ nanosheets retain the oxide coordination framework, preserved Mn-O local coordination, and limited defect-related loss channels, providing the material basis for a low-loss MIR response. At the optical and radiative level, the 2D YMnO₃/SLS sample shows a solar-band reflectance of 21% and a MIR emissivity of 13%, indicating suppressed MIR radiative coupling. The YMnO₃-coated PE film exhibits an apparent temperature essentially identical to that of pristine PE, further confirming that the coating itself does not introduce resolvable additional MIR radiative losses. In the thickness-dependent measurement, increasing the YMnO₃ film thickness from 7 μm to 35 μm only increases the brightness temperature from 52.2 $^{\circ}\text{C}$ to 56.7 $^{\circ}\text{C}$, which remains far below the actual hot-plate temperature of approximately 93 $^{\circ}\text{C}$. This weak thickness dependence indicates that increasing the amount of YMnO₃ does not lead to strong MIR absorption within the examined thickness range. These data are consistent with the manuscript's existing description that the coating/substrate configuration is used to evaluate the effective radiative behavior while the coating itself remains low-loss in the MIR.

The composition-dependent results further support that the MIR window is governed by the Y-Mn-O phase/structure rather than being simply imposed by the metallic substrate. For $x_{\text{Mn}} = 0.1\text{-}0.5$, the coatings retain a stable MIR high-reflectance plateau with an average reflectance of approximately 0.89 and an apparent radiation temperature of approximately 50 $^{\circ}\text{C}$ on a 100 $^{\circ}\text{C}$ hot plate. In contrast, when x_{Mn} reaches 0.6, the emergence of Mn₃O₄ is accompanied by a decrease in the average MIR reflectance to 0.568 and an increase in apparent temperature to 55.9 $^{\circ}\text{C}$. This phase-dependent degradation of the MIR window supports that the MIR-transparent/low-loss behavior originates from the material structure itself, rather than from the metallic substrate alone.

Following the reviewer's suggestion, we have added a clarifying statement in the discussion of Fig. 5 to make this relationship explicit: "This low-loss MIR response arises from the 2D YMnO₃ layer itself, whereas its device-level consequence in the

coating/substrate geometry is the preservation of the low-emissivity radiative boundary of the underlying metal.” This revision clarifies that the coating/substrate measurements are not used to define MIR transparency as a substrate-induced effect, but to validate that the MIR-transparent 2D YMnO₃ coating can preserve the underlying metallic radiative boundary under practical infrared-camouflage conditions. The relevant explanations have been incorporated into the *Revised Manuscript*.

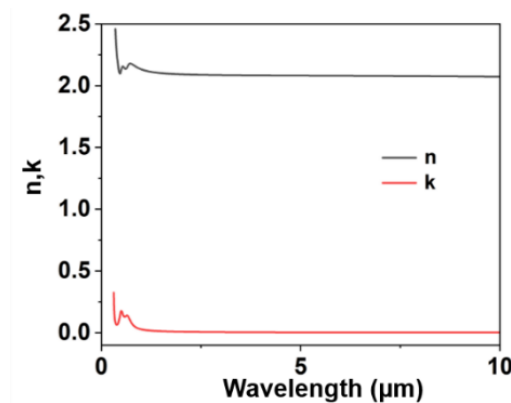


Fig. R11. Wavelength-dependent optical constants of the YMnO₃ layer extracted from optical analysis.

Comment 3-3: The manuscript makes repeated use of apparent temperature from infrared thermography to support the optical performance. Since much of the performance argument relies on this thermographic contrast, it would strengthen this part if the authors more explicitly relate the observed temperature differences to the measured MIR reflectance/emittance. In particular, it would be helpful to clarify how the effective emissivity of the coating/substrate system is expected to translate into the reported apparent-temperature differences under the present measurement conditions.

Response: We appreciate the reviewer’s insightful and technically important comment. We agree that a clearer radiometric interpretation is necessary to explicitly relate the apparent-temperature contrast observed by infrared thermography to the measured MIR reflectance/effective emittance of the YMnO₃ coating/substrate system. In infrared thermography, the apparent temperature is not a direct measurement of the thermodynamic surface temperature. Instead, it is a blackbody-equivalent temperature converted from the band-integrated infrared radiance received by the camera. According to the standard radiometric model of infrared thermography, the radiation received by the camera includes contributions from object emission, reflected background radiation, and atmospheric radiation. Under the present short-distance measurement conditions, with identical camera settings, viewing geometry, and environmental conditions for the compared regions, the atmospheric contribution and camera-related parameters can be treated as common factors. Therefore, the emissivity-dependent part of the band-integrated radiance received by the camera can be approximately described as:

$$L_{camera} \approx \varepsilon_{eff} L_{bb}(T_s) + (1 - \varepsilon_{eff}) L_{bb}(T_{bg})$$

where L_{camera} is the infrared radiance received by the camera, ε_{eff} is the effective emissivity of the coating/substrate system, $L_{bb}(T_s)$ is the blackbody radiance corresponding to the actual surface temperature T_s and $L_{bb}(T_{bg})$ represents the reflected-background radiance expressed using an equivalent background temperature. The symbol “ $\approx$ ” is used because this expression is a simplified form of the full radiometric model, in which atmospheric transmission, atmospheric self-emission, detector response, and other common measurement factors are not explicitly expanded.

This relationship explains how the effective emissivity is converted into the reported apparent-temperature difference. A lower ε_{eff} , which is consistent with the measured high MIR reflectance and the corresponding low effective emittance of the $YMnO_3$ coating/substrate system, reduces the MIR radiance emitted from the heated surface toward the infrared camera. The camera then converts this lower detected radiance into a lower blackbody-equivalent apparent temperature. Therefore, the apparent-temperature contrast does not arise from a direct measurement of a lower thermodynamic surface temperature, but from the lower band-integrated MIR radiance associated with the low effective emissivity of the coating/substrate system. This radiometric interpretation directly links the spectral and thermographic results in this work. The measured high MIR reflectance and low effective emittance indicate suppressed MIR radiative coupling of the $YMnO_3$ coating/substrate system. Consequently, under the same hot-plate conditions, the coated region provides lower MIR radiance to the infrared camera than the higher-emissivity reference region, resulting in a lower apparent temperature in the infrared images. As the hot-plate temperature increases, $L_{bb}(T_s)$ increases nonlinearly; therefore, the radiance contrast between the uncoated and low-emissivity coated regions becomes larger and is converted by the camera into an enlarged apparent-temperature difference. This explains why the reported ΔT increases at higher hot-plate temperatures in Fig. 5e.

To make this point clear in the revised manuscript, we revised the discussion of Fig. 5e. The original sentence: “This trend is consistent with the sensitivity of radiometric thermometry to effective emissivity: as temperature increases, radiance grows more strongly, and the constraining role of a low-emissivity boundary condition on the observed signal becomes increasingly prominent” has been revised to: “According to the standard radiometric principle of infrared thermography, the apparent temperature is obtained by converting the band-integrated infrared radiance received by the camera into a blackbody-equivalent temperature. Under identical measurement conditions, the high MIR reflectance and low effective emittance of the $YMnO_3$ coating/substrate system reduce the detected MIR radiance, thereby producing a lower apparent temperature and an enlarged ΔT at higher hot-plate temperature.”

This revision clarifies the conversion pathway from effective emissivity to apparent-temperature difference: the effective emissivity first determines the MIR radiance received by the infrared camera, and this detected radiance is then converted into the reported blackbody-equivalent apparent temperature. Thus, the thermographic contrast is interpreted as a radiance-based consequence of the low effective emissivity

of the YMnO_3 coating/substrate system, rather than as direct evidence of a reduced thermodynamic surface temperature. A reference on the standard radiometric principle of infrared thermography has also been added to support this clarification. (DOI: **10.3390/s140712305**.) The corresponding revisions have been incorporated into the *Revised Manuscript* and are highlighted in red for clarity.

◆ Reviewer #4:

General Comment: The manuscript presents an interesting and potentially impactful approach to achieving blue coloration with MIR transparency in thermally stable oxides. The experimental work is comprehensive and relevant for high-temperature camouflage applications. However, the central mechanism is largely supported by indirect evidence, with insufficient validation of phonon-related claims. Additional quantitative analysis and stronger justification are needed. The manuscript presents interesting results and is generally well-structured. After addressing the highlighted concerns and improving the clarity of the mechanistic explanations, the work will be suitable for publication in *Nature Communications* following revision.

Response: We thank the reviewer for the rigorous and constructive assessment of our manuscript. The comments identify an important limitation of the original version: the mechanistic interpretation of visible-coloration/MIR-response decoupling, particularly the phonon-related discussion, required clearer evidential boundaries and stronger quantitative support. In the revised manuscript, we have therefore refined the scope and wording of the central claims, strengthened the quantitative comparisons and supporting analyses, and more explicitly distinguished direct experimental observations from physically motivated interpretations. We have also incorporated the suggested recent literature to better contextualize the present work. The corresponding revisions have been incorporated into the *Revised Manuscript* and are highlighted in red for clarity. Detailed point-by-point responses are provided below.

Comment 4-1: The manuscript claims decoupling of visible coloration and MIR response-how does this fundamentally differ from prior phonon-engineering approaches in polar oxides?

