

Response to comments from the Reviewer #3

General Response: We sincerely thank the Referee for the careful evaluation of our manuscript and for the constructive comments. These comments have helped us clarify the relationship between the Raman measurements and the radiative model, refine the interpretation of the carrier–lattice response, better define the temperature range and scope of the present experiment, and improve several descriptions in the manuscript. Our point-by-point responses are provided below.

Comment 1: The connection between the redshift in the Raman spectra and the near-field radiative transfer spectra is not well established. Is it accounted for in the radiative model? To what extent does it matter?

Response 1: We thank the Reviewer for this important comment. We appreciate the opportunity to clarify (i) why Raman spectroscopy was employed, (ii) how the Raman measurements are related to the radiative model, (iii) influence of the redshift in the Raman spectra on near-field radiative transfer spectra, and (iv) how the Raman evidence supports the central novelty of this work. The original manuscript did not sufficiently distinguish these different roles. We have now clarified them as follows.

1. Why Raman spectroscopy provides appropriate supporting evidence

Raman spectroscopy is a well-established method for characterizing carrier-induced modifications of optical phonons in polar semiconductors. In doped GaAs, the macroscopic polarization field associated with lattice vibrations interacts with collective free-carrier oscillations through the long-range Coulomb interaction. This interaction modifies the frequency, linewidth, intensity, and line shape of the optical phonon response.

The use of Raman spectroscopy for this purpose has a substantial experimental and theoretical foundation. Mooradian and Wright reported plasmon–phonon interactions in Raman spectra of GaAs [R1], and Tell and Martin subsequently analyzed Raman scattering by coupled optical-phonon–plasmon modes [R2]. Chandrasekhar and Ramdas systematically investigated the carrier-concentration dependence of coupled plasmon–phonon modes in n-type GaAs [R3]. More recently, Takeuchi et al. combined Raman scattering and terahertz time-domain spectroscopy to characterize phonon–plasmon coupled modes in undoped-GaAs/n-type-GaAs structures [R4]. First-principles calculations by Lihm and Park further reproduced carrier-dependent plasmon–phonon hybridization in doped GaAs and showed agreement with available Raman measurements [R5]. These studies establish Raman spectroscopy as a physically well-founded probe of carrier-dependent changes in the optical phonon response of GaAs.

2. How the Raman measurements are related to the radiative model

In the present work, Raman spectroscopy is not treated as a direct measurement of the evanescent surface modes or the NFRHT spectrum. The measured Raman peak positions were neither inserted directly into nor used to fit the radiative heat-transfer calculations. Instead, the dielectric function used in the fluctuational-electrodynamics model was constructed using independently reported, carrier-concentration-dependent optical parameters for n-type GaAs [R3]. As summarized in Table S1 of the Supplementary Material, these parameters include the TO and LO phonon frequencies, phonon damping constant, plasma frequency, and carrier relaxation rate. They were interpolated to the carrier concentrations of our samples, which were independently determined by Hall measurements. Therefore, the carrier-concentration dependence of both the lattice and free-carrier responses is explicitly included in the radiative model, independently of the Raman measurements performed in this work.

The Raman spectra instead provide complementary evidence. The observed shift and broadening of the phonon-related Raman feature demonstrate that doping modifies the optical-phonon response of the actual GaAs wafers used in the NFRHT experiments. This observation supports an important part of our physical interpretation: doping is accompanied not only by a free-carrier response but also by a modification of the lattice response. The independently parameterized dielectric model then determines how these carrier-dependent material properties affect the surface-polariton dispersion, photon-transmission probability, spectral heat flux, and integrated heat flux. Thus, the Raman measurements and radiative calculations provide complementary, rather than interchangeable, evidence.

3. Influence of the redshift in the Raman spectra on NHRHT

The connection between the Raman response and the NFRHT spectrum is a sequential physical relationship rather than a direct correspondence between their spectral peaks. The physical relationship can be summarized as a progressive sequence: doping modifies the optical-phonon response; this modification changes the dielectric response of GaAs; the resulting dielectric response reshapes the surface-polaritonic modes; and these changes ultimately modify the evanescent transmission channels and the NFRHT spectrum. The Raman results provide experimental evidence for the first part of this physical sequence: carrier doping modifies the optical-phonon response of the actual GaAs wafers used in the heat-transfer experiments.

Nevertheless, the Raman peak and the NFRHT spectral feature should not be expected to exhibit a one-to-one correspondence. Raman spectroscopy predominantly probes zone-center longitudinal optical excitations, whereas NFRHT is governed by surface electromagnetic modes over a broad range of frequencies and in-plane wavevectors. Moreover, in doped GaAs, the surface modes are

determined jointly by the lattice and free-carrier contributions to the complete NFRHT spectral feature.