Response: We thank the reviewer for raising this important conceptual question. The present strategy is indeed related to phonon engineering, but its design objective and mechanistic scope differ from conventional phonon-engineering approaches used for infrared optical control in polar oxides.

In the context of infrared optical regulation, prior phonon-engineering strategies have mainly focused on modifying the MIR phonon response itself, such as shifting phonon resonances, tuning Reststrahlen responses, adjusting infrared dielectric functions, or engineering phonon-polariton behavior. These approaches primarily optimize how a polar material interacts with infrared radiation, as reflected in representative studies on infrared dielectric-function control, Reststrahlen responses in thin polar media, dimensional effects on optical-phonon splitting, and phonon-polariton behavior [Refs. 8-10, 13]. However, they usually do not take the coupling between visible coloration and MIR polar-phonon response as the central design target.

In contrast, the present manuscript addresses a cross-spectral coupling problem.

Within the design framework discussed in this work, the electronic processes responsible for selective visible reflectance can be accompanied by local charge redistribution and enhanced lattice polarization. Such polarization enhancement may increase the long-range Coulomb contribution to LO-TO splitting and broaden the Reststrahlen band, thereby degrading MIR transparency. Therefore, the purpose of the present work is not simply to tune an isolated MIR phonon feature, but to weaken the coupling between the visible electronic-coloration process and the MIR polar-phonon response.

This distinction is embodied in the two-part structural design of 2D YMnO_3 . Dimensional confinement is proposed to reduce the long-range polarization contribution to LO-TO splitting, consistent with the dimensional dependence of optical-phonon splitting in polar 2D systems reported by Sohler et al. [Ref. 10]. Meanwhile, the retained Mn-O coordination integrity supports the short-range bonding framework and is consistent with the TO-mode-related lattice response discussed in the manuscript. The key difference from conventional MIR-centered phonon engineering is therefore the cooperative regulation of long-range polarization and short-range bonding to weaken the coupling between visible coloration and MIR response, rather than the optimization of MIR phonon behavior alone.

Thus, the decoupling claimed in this work does not mean that phonon effects are absent. Rather, it means that the phonon-related MIR response is structurally regulated so that visible coloration does not necessarily lead to pronounced degradation of the MIR transparency window.

Comment 4-2: Can the authors quantitatively compare their strategy with existing blue pigments or MIR-transparent coatings to clearly establish novelty?

Response: We appreciate the reviewer's request for a clearer quantitative benchmark to substantiate the novelty of the present strategy. We fully agree that such comparison is essential because blue materials capable of simultaneously exhibiting low-loss mid-infrared (MIR) behavior and high thermal stability remain extremely rare. To the best of current knowledge, previously reported blue MIR low-emissivity or MIR-transparent systems are largely limited to Prussian-Blue-based architectures, whose optical functionality generally relies on multilayer coating configurations and organic/inorganic hybrid designs rather than intrinsically thermally robust oxide pigments. In contrast, conventional thermally stable blue ceramic pigments, such as CoAl_2O_4 spinels, provide visible coloration but do not exhibit low-MIR-emissivity or MIR-transparent characteristics. Therefore, the simultaneous integration of blue appearance, low-loss MIR response, and high-temperature stability remains largely unresolved in existing material systems.

In response, we have revised Table S1 in the Supporting Information to provide a systematic comparison between the present 2D YMnO_3 coating and representative blue pigments or MIR-transparent/low-emissivity coating systems. The revised table now includes material type, visible optical origin, visible appearance, reported or effective MIR emissivity/optical metric, sample configuration, and reported maximum

thermal-stability temperature. The sample configuration is explicitly listed because the MIR response of low-emissivity or MIR-transparent coatings is strongly dependent on the coating/substrate geometry. For clarity and ease of evaluation, the revised benchmark table is also reproduced in this response as **Table R7**.

The comparison shows that existing blue pigment systems and MIR-control coating systems satisfy different subsets of the required functions. Prussian-Blue-containing low-emissivity films can provide blue appearance and an effective MIR emissivity of approximately 0.1 in an Al-backed bilayer coating configuration; however, the low-emissivity behavior is achieved through the coating architecture rather than through a thermally robust oxide pigment. CoAl_2O_4 is a representative thermally stable blue spinel pigment with coloration arising from Co^{2+} -related electronic transitions, but it does not provide a low-MIR-emissivity or MIR-transparent coating function. Therefore, conventional blue pigment systems do not simultaneously combine blue appearance, low-loss MIR behavior, and high-temperature stability.

Conversely, reported MIR-transparent or low-emissivity systems also show incomplete functionality with respect to the present design target. For example, 2D Y_2O_3 coatings provide a useful MIR-transparent/low-loss reference, but the material is white rather than blue. Compared with these representative systems, the 2D YMnO_3 coating developed in this work combines blue visible appearance, a low effective MIR emissivity of approximately 0.13 in the coating-on-SLS configuration, and thermal stability up to 900 °C. This benchmark clarifies that the novelty of the present strategy lies in integrating blue coloration with low-loss MIR behavior and thermal robustness through a 2D hexagonal oxide design, rather than optimizing any single property in isolation. The corresponding revisions have been incorporated into Table S1 of the revised Supporting Information, providing a direct quantitative benchmark to support the proposed cross-spectral decoupling strategy.

Table R7. Benchmark comparison of representative blue inorganic materials, MIR-transparent oxides for dual-band camouflage applications.

Material	Material type	Visible optical origin	Visible appearance	MIR metric / emissivity	Configuration	Max. T	Ref.
Prussian Blue	Coordination framework	Intervalence charge transfer	Blue	$\epsilon \approx 0.1$	PB/PE/Al bilayer low-emissivity film	300 °C	Nat. Sustain. 2022, 5, 339-347
CoAl_2O_4	Spinel oxide	Co^{2+} d-d transition	Blue	$\epsilon > 0.9$	Reported ceramic pigment	1400 °C	RSC Adv. 2015, 5, 87932-87939
Y_2O_3	Cubic oxide	—	White	$\epsilon \approx 0.15$	2D Y_2O_3 film/coating	1100 °C	J. Mater. Chem. A 2025, 13, 39330
2D YMnO_3 (this work)	2D hexagonal oxide	Mn-O electronic modulation	Blue	$\epsilon \approx 0.13$	2D YMnO_3 Coating on SLS	900 °C	This work

Comment 4-3: Is the suppression of LO-TO splitting experimentally verified or only inferred from indirect observations?

Response: The reviewer’s insightful comment is highly valuable and well taken, as it highlights the need to distinguish indirect MIR-optical evidence from quantitative

experimental validation of the phonon dielectric response. We agree that, in the original manuscript, the suppression of LO-TO splitting was mainly inferred from the MIR optical response and structural evidence, rather than being quantitatively extracted from the phonon dielectric response. To address this issue, we have now added polarized infrared reflectance measurements and dielectric-function fitting for both bulk and 2D YMnO₃.

In the revised analysis, the TO and LO phonon frequencies were not assigned from apparent reflectance maxima or minima. Instead, they were extracted by fitting the experimentally measured polarized infrared reflectance spectra using a factorized dielectric-function model. This procedure allows the LO-TO splitting, defined as $\Delta\omega = \omega_{\text{LO}} - \omega_{\text{TO}}$, to be quantitatively evaluated for the infrared-active polar phonon modes. To ensure a meaningful comparison, we focused on the modes that could be consistently resolved and reliably fitted in both bulk and 2D YMnO₃.

The extracted phonon parameters show that the consistently resolved representative polar modes in 2D YMnO₃ possess smaller LO-TO splitting than their bulk counterparts. This trend was obtained from the fitted dielectric function under the same analysis protocol, rather than from visual inspection of the reflectance spectra. Therefore, the suppression of LO-TO splitting is now supported by model-based experimental quantification through polarized infrared reflectance and dielectric-function fitting, rather than only by indirect inference from MIR transparency. The detailed fitting procedure, dielectric-function analysis, and extracted phonon parameters are provided in our response to Comment 4-4, **Fig. R12**, and **Table R8**.

Comment 4-4: The claim of decoupling visible coloration and MIR response is central; however, no direct measurement of LO-TO splitting is presented. This should be experimentally validated.

Response: We appreciate the reviewer's constructive comment. We agree that the proposed visible-MIR decoupling should be supported by experimental validation of the polar-phonon response, rather than relying only on phenomenological MIR-transparency observations. In the revised manuscript, we therefore added polarized infrared reflectance measurements and effective factorized dielectric-function fitting to quantitatively extract the LO-TO splitting in bulk and 2D YMnO₃.

Polarized infrared reflectance spectra of bulk YMnO₃ and 2D YMnO₃ were collected under the same spectral range, polarization configuration, and incidence geometry, so that the extracted phonon parameters could be compared on an equivalent experimental basis. The phonon-response region was fitted using an effective factorized dielectric-function model:

$$\varepsilon(\omega) = \varepsilon_{\infty} \prod_j \frac{\omega_{\text{LO},j}^2 - \omega^2 - i\gamma_{\text{LO},j}\omega}{\omega_{\text{TO},j}^2 - \omega^2 - i\gamma_{\text{TO},j}\omega}$$

where $\omega_{\text{TO},j}$ and $\omega_{\text{LO},j}$ denote the transverse and longitudinal optical phonon frequencies of the j -th infrared-active polar mode, respectively; $\gamma_{\text{TO},j}$ and $\gamma_{\text{LO},j}$ are the

corresponding damping constants; and ϵ_∞ is the high-frequency dielectric constant. The calculated reflectance was obtained from the fitted dielectric response through the Fresnel equations using the same polarization and incidence geometry as in the experiment.

In this analysis, the LO-TO splitting was not obtained by directly reading apparent maxima or minima from the reflectance spectra. Instead, the TO and LO frequencies were extracted from the fitted dielectric response, where TO modes correspond to the poles of the dielectric function and LO modes correspond to its zeros. The LO-TO splitting for each polar mode was then calculated as:

$$\Delta\omega_{\text{LO-TO},j} = \omega_{\text{LO},j} - \omega_{\text{TO},j}$$

The fitting quality was evaluated by comparing the measured and calculated reflectance spectra, together with the corresponding residuals. As shown in **Fig. R12 a, b**, the effective FPSQ fitting captures the principal Reststrahlen-type features of both bulk and 2D YMnO₃, particularly in the phonon-response region that is most relevant to MIR lattice absorption. The representative extracted TO and LO frequencies and the corresponding LO-TO splitting values are summarized in **Table R8**.

The extracted parameters show a systematic contraction of the LO-TO splitting in 2D YMnO₃ relative to bulk YMnO₃ for the representative and well-resolved infrared-active modes listed in Table R8. These modes were selected because their TO and LO components could be reliably extracted from the FPSQ fitting and compared between bulk and 2D YMnO₃ on the same fitting basis. For the lower-frequency polar modes, the LO-TO splitting decreases from 19.78 to 16.31 cm⁻¹ and from 14.94 to 12.69 cm⁻¹, corresponding to reductions of approximately 17.54% and 15.06%, respectively. Although the absolute splitting values of these low-frequency modes are relatively small, their consistent decrease indicates that the reduction of polar coupling is not limited to a single isolated fitting feature.

More importantly, the contraction becomes more pronounced for the modes associated with the stronger Reststrahlen-type response. For example, the LO-TO splitting decreases from 94.16 cm⁻¹ in bulk YMnO₃ to 51.76 cm⁻¹ in 2D YMnO₃, corresponding to a reduction of 45.03%. For the high-frequency Reststrahlen-related mode, the splitting decreases from 368.62 to 239.86 cm⁻¹, corresponding to a reduction of 34.93%. Since the Reststrahlen band width and intensity in polar oxides are strongly governed by the separation between TO and LO phonon frequencies, this pronounced reduction provides direct experimental evidence for a compressed polar-phonon response in 2D YMnO₃. This result supports the proposed weakening of the effective long-range Coulomb contribution to the lattice response after dimensional reduction.

These experimentally extracted LO-TO splitting values directly support the mechanism proposed in the manuscript. In polar oxides, the MIR Reststrahlen response originates from infrared-active lattice vibrations and the macroscopic electric field associated with longitudinal optical modes. Therefore, a reduced LO-TO splitting indicates a narrowed polar-phonon loss channel and a weakened Reststrahlen-type MIR response. Because the visible blue coloration of YMnO₃ originates primarily from visible-region electronic transitions and selective reflection,

whereas the MIR response is governed by polar lattice vibrations, the experimentally verified contraction of LO-TO splitting supports the proposed decoupling between visible coloration and MIR radiative response in 2D YMnO₃. Accordingly, the *Revised Manuscript* now includes the polarized infrared reflectance fitting, residual analysis, and representative LO-TO splitting comparison, All relevant revisions have been highlighted in red in the *Revised Manuscript*.

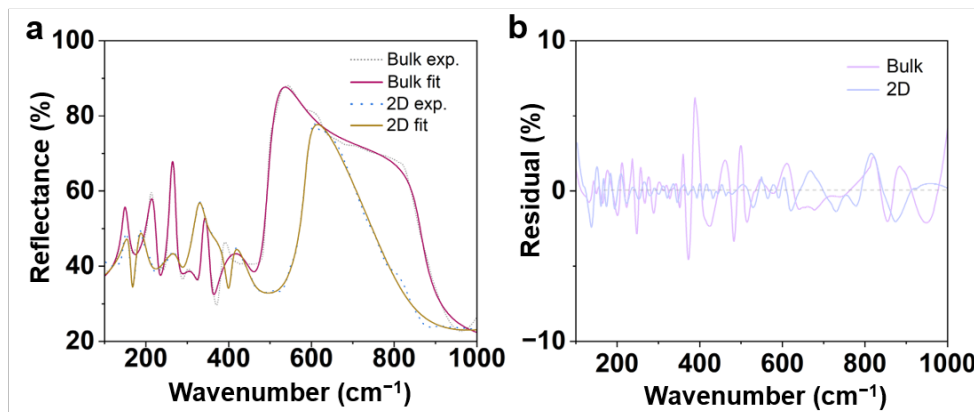


Fig. R12. Polarized infrared reflectance fitting and residual analysis of bulk and 2D YMnO₃. **a.** Experimental and FPSQ-fitted polarized infrared reflectance spectra of bulk and 2D YMnO₃. **b.** Corresponding fitting residuals, defined as $\Delta R = R_{\text{exp}} - R_{\text{fit}}$.

Table R8. Representative LO-TO splitting values extracted from effective FPSQ fitting of polarized infrared reflectance spectra.

Mode	Sample	$\omega_{\text{TO}} / \text{cm}^{-1}$	$\omega_{\text{LO}} / \text{cm}^{-1}$	$\Delta\omega_{\text{LO-TO}} / \text{cm}^{-1}$	Reduction of $\Delta\omega_{\text{LO-TO}}$ relative to bulk
1	Bulk	208.01	227.79	19.78	—
1	2D	184.90	201.21	16.31	17.54%
2	Bulk	258.25	273.19	14.94	—
2	2D	272.65	285.33	12.69	15.06%
3	Bulk	369.00	463.15	94.16	—
3	2D	407.65	459.41	51.76	45.03%
4	Bulk	493.92	862.53	368.62	—
4	2D	587.73	827.59	239.86	34.93%

Comment 4-5: Can the reduction in long-range polarization be directly evidenced (e.g., dielectric function or Born effective charge analysis)?

Response: We are grateful for the reviewer’s insightful comment, as it highlights an important mechanistic point in our interpretation. As described in our response to Comment 4-4, we have added polarized infrared reflectance measurements and factorized dielectric-function fitting to extract representative TO and LO phonon frequencies for bulk and 2D YMnO₃. In the present response, we further focus on the physical implication of the extracted dielectric-function parameters, especially their relevance to the reduction in long-range polarization.

In polar crystals, LO-TO splitting originates from the macroscopic electric field generated by long-wavelength polar lattice vibrations. Therefore, the TO-LO

frequency separation provides an experimentally accessible indicator of the long-range Coulomb contribution to the polar-phonon response. The extracted fitting results show that 2D YMnO₃ exhibits smaller LO-TO splitting than bulk YMnO₃ for the representative infrared-active modes. The splitting is reduced by 17.54%, 15.06%, 45.03%, and 34.93% for the selected modes, respectively. This compression of LO-TO splitting provides phonon-level experimental evidence that the long-range polar contribution is weakened in the 2D structure.

To further substantiate this interpretation, we have also added the real and imaginary parts of the dielectric function, ϵ_1 and ϵ_2 , extracted from the fitted polar-phonon response. The corresponding dielectric-function spectra are shown in **Fig. R13**. The ϵ_1 spectrum reflects the dispersive dielectric response associated with polar optical phonons, while the ϵ_2 spectrum represents the corresponding phonon-induced dielectric loss. As shown in Fig. R13, compared with bulk YMnO₃, 2D YMnO₃ exhibits a weakened effective Reststrahlen-type dielectric response, characterized by a less pronounced negative- ϵ_1 region and a reduced dominant ϵ_2 phonon-loss feature. These changes indicate that the macroscopic field associated with infrared-active polar vibrations is suppressed in 2D YMnO₃.

This dielectric-function analysis directly supports the mechanism proposed in the manuscript: dimensional confinement weakens the long-range polar contribution, compresses the LO-TO splitting, narrows the Reststrahlen-type response, and thereby reduces MIR phonon loss. Thus, the MIR transparency is no longer discussed only as a phenomenological observation, but is now linked to the experimentally extracted polar-phonon dielectric response.

Accordingly, we have revised the manuscript to clarify that the reduction in long-range polarization is supported by the experimentally extracted dielectric-function response, specifically the compressed LO-TO splitting and the weakened Reststrahlen-type dielectric response. This revision establishes a direct link between the measured polarized infrared reflectance, the polar-phonon dielectric response, and the improved MIR transparency of 2D YMnO₃. All relevant revisions have been highlighted in red in the *Revised Manuscript*.