Raman spectroscopy nevertheless fills an important evidentiary role in the present study. Conventional descriptions of NFRHT in doped semiconductors often focus primarily on the additional Drude response and the resulting surface plasmon polaritons. Our Raman measurements demonstrate that, in polar GaAs, carrier doping also modifies the optical-phonon response of the experimental samples. Raman spectroscopy supplies evidence for the carrier-induced lattice modification underlying our material-level interpretation.

4. How the Raman evidence supports the central novelty of this work

The Raman measurements provide an important experimental link in establishing the central advance of this work. In many previous NFRHT studies involving doped semiconductors, the principal role of free carriers has been understood as introducing a Drude response and thereby enabling or tuning surface plasmon polaritons. When interactions between plasmonic and phononic polaritons are desired, they are often realized by combining distinct plasmonic and phonon-polaritonic materials, or by introducing engineered structures such as multilayer photonic crystals, heterostructures, or nanoscale resonators. In such systems, the polariton interaction is created or strengthened primarily through structural design.

The physical situation identified here is different. The Raman spectra show that carrier doping does not merely add a free-carrier response to GaAs; it is also accompanied by changes in the frequency and linewidth of the optical-phonon response of the same material. Because polar optical phonons generate the lattice dielectric response that supports surface phonon polaritons, modifying the optical-phonon resonance provides a direct material-level route for tailoring the corresponding surface phonon-polariton response. At the same time, the introduced free carriers support the plasmonic response. Consequently, carrier concentration simultaneously modifies both the carrier-related and phonon-related parts of the dielectric function, producing the coupled surface-polaritonic response within doped GaAs.

This is where Raman spectroscopy adds essential evidence to the present study. Without the Raman measurements, the effect of doping could be interpreted simply as the introduction of an additional Drude-type plasmonic response. The observed modification of the optical-phonon feature demonstrates that the lattice response of the actual GaAs samples also evolves with doping. The Raman and radiative results complete a consistent physical chain from carrier doping, through optical-phonon modification and surface-polariton tailoring, to the experimentally observed enhancement of NFRHT.

This intrinsic material-level approach does not require an additional photonic crystal, multilayer structure, or nanocavity to create the plasmon–phonon coupled response. Carrier concentration itself serves as the control parameter, while the system retains a simple planar and uniformly doped semiconductor configuration. This structural simplicity avoids additional resonator fabrication and interfacial complexity and is naturally compatible with established semiconductor doping and wafer-processing technologies.

Following the Reviewer’s comment, we have added the following clarification to the 3rd paragraph on the 4th page:

“The observed shift and broadening of Raman feature provide experimental evidence that introducing carriers modifies the longitudinal lattice response, rather than merely adding a free-carrier plasmonic response to an unchanged phononic background. These observations support the carrier–lattice interaction underlying the intrinsic plasmon–phonon self-hybridization mentioned in this work. Because the optical-phonon response contributes to the dielectric function, its interaction with free carriers reshapes the phonon-derived surface-polaritonic modes and consequently the thermal-photon tunneling spectrum. However, it is worth noting that the measured Raman peak positions cannot be used to fit the radiation heat transfer model. The dielectric parameters related to the carrier concentration used in the fluctuating electrodynamics calculation, including the TO and LO phonon frequencies, phonon damping constants, plasma frequency, and carrier relaxation rate, were obtained by fitting the Hall measurement (SM³³, Table S1). Therefore, Raman spectroscopy thus supports the material-level origin of the proposed mechanism, although it does not directly measure the surface modes responsible for NFRHT.”

References cited in this response

- [R1] A. Mooradian and G. B. Wright, “Observation of the Interaction of Plasmons with Longitudinal Optical Phonons in GaAs,” *Physical Review Letters* 16, 999–1001 (1966).
 - [R2] B. Tell and R. J. Martin, “Raman Scattering by Coupled Optical-Phonon-Plasmon Modes in GaAs,” *Physical Review* 167, 381–386 (1968).
 - [R3] H. R. Chandrasekhar and A. K. Ramdas, “Nonparabolicity of the Conduction Band and the Coupled Plasmon-Phonon Modes in n-GaAs,” *Physical Review B* 21, 1511–1515 (1980).
 - [R4] H. Takeuchi, T. Sumioka, and M. Nakayama, “Longitudinal Optical Phonon-Plasmon Coupled Mode in Undoped GaAs/n-Type GaAs Epitaxial Structures Observed by Raman Scattering and Terahertz Time-Domain Spectroscopic Measurements,” *IEEE Transactions on Terahertz Science and Technology* 7, 124–130 (2017).
 - [R5] J.-M. Lihm and C.-H. Park, “Plasmon-Phonon Hybridization in Doped Semiconductors from First Principles,” *Physical Review Letters* 133, 116402 (2024).
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Comment 2: There is no direct experimental evidence provided for this abstract claim: "a coupled plasmonic-phononic response arising from intrinsic carrier-lattice interactions, leading to a broadened spectral distribution and enhanced thermophotonic tunneling, rather than a simple superposition of independent plasmonic and phononic modes." The case of independent modes is not clearly presented. The Raman evidence is indirect and not clearly connected to the radiative transfer calculation and experiments.