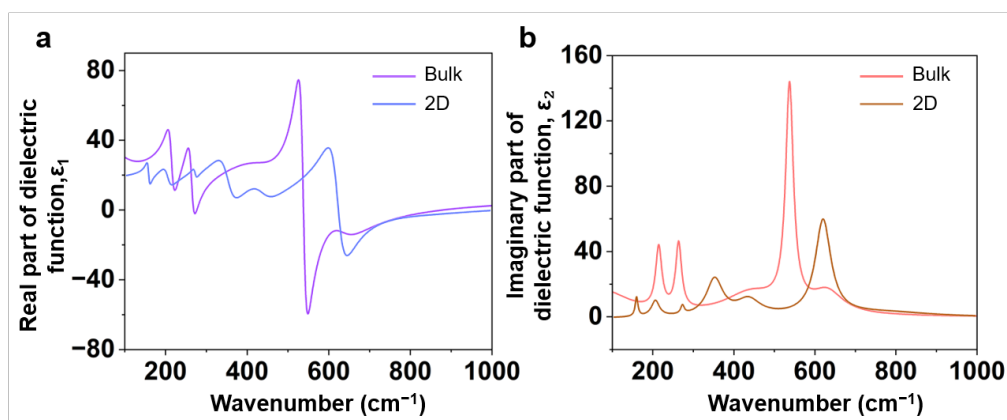


Fig. R13. Extracted dielectric functions of bulk and 2D YMnO₃. a, Real part, ϵ_1 ; narrowed and weakened negative- ϵ_1 Reststrahlen response in 2D YMnO₃. b,

Imaginary part, ϵ_2 ; suppressed polar-phonon dielectric loss in 2D YMnO₃.

Comment 4-6: The manuscript attributes MIR transparency to suppressed Reststrahlen band expansion, but dielectric function (ϵ_1 , ϵ_2) data are not provided. This is a critical missing link.

Response: The reviewer's careful assessment has helped us identify an important mechanistic gap in the original manuscript, namely the absence of dielectric-function evidence linking the MIR optical response to the suppression of Reststrahlen-band expansion. To address this issue, we have added the real and imaginary parts of the dielectric function, ϵ_1 and ϵ_2 , extracted from the fitted polarized infrared reflectance spectra of bulk and 2D YMnO₃. These dielectric-function spectra are now presented in **Fig. R14**.

The dielectric-function analysis provides a direct description of the polar-phonon contribution to the Reststrahlen response. In the factorized dielectric-function model, transverse optical phonons correspond to poles of $\epsilon(\omega)$, whereas longitudinal optical phonons are associated with zeros of $\epsilon(\omega)$. Under finite damping, this phonon response appears as strong dispersion in ϵ_1 and as phonon-loss features in ϵ_2 . In particular, the spectral region where ϵ_1 becomes negative corresponds to a Reststrahlen-type response, in which electromagnetic propagation is strongly suppressed and infrared reflection is enhanced. Therefore, the width and magnitude of the negative- ϵ_1 region provide a dielectric-function measure of Reststrahlen-band expansion.

As shown in Fig. R14a, the extracted ϵ_1 spectra show that the Reststrahlen-type response is substantially weakened in 2D YMnO₃ compared with bulk YMnO₃. Specifically, the negative- ϵ_1 region becomes narrower and shallower in the 2D sample, indicating that the infrared region with metallic-like dielectric response is compressed. Since a larger LO-TO separation generally broadens the frequency interval over which ϵ_1 remains negative, the reduced LO-TO splitting in 2D YMnO₃ provides direct dielectric-function evidence for suppressed Reststrahlen-band expansion.

The ϵ_2 spectra further support this interpretation, as shown in Fig. R14b. Because ϵ_2 represents the phonon-induced dielectric loss, the reduced dominant ϵ_2 loss feature in 2D YMnO₃ indicates a weaker polar-phonon absorption response than that of bulk YMnO₃. Thus, the 2D structure not only compresses the negative- ϵ_1 Reststrahlen-type region, but also decreases the associated phonon-loss contribution in the infrared region.

These newly added dielectric-function results fill the missing mechanistic link identified by the reviewer. They show that the MIR transparency of 2D YMnO₃ is not merely inferred from phenomenological optical spectra, but is supported by the experimentally extracted polar-phonon dielectric response. Specifically, dimensional confinement compresses the LO-TO splitting, suppresses the expansion of the negative- ϵ_1 Reststrahlen region, weakens the ϵ_2 phonon-loss response, and thereby reduces the polar-phonon contribution to MIR optical loss. The relevant discussion

and dielectric-function plots have been added to the *Revised Manuscript*.

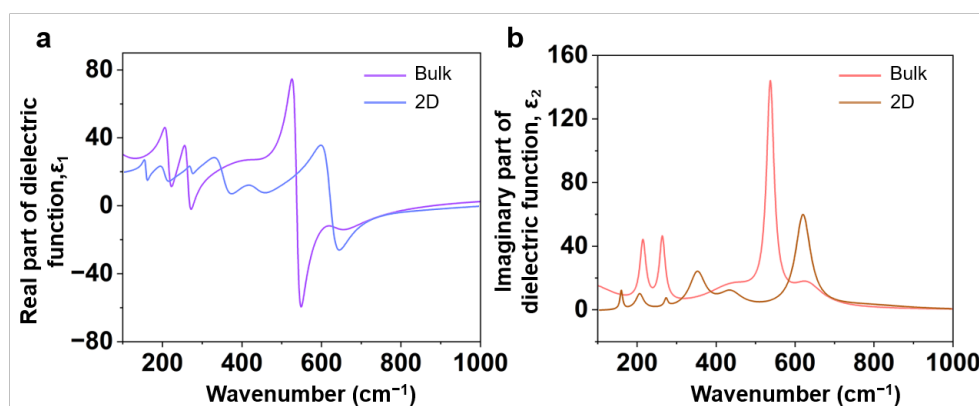


Fig. R14. Extracted dielectric functions of bulk and 2D YMnO₃. a, Real part, ϵ_1 ; narrowed and weakened negative- ϵ_1 Reststrahlen response in 2D YMnO₃. b, Imaginary part, ϵ_2 ; suppressed polar-phonon dielectric loss in 2D YMnO₃.

Comment 4-7: The absence of DFT or phonon dispersion calculations makes the mechanistic explanation largely speculative. Can the authors provide theoretical validation?

Response: We appreciate the reviewer’s constructive comment regarding the theoretical basis of the proposed mechanism. We agree that the original manuscript did not provide sufficient theoretical support for the dimensional-confinement effect in 2D YMnO₃. In the revised manuscript, we have therefore added DFT-based Bader charge and electron localization function analyses to provide electronic-structure-level support for the proposed mechanism.

The purpose of these calculations is to evaluate two electronic-structure prerequisites that are directly related to the proposed MIR-regulation mechanism. First, if dimensional confinement is responsible for weakening polarization-related MIR lattice responses, the 2D configuration should not exhibit enhanced static ionicity or stronger cation-anion charge separation, because such a trend would be unfavorable for reducing the long-range polarization contribution to infrared-active lattice vibrations. Second, the local Y-O/Mn-O coordination framework should remain structurally and electronically intact, since a severe disruption of these oxide coordination units would generate additional vibrational disorder and MIR-loss channels rather than improve MIR transparency.

The Bader charge analysis shows that the net charges of Y, Mn, and averaged O are +2.18, +1.61, and -1.26 e for bulk YMnO₃, respectively, whereas the corresponding values for 2D YMnO₃ are +2.13, +1.58, and -1.24 e. In addition, the O-site charge distribution narrows from -1.30 to -1.24 e in bulk YMnO₃ to -1.26 to -1.22 e in 2D YMnO₃. Although these variations are moderate, they are internally consistent and indicate that the transition from bulk to 2D YMnO₃ does not induce pronounced electronic reconstruction. Instead, the 2D configuration slightly attenuates the static charge separation between the cationic and anionic sublattices. This trend is consistent

with the proposed dimensional-confinement picture, in which the polarization-related contribution to the MIR lattice response is reduced rather than reinforced.

The ELF maps provide complementary information on the local bonding environment. When plotted using the same 0-1 scale, bulk and 2D YMnO₃ exhibit comparable electron-localization features around the Y-O and Mn-O coordination units. This similarity indicates that the dimensional reduction does not lead to a collapse of the local oxide coordination framework. Therefore, the improved MIR response of 2D YMnO₃ is unlikely to arise from extensive bond disruption or amorphization-like structural disorder. Rather, the DFT results support a more specific structural picture: dimensional confinement slightly weakens the static charge-separation tendency of the polar oxide lattice while retaining the local Y-O/Mn-O coordination framework required for stable lattice vibrations.

These DFT analyses therefore provide targeted electronic-structure evidence for the two mechanistic prerequisites discussed above, namely reduced static ionicity/charge separation and preserved local oxide coordination in 2D YMnO₃. Together with the revised MIR optical analysis discussed in the manuscript, these results provide a more grounded theoretical basis for the role of dimensional confinement in regulating the MIR response of 2D YMnO₃ and reduce the speculative nature of the original mechanistic interpretation. The relevant results have been added as **Fig. R15** and **Table. R9** in the *Revised Supporting Information*, and the corresponding discussion has been incorporated into the *Revised Manuscript* with the changes highlighted in red.

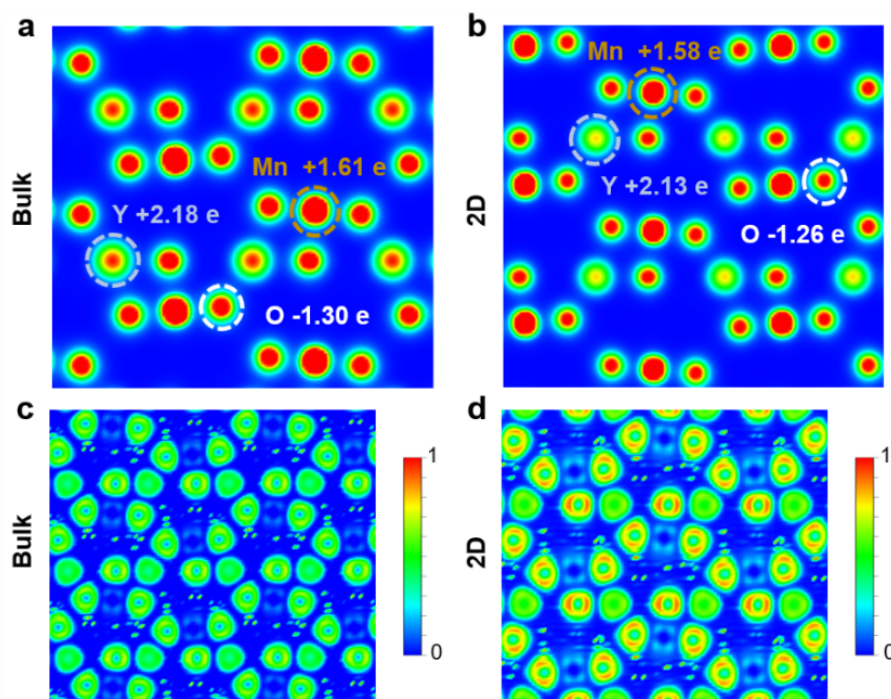


Fig. R15. DFT-based comparison of bulk and 2D YMnO₃. a, b, Bader charge distributions. c, d ELF maps plotted on a common 0-1 scale.

Table R9. Bader net-charge comparison of bulk and 2D YMnO₃.

Structure	q_Y / e	q_{Mn} / e	$q_{O,avg} / e$	O-site charge range / e
Bulk	+2.18	+1.61	-1.26	-1.30 to -1.24
2D	+2.13	+1.58	-1.24	-1.26 to -1.22

Comment 4-8: How reproducible is the microwave-shock synthesis in terms of phase purity and nanosheet thickness?

Response: We appreciate the reviewer’s careful attention to the reproducibility of the microwave-shock synthesis, which is critical for validating the reliability of this non-equilibrium synthetic route. To address this issue in a direct and experimentally grounded manner, we evaluated reproducibility along the two dimensions specified by the reviewer: phase purity and nanosheet thickness. Specifically, phase-purity reproducibility was assessed using repeated XRD/Rietveld refinements of products obtained from three independent synthesis batches, while nanosheet-thickness reproducibility was examined using additional AFM topographic images and line-scan height profiles.

For phase formation, we repeated the microwave-shock synthesis under identical experimental conditions and collected XRD patterns from three independent synthesis batches. The corresponding XRD/Rietveld refinement results have been added to the revised Supporting Information as Fig. S5 and are reproduced here as **Fig. R16** for ease of evaluation. All three batches show consistent diffraction peak positions assignable to hexagonal YMnO₃, with no detectable crystalline secondary phases within the XRD detection limit. The Rietveld refinements also give comparable reliability factors: $R_{wp} = 7.2\%$, $R_p = 5.79\%$, and $\chi^2 = 3.12$ for Batch 1; $R_{wp} = 6.3\%$, $R_p = 4.87\%$, and $\chi^2 = 2.91$ for Batch 2; and $R_{wp} = 6.9\%$, $R_p = 5.29\%$, and $\chi^2 = 2.87$ for Batch 3. These results demonstrate that the microwave-shock process reproducibly yields hexagonal YMnO₃ under the present synthesis conditions.

For nanosheet thickness, we added additional AFM topographic images with overlaid line-scan height profiles to the revised Supporting Information as Fig. S2, and these data are also reproduced here as **Fig. R17**. The representative nanosheets show apparent edge step heights of approximately 0.6-0.9 nm, consistent with the initially reported ultrathin morphology. We describe these values as apparent AFM topographic heights because height measurements of ultrathin nanosheets can be affected by surface adsorption, tip-sample interaction, and local substrate effects. Therefore, the AFM results are used to support the reproducible formation of ultrathin nanosheet morphology, rather than to claim definitive crystallographic confirmation of a strictly monolayer structure.

Taken together, the newly added repeated-batch XRD/Rietveld refinements and AFM height-profile data demonstrate that the microwave-shock synthesis reproducibly produces hexagonal YMnO₃ with an ultrathin nanosheet morphology under the present experimental conditions. We have revised the manuscript accordingly and added the corresponding data to the Supporting Information as Fig. S2 and Fig. S5. All changes are highlighted in red.

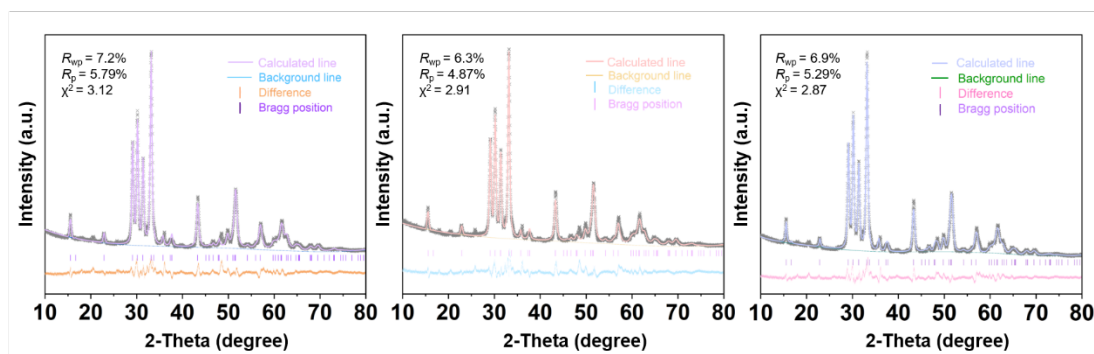


Fig. R16. Reproducibility of phase formation in repeated microwave-shock syntheses. XRD patterns and Rietveld refinements of YMnO_3 products from three independent synthesis batches.

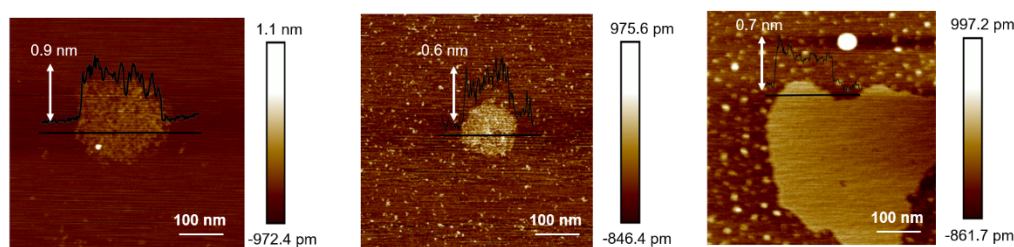


Fig. R17. AFM height profiles of YMnO_3 nanosheets. AFM topographic images with overlaid line-scan profiles showing apparent step heights of 0.9, 0.6, and 0.7 nm. Scale bars, 100 nm.

Comment 4-9: What is the scalability and batch-to-batch consistency of the non-equilibrium synthesis method?

Response: We appreciate the reviewer's thoughtful comment on the scalability and batch-to-batch consistency of the non-equilibrium synthesis method. This issue is important for evaluating the reliability of a microwave-shock route, because the reaction involves rapid microwave-energy input, transient local heating, and gas-assisted structural evolution.

To clarify the processing scale and experimental window, we have revised the Methods section to specify the key synthesis parameters. In the optimized procedure, 0.4 g glucose, 0.5 g NH_4Cl , 0.12 g $\text{YCl}_3 \cdot 6\text{H}_2\text{O}$, and 0.078 g $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$ were used, corresponding to a total precursor loading of approximately 1.10 g per batch. The precursor mixture was ground for 1 h to improve compositional homogeneity and then treated by microwave irradiation at 650 W for 10 min under ambient conditions, with a representative peak temperature of approximately 850 °C. These parameters define the gram-scale laboratory processing window used to obtain the YMnO_3 nanosheets reported in this work.

Within this defined processing window, the synthesis showed consistent formation of hexagonal YMnO_3 nanosheets at the laboratory scale. As discussed in the responses above, repeated XRD measurements from independent synthesis batches confirmed

the formation of hexagonal YMnO_3 without detectable crystalline secondary phases within the XRD detection limit, while the additional AFM height profiles further supported the reproducible formation of ultrathin nanosheet morphologies. These results support the batch-to-batch consistency of the present synthesis under the controlled gram-scale conditions used in this study.

Regarding scalability, the present synthesis should be interpreted as a gram-scale laboratory demonstration with a defined precursor loading of approximately 1.10 g per batch, rather than as an industrial-scale manufacturing process. Nevertheless, the process contains several features that provide a practical basis for future throughput-oriented optimization, including ambient-pressure operation, a solid precursor mixture, short microwave irradiation time, and the absence of vacuum, protective atmosphere, or high-pressure reaction vessels.

At the same time, we agree that the scale-up of a microwave-shock process cannot be achieved simply by increasing the powder mass or precursor-bed thickness. Because the formation of ultrathin YMnO_3 nanosheets depends on rapid microwave-energy coupling, transient local heating, and gas evolution assisted by glucose and NH_4Cl , larger-scale implementation would require preservation of a similar local reaction environment throughout the precursor bed. Therefore, the key scale-up considerations are not changes in the reaction chemistry itself, but engineering control of microwave-field uniformity, precursor-bed geometry, heat dissipation, and homogeneous gas release. This distinction clarifies that the present work demonstrates controlled gram-scale batch consistency, while larger-scale production would require further process-engineering optimization.

Comment 4-10: The reported thickness (~0.7 nm) suggests a monolayer structure, but no direct crystallographic or layer-number confirmation (e.g., cross-sectional TEM or AFM statistics) is provided.

Response: We are grateful for the reviewer's careful assessment of the thickness assignment and for requesting clearer evidence regarding the layer-number interpretation. We agree that an apparent AFM height of approximately 0.7 nm should be interpreted as topographic evidence for an ultrathin morphology, rather than as definitive crystallographic confirmation of a strictly monolayer structure.

In the revised Supporting Information, we have updated Fig. S2 by including AFM topographic images with overlaid line-scan height profiles obtained from independently sampled YMnO_3 nanosheets. For ease of evaluation, the same data are also reproduced in this response as **Fig. R18**. The apparent edge step heights obtained from these measurements are approximately 0.6-0.9 nm, consistent with the originally reported value of approximately 0.7 nm. These AFM measurements provide additional topographic evidence that the nanosheets exhibit sub-nanometer apparent heights, supporting an ultrathin 2D morphology rather than thick or bulk-like particles.

It should be noted that AFM height analysis provides topographic evidence for the ultrathin morphology, whereas the crystallographic structure and phase assignment are supported separately by HRTEM/SAED and Rietveld-refined XRD. HRTEM and

SAED confirm the crystalline in-plane ordering of the nanosheets, and the Rietveld-refined XRD pattern verifies the hexagonal YMnO₃ phase. Taken together, these results support the assignment of crystalline ultrathin 2D YMnO₃ nanosheets, while avoiding an overinterpretation of AFM height as direct crystallographic layer-number confirmation.

Accordingly, we have revised the relevant statement in the Results section to describe the AFM data as apparent edge step heights of approximately 0.6-0.9 nm, rather than as a definitive layer-number assignment. We have also updated Section 1 of the Supporting Information and revised Fig. S2 and its caption to include the additional independent AFM line-scan measurements. All corresponding revisions in the *Revised Manuscript* and *Revised Supporting Information* have been highlighted in red.

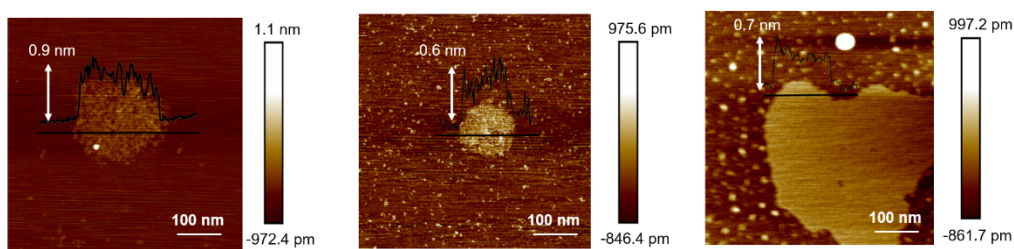


Fig. R18. AFM height profiles of YMnO₃ nanosheets. AFM topographic images with overlaid line-scan profiles showing apparent step heights of 0.9, 0.6, and 0.7 nm. Scale bars, 100 nm.

Comment 4-11: The proposed mechanism relies on reduced *Born effective charge (Z)***, yet no quantitative or computational evidence is shown—this weakens the core argument.

Response: We sincerely appreciate this insightful comment, which correctly identifies the need for quantitative evidence supporting the proposed reduction in dynamic charge response. In the original manuscript, the weakened long-range polar response was mainly discussed on the basis of the reduced LO-TO splitting and the suppressed Reststrahlen-band expansion, whereas the associated dynamic effective charge response was not quantitatively evaluated. To address this important point, we have now added an estimation of the mean effective charges for bulk and 2D YMnO₃ using the infrared phonon oscillator strengths extracted from the polarized infrared reflectance fitting.

The analysis follows the polarized-reflectivity method previously reported for hexagonal YMnO₃, in which the mean effective charges were estimated from infrared phonon fit parameters. (DOI: [10.1103/PhysRevB.78.184305](https://doi.org/10.1103/PhysRevB.78.184305)) In this treatment, the total oscillator strength of the infrared-active polar phonons is related to the dynamic charge response by:

$$\sum_i \omega_{p,i}^2 = \frac{4\pi}{V_{cell}} \sum_k \frac{N_k (Z_k^*)^2}{m_k}$$

where Z_k^* is the effective charge of atom k , m_k is the corresponding atomic mass, N_k is

the number of atom k in the crystallographic unit cell, V_{cell} is the unit-cell volume, and $\omega_{p,i}$ is the effective plasma frequency of the i -th infrared-active phonon mode obtained from the reflectance fitting. The expression is written in the Gaussian-unit convention commonly used in infrared phonon oscillator-strength analysis, and the same unit convention was applied to both bulk and 2D YMnO₃.

Equivalently, we define:

$$S = \sum_k \frac{N_k (Z_k^*)^2}{m_k} = \frac{V_{\text{cell}}}{4\pi} \sum_i \omega_{p,i}^2$$

This relation provides an experimental route to estimate the mean dynamic effective charge from the oscillator strength of polar phonon modes, rather than relying solely on qualitative interpretation of the reflectance spectra.

For hexagonal YMnO₃, the crystallographic unit cell contains 6 Y, 6 Mn, and 18 O atoms. Therefore,

$$S = \frac{6(Z_Y^*)^2}{m_Y} + \frac{6(Z_{Mn}^*)^2}{m_{Mn}} + \frac{18(Z_O^*)^2}{m_O}$$

Because YMnO₃ is a ternary compound, Z_Y^* , Z_{Mn}^* , and Z_O^* cannot be independently determined from this relation alone without additional constraints. Therefore, following the reported treatment for hexagonal YMnO₃, we used the electroneutrality condition:

$$6Z_Y^* + 6Z_{Mn}^* + 18Z_O^* = 0$$

and adopted a mean-cation approximation:

$$Z_Y^* = Z_{Mn}^* = Z_{Y/Mn}^*$$

This approximation does not imply that the atom-specific effective charges of Y and Mn are identical. Rather, it provides a mean cationic dynamic effective charge that allows a consistent comparison between bulk and 2D YMnO₃ under the same fitting protocol. Under this approximation,

$$Z_O^* = -\frac{2}{3} Z_{Y/Mn}^*$$

Thus, the expression for S becomes:

$$S = (Z_{Y/Mn}^*)^2 \left(\frac{6}{m_Y} + \frac{6}{m_{Mn}} + \frac{8}{m_O} \right)$$

and the mean effective charge of the cation sublattice can be obtained as:

$$Z_{Y/Mn}^* = \left[\frac{S}{\frac{6}{m_Y} + \frac{6}{m_{Mn}} + \frac{8}{m_O}} \right]^{1/2}$$

All resolved infrared-active phonon modes within the fitted spectral region were included in the oscillator-strength summation for bulk and 2D YMnO₃ under the same polarization geometry. Using the fitted infrared phonon plasma frequencies, the mean $Z_{Y/Mn}^*$ decreases from 4.92 in bulk YMnO₃ to 4.55 in 2D YMnO₃, corresponding to a reduction of approximately 7.5%. The magnitude of the oxygen effective charge also decreases from 3.28 to 3.03 as shown in **Table. R10**. These results indicate that the dynamic charge response associated with polar lattice vibrations is weakened in the 2D structure.

This result is consistent with the independently extracted LO-TO splitting. The representative infrared-active modes in 2D YMnO₃ show smaller LO-TO splittings than those in bulk YMnO₃, indicating a reduced effective long-range dipole–dipole contribution to the polar phonon response. Therefore, the reduced mean effective charge, reduced LO-TO splitting, and narrowed negative- ϵ_1 Reststrahlen region provide mutually consistent evidence that dimensional confinement weakens the effective long-range polarization response in 2D YMnO₃. We have revised the manuscript accordingly and now present these values as mean effective charges estimated from infrared phonon oscillator strengths. This quantitative analysis strengthens the mechanistic link between the reduced dynamic charge response, suppressed LO-TO splitting, narrowed Reststrahlen-band expansion, and enhanced MIR transparency in 2D YMnO₃.

Table R10. Mean dynamic effective charges estimated from infrared phonon oscillator strengths for bulk and 2D YMnO₃.

Sample	Mean $Z_{Y/Mn}^*$ estimated from IR oscillator strength	Mean Z_O^* estimated from charge neutrality
Bulk	4.92	−3.28
2D	4.55	−3.03

Comment 4-12: Can the authors clarify the role of glucose-derived carbon and whether residual carbon affects optical properties?

Response: We are grateful to the reviewer for raising this important point. Clarifying the role of glucose-derived carbon is essential for distinguishing the synthesis pathway of 2D YMnO₃ from the origin of its final visible/MIR optical response. In particular, if residual carbonaceous species were retained as a substantial phase, they could complicate the interpretation of the measured optical properties. We have therefore revised the manuscript to clarify that glucose-derived species mainly function as transient structure-directing intermediates during the microwave-pulse synthesis, and to address whether residual carbonaceous species are likely to govern the optical response of the final coating.

In our microwave-pulse synthesis, glucose is not introduced as a final optical functional component. Instead, it acts primarily as a sacrificial carbonaceous precursor and microwave-responsive fuel during the rapid heating process. Under microwave irradiation, glucose can generate transient carbonaceous intermediates through dehydration/carbonization-related processes, which may absorb microwave energy and create localized high-temperature microenvironments. More importantly, these glucose-derived intermediates provide a temporary confining matrix for the Y/Mn-containing precursor species, which helps disperse the metal precursors, restricts extensive mass transport, and suppresses uncontrolled particle coalescence during the short reaction period. Meanwhile, gas evolution from NH₄Cl decomposition produces a local expanding/bubbling environment. The coupling between the transient carbonaceous confinement and gas-assisted expansion favors interfacial growth and lamellar separation, thereby promoting the formation of two-

dimensional YMnO_3 nanosheets rather than densely aggregated particles.

To support this structure-directing role, we added comparative SEM observations of products synthesized with and without glucose under otherwise comparable conditions. **(Fig. R19)** In the absence of glucose, the product shows a more aggregated and poorly defined morphology, whereas the glucose-assisted synthesis yields nanosheet-like YMnO_3 . This comparison indicates that glucose-derived species mainly participate in morphology regulation during the microwave-pulse process, rather than serving as a retained optical functional phase in the final material.

We further used Raman spectroscopy to examine whether a Raman-detectable carbonaceous phase remains in the final product. **(Fig. R20)** In the typical D/G-band region, no discernible carbon-related bands are observed around 1350 and 1580 cm^{-1} , where defect-activated and graphitic sp^2 carbon vibrations are commonly detected. This result argues against the presence of a substantial separated sp^2 -rich carbonaceous phase in the as-prepared YMnO_3 sample. We note that trace surface carbonaceous species below the Raman detection limit cannot be completely excluded. Nevertheless, the absence of detectable D/G bands does not support the presence of a continuous or dominant residual carbon phase that would govern the visible/MIR optical response.

This interpretation is further consistent with the high-temperature functional stability of the coating. If the MIR thermal-management behavior were primarily dominated by labile glucose-derived carbon residues, a pronounced change would be expected after prolonged annealing in air, where carbonaceous species are prone to oxidative removal. However, the YMnO_3 coating retains a clear infrared-thermographic contrast after annealing at $900\text{ }^\circ\text{C}$ for 8 h. Although this result is not a direct quantification of residual carbon, it provides a relevant functional-level constraint, supporting that the observed MIR response is not primarily dependent on unstable glucose-derived carbon residues.

Accordingly, we have revised the manuscript to clarify that glucose-derived species mainly act as transient structure-directing intermediates during the microwave-pulse synthesis, while the visible/MIR optical properties of the final YMnO_3 coating are unlikely to be governed by a substantial residual carbonaceous phase. The comparative spectra have been added in the *Revised Supporting Information*, and the corresponding discussion has been incorporated into the *Revised Manuscript* in red.

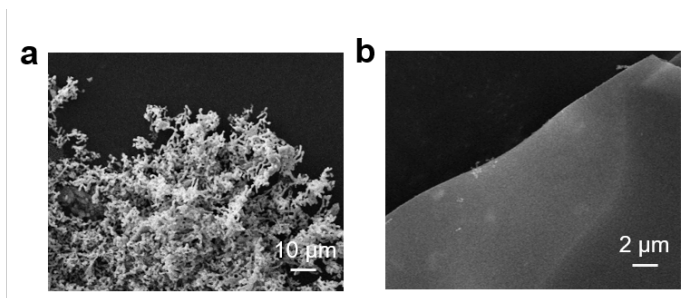


Fig. R19. SEM comparison of YMnO_3 synthesized without and with glucose. a, Without glucose. b, With glucose.

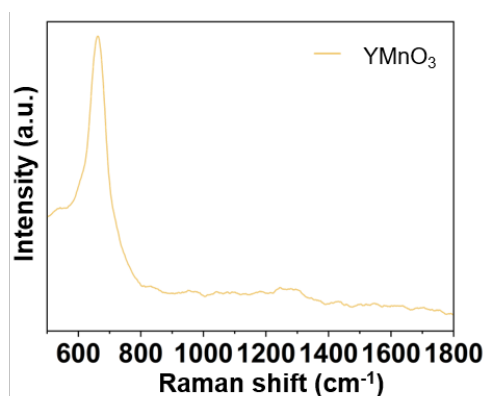


Fig. R20. Extended Raman spectrum of 2D YMnO₃ for evaluating residual carbonaceous species.

Comment 4-13: The high-temperature stability test (8 h at 100 °C, 900 °C annealing) is limited-long-term cycling and oxidation resistance are not sufficiently explored.

Response: We are grateful to the reviewer for raising this important and practically relevant concern. We agree that long-term thermal cycling and oxidation resistance are critical considerations for evaluating high-temperature functional coatings under practical service conditions. We would first like to clarify that the 8 h test at 100 °C in the original manuscript was a continuous thermal-exposure experiment rather than a transient measurement, and therefore provided an initial sustained-heating baseline for assessing the stability of the MIR radiative response.

From a materials perspective, YMnO₃ is a thermally formed oxide ceramic phase, in which the constituent cations are incorporated into a high-temperature oxide lattice. This is fundamentally different from low-temperature organic, carbonaceous, or ultrathin metallic functional layers that may undergo thermal decomposition, oxidative consumption, dewetting, or rapid loss of film continuity under high-temperature air exposure. This oxide-ceramic nature provides a materials-level rationale for the intrinsic thermal tolerance of the YMnO₃ framework. Nevertheless, we agree that this rationale cannot replace direct stability evaluation of the coating/substrate system, because prolonged exposure in air may still affect the broadband optical response, interfacial radiative boundary condition, and device-level MIR camouflage performance.

To further address this concern, we extended the 900 °C air-annealing duration from 8 h to 24 h. The corresponding broadband reflectance spectrum and infrared thermography comparison before and after annealing are provided in **Fig. R21** and **R22**, respectively. This prolonged isothermal air-annealing experiment was designed to directly examine whether high-temperature oxidative aging would compromise the broadband optical response or device-level MIR radiative function of the YMnO₃-coated metallic system. We note that this experiment does not constitute a complete long-term thermal-cycling lifetime test. Rather, it specifically addresses the stability

of the coated metallic system under prolonged high-temperature air exposure, which is one of the key degradation scenarios raised by the reviewer.

The additional results show that the functional optical response is largely preserved after 900 °C/24 h air annealing. The broadband reflectance spectrum maintains the characteristic optical profile of the YMnO₃ coating, without obvious spectral-shape degradation or the emergence of new MIR loss features within the tested range. More importantly, infrared thermography shows that the apparent temperature of the coated sample remains nearly unchanged after annealing, increasing only slightly from 53.6 °C to 53.8 °C under the same measurement conditions. This negligible difference indicates that the effective MIR radiative boundary condition of the coated metallic system is largely retained after prolonged high-temperature oxidative exposure.

Therefore, these additional data support the prolonged isothermal thermal/oxidative aging stability of the YMnO₃-coated metallic system under the tested conditions. At the same time, to avoid overextending the conclusion, we clarify that these results should not be interpreted as a complete long-term thermal-cycling lifetime assessment. Systematic thermal-cycling tests, longer-duration environmental-aging experiments, and post-aging microstructural analysis will be valuable in future work for evaluating service reliability under more demanding operational conditions. All corresponding revisions have been highlighted in red in the *Revised Manuscript*.

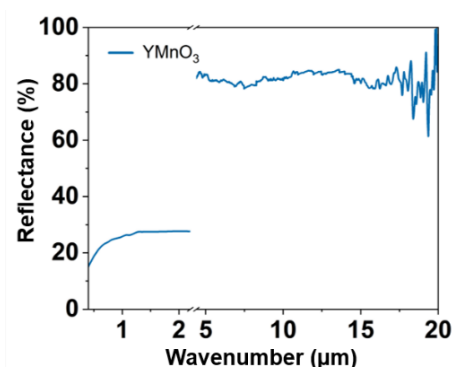


Fig. R21. Broadband reflectance spectrum of the YMnO₃-coated sample after prolonged air annealing.

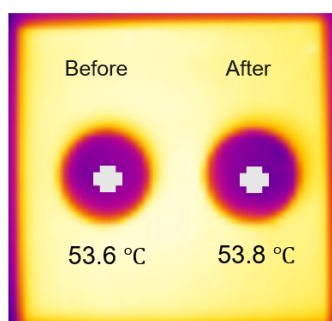


Fig. R22. Infrared thermographs of the coated sample before and after 900 °C/24 h air annealing under identical measurement conditions.

Comment 4-14: The ethanol flame test introduces carbon contamination, which the authors acknowledge-this raises concerns about the reliability of post-test optical data.

Response: We appreciate the reviewer's important comment. We agree that carbon deposition introduced during ethanol-flame exposure can complicate the interpretation of post-flame MIR-radiative data, particularly if a carbon-contaminated surface state is used to infer the intrinsic MIR stability of pristine YMnO_3 . This is because an ethanol flame imposes not only rapid thermal and convective loading, but also a chemically non-ideal environment in which incomplete combustion may introduce surface carbonaceous deposits.

To address this ambiguity, we have revised the manuscript to distinguish short-duration flame impingement from prolonged flame exposure. The ethanol-flame experiment is now described as an examination of the coating response to rapid flame impingement, rather than as a contamination-free high-temperature stability test. The flame-impingement test was designed to represent a practical transient thermal hazard at approximately 750 °C, rather than a chemically clean and quantitative high-temperature stability assessment. Because fuel-based flame environments can introduce carbonaceous deposits under locally incomplete-combustion or prolonged-exposure conditions, the prolonged 53 s result is treated as an extrinsically contaminated surface state, whereas the intrinsic thermal stability is supported by the separate controlled annealing experiments. Under the short exposure condition ($t = 5$ s; ~ 750 °C), the purpose is to evaluate whether rapid flame impingement causes a detectable degradation of the MIR-radiative response of the YMnO_3 -coated metallic surface.

In contrast, when the exposure is extended to 53 s, slight carbon deposition is observed on the flame-exposed surface. We therefore interpret this prolonged-exposure condition as a flame-exposed, carbon-contaminated surface state, rather than as evidence for the intrinsic optical/MIR stability of pristine YMnO_3 . Accordingly, the 53 s result is not used to substantiate the intrinsic MIR performance of YMnO_3 , but is discussed only as a possible extrinsic contamination pathway under prolonged flame exposure.

The main thermographic comparison in Fig. 5c,d is based on the bare metallic surface, the YMnO_3 -coated surface, and the YMnO_3 -coated surface after short-duration flame impingement. Under identical thermography settings, the apparent temperature changes only slightly from 44.0 °C to 44.3 °C after the 5 s flame exposure, indicating that short-duration flame impingement does not cause a substantial change in the measured MIR-radiative response. This restricted interpretation avoids using the carbon-contaminated prolonged-exposure state to support the intrinsic MIR performance claim of pristine YMnO_3 . This clarification has been incorporated into the *Revised Manuscript* through revisions to the main-text discussion and the Fig. 5b caption, with all changes highlighted in red.

Comment 4-15: The coating performance is demonstrated on controlled

substrates, but real aerospace conditions (mechanical stress, erosion, oxidation) are not simulated or discussed.

Response: We are grateful for the reviewer's careful assessment of the service-environment boundary of the coating system. The present work was designed to address a materials-level challenge in visible-MIR spectral management: integrating visible coloration, MIR radiative regulation, and high-temperature robustness within a single YMnO_3 -based coating system. The controlled substrate-level demonstrations were therefore used to verify the essential optical and thermal-radiative functions required for this coating concept under well-defined laboratory conditions.

The practical motivation for this design is closely related to the broader requirement for multispectral signature management on military aircraft surfaces. Public information on the B-2 Spirit indicates that low-observable aircraft rely on simultaneous reduction of infrared, visual, electromagnetic, acoustic, and radar signatures, with special coatings contributing to the overall low-observable performance. In addition, low-emissivity topcoats with visible camouflage colors, such as those based on Aerospace Material Specification Standard 595 36375, have been developed for military platforms to reduce emissivity in thermal-infrared atmospheric windows. These examples are cited to contextualize the practical relevance of combining visible appearance control with thermal-infrared signature suppression, rather than to imply that the present laboratory tests reproduce full aircraft-service conditions.

We also agree with the reviewer that translating such a coating concept to real aerospace service requires evaluation under substantially more complex coupled conditions than those reproduced in the present controlled experiments. In practical aircraft operation, exterior coatings may experience mechanical stress, aerodynamic loading, thermal cycling, particle/rain erosion, interfacial fatigue caused by thermal-expansion mismatch, and oxidative or environmental degradation. These factors are often localized and strongly dependent on aircraft type, surface location, flight condition, coating thickness, and substrate/coating adhesion. For example, public F-35 flight-test reports show that horizontal-tail coatings were modified and instrumented with temperature sensors after coating damage had been observed during flight tests involving extended afterburner use, although such conditions were noted as not being representative of normal operational use. In another high-speed-flight example, NASA reports on SR-71 Mach-3-class operation indicate that the aircraft was designed to cruise at approximately Mach 3.2, above 2,200 mph and up to 85,000 ft, while related thermal-experiment conditions could generate heat-soak temperatures exceeding 600 °F (>316 °C). These examples are not cited to imply that the present laboratory tests reproduce full aircraft-service conditions, but to illustrate why localized heating, coating degradation, and service-induced damage must be considered in future aerospace-coating qualification.

Within this context, the high-temperature annealing, hot-plate thermography, and flame-exposure tests presented in this work should be interpreted as controlled evaluations of the thermal robustness and MIR spectral function of the YMnO_3 -based

coating, rather than as a full simulation of the aerospace service environment. Specifically, the coating was evaluated by long-term hot-plate thermography at 100 °C for 8 h, high-temperature annealing at 900 °C for up to 8 h, hot-plate measurements over 100-350 °C with 50 °C intervals, and ethanol-flame exposure at approximately 750 °C for short and prolonged durations, including 5 s and 53 s flame-impingement tests. These experiments establish that the coating retains the key visible-MIR spectral-management functions under selected high-temperature laboratory conditions. However, we agree that they do not fully reproduce flight-induced mechanical stress, high-speed airflow, particle/rain erosion, interfacial fatigue, or long-term oxidative/environmental degradation.

To address this point, we have revised the manuscript to explicitly clarify the application boundary of the coating system. Specifically, we now state that although the present results demonstrate visible-MIR spectral management and high-temperature robustness under controlled laboratory conditions, practical aerospace deployment will require further application-level qualification under coupled mechanical loading, thermal cycling, erosion, and oxidative/environmental exposure. Future qualification should include adhesion tests on aerospace metallic substrates, thermal-cycling fatigue, mechanical bending or vibration, sand/rain erosion, and long-term oxidative aging under representative aircraft-surface temperature windows. We regard these coupled service-environment issues raised by the reviewer as an essential next-stage research direction for translating the YMnO_3 -based spectral design from a materials-level concept to an application-qualified aerospace coating system. These reliability and durability evaluations are therefore being considered as the focus of our subsequent work.

Comment 4-16: The authors are encouraged to include relevant recent literature in the Introduction to strengthen the scientific background and highlight the current significance of the study.
<https://doi.org/10.1016/j.ceramint.2025.09.340>
<https://doi.org/10.1016/j.jallcom.2025.180821>
<https://doi.org/10.1103/PhysRevMaterials.9.024408>

Response: We appreciate the reviewer's constructive suggestion. We agree that incorporating recent literature is important for strengthening the scientific background and clarifying the current significance of this study. In the revised Introduction, we have added the three suggested recent references at the position where high-temperature-stable inorganic material systems and their functional constraints are discussed.

These references were incorporated not merely as additional citations, but to place our work within the broader and rapidly developing context of YMnO_3 and related hexagonal manganites. The recent study on nanocrystalline YMnO_3 prepared by microwave reaction followed by calcination demonstrates that YMnO_3 -based oxides can exhibit tunable structural, optical, luminescent, and chromaticity characteristics, thereby supporting their relevance for visible optical regulation and color-related

applications. The work on Yb-doped h-YMnO₃ further shows that lattice distortion induced by rare-earth-site substitution can effectively modulate the structural, magnetic, and ferroelectric properties of h-YMnO₃, highlighting the importance of structure-property coupling in this material family. In addition, the Physical Review Materials study further shows that the electronic properties of hexagonal manganites can be regulated through aliovalent doping and atmosphere-dependent thermal treatment, emphasizing the strong coupling among cation substitution, anion defects, charge-carrier behavior, and processing conditions.

Together, these studies demonstrate that YMnO₃ and related hexagonal manganites are currently active platforms for functional regulation across optical, structural, multiferroic, and electronic dimensions. At the same time, they also clarify the remaining gap addressed in our work: previous studies have mainly focused on individual optical, multiferroic, or electronic properties, whereas the simultaneous integration of blue visible appearance, MIR transparency, and high-temperature radiative stability has not been established. Therefore, the newly added references strengthen the Introduction by both updating the recent literature background and highlighting the specific significance of our structure-based strategy for decoupling visible and MIR optical constraints in thermally stable oxides. The corresponding citations and reference-list entries have been added and highlighted in red in the *Revised Manuscript*.

Response to the Referees' comments

We sincerely thank the Editor and all Reviewers for their careful evaluation of our manuscript and for the positive recommendations regarding its publication. We are grateful for the constructive comments and suggestions provided throughout the review process. These insightful remarks have significantly improved the quality, clarity, and presentation of the manuscript. Following the reviewers' recommendations and the editorial guidance, we carefully revised the manuscript, addressed all comments point-by-point, and completed all requested modifications.

We are pleased that the reviewers consider the revised manuscript satisfactory and suitable for publication. We greatly appreciate the time, effort, and expertise devoted by the reviewers and the editorial team during the assessment of our work.