Response 2: We thank the Reviewer for this important comment. We agree that the original statement went beyond what was directly established by the experiments. In particular, we did not experimentally isolate independent plasmonic and phononic modes, nor did we compare the measured response with the sum of two independent heat-transfer contributions. **Such an experimental separation is not physically available in the present material system. In uniformly doped GaAs, the free-carrier and polar-lattice responses coexist within the same material and jointly determine its total dielectric function.** On a physical level, it is extremely difficult to isolate the two components and observe their near-field radiative heat transfer capabilities separately.

We have therefore removed the phrase **"rather than a simple superposition of independent plasmonic and phononic modes"** from the abstract.

We also recognized that the original presentation of Fig. 4(e) may have contributed to the impression that the plasmonic and phononic contributions can be independently separated. This was not our intention. Figure 4(e) presents a theoretical comparison between the result obtained using the complete carrier-dependent dielectric function and a reference calculation in which the free-carrier Drude term is omitted. The increasing difference between these calculations with increasing carrier concentration indicates that the Drude response exerts an increasingly pronounced influence on the overall NFRHT response. This comparison does not provide a quantitative decomposition of the total heat flux into independent plasmonic and phononic contributions. We have added an explicit clarification to the discussion of Fig. 4(e) to prevent this interpretation:

"The comparison in Fig. 4(e) is intended only to illustrate that the influence of the free-carrier Drude response becomes increasingly pronounced with increasing carrier concentration. It should not be interpreted as a quantitative decomposition of the total heat flux into independent plasmonic and phononic contributions, because the carrier and lattice responses jointly determine the complete surface-polariton and near-field photon-transmission characteristics."

Comment 3: The experimental range of hot-side temperatures is limited ($<100\text{ }^{\circ}\text{C}$). It would be beneficial for both practical relevance and model validation to extend this range to above $300\text{ }^{\circ}\text{C}$.

Response 3: We thank the Reviewer for this constructive suggestion. We agree that extending the emitter temperature above 300 °C would be valuable for evaluating the high-temperature applicability of the proposed mechanism and for examining temperature-dependent effects that are not accessible within the present experimental range.

The temperature range used in this work was selected to establish the carrier-concentration dependence of NFRHT under well-controlled conditions. With the receiver maintained at 297 K and the maximum temperature difference reaching approximately 55 K, the emitter temperature reaches approximately 352 K, or 79 °C. **This near-room-temperature to moderate-temperature range is widely used in quantitative NFRHT experiments, particularly for macroscopic parallel surfaces.** For example, Ottens et al. performed measurements near 300 K with temperature differences of 2.5–30 K [R1]; Watjen et al. measured NFRHT between macroscopic doped-Si plates at average emitter and receiver temperatures of 318.5 and 302.3 K, respectively [R2]; and Ghashami et al. investigated phonon-polaritonic heat transfer between quartz plates at emitter and receiver temperatures of approximately 349 and 301 K [R3]. The temperature range of our experiments is therefore consistent with an established regime for quantitatively testing NFRHT while maintaining accurate control of a nanoscale gap. The present results therefore validate the carrier-dependent radiative model within the experimentally investigated temperature window. We agree, however, that this validation should not be extrapolated to temperatures above 300 °C without additional measurements and temperature-dependent optical parameters.

Increasing the temperature above 300 °C may introduce additional and potentially important physical behavior. In addition to increasing and redistributing the thermally excited photon population, elevated temperatures can modify the optical-phonon frequency and damping, carrier relaxation rate, and consequently the plasmonic–phononic response of GaAs. High-temperature NFRHT experiments in other material systems have already demonstrated that near-field heat transfer can exhibit material- and gap-dependent temperature scaling that differs substantially from the conventional far-field ($Q \propto T^4$) dependence [R4]. It would therefore be particularly interesting to determine whether the carrier-induced enhancement and spectral broadening observed here remain unchanged, become stronger, or weaken at elevated temperatures.

The present apparatus, however, was designed and calibrated for emitter temperatures below 100 °C and cannot yet provide reliable measurements above 300 °C. For our macroscopic wafer configuration, simply increasing the heater set point would not be sufficient. **Differential thermal expansion and temperature-induced wafer deformation could change the size of vacuum gap. This thermal-induced uncertainty can be very serious.** The thermal conductance of the supporting nanopillars, interfacial thermal resistances, and sensor calibrations would also become temperature

dependent. Without high-temperature-compatible components and reliable characterization of the gap at the operating temperature, it would be difficult to distinguish an intrinsic temperature-dependent radiative effect from a thermally induced change in the gap or parasitic heat transfer.

Room-temperature and extreme-temperature NFRHT measurements are consequently treated as distinct experimental stages requiring different platform designs. From measurements taken at near-room temperature to specially designed experiments conducted under extreme temperature conditions, this phased approach is very common in near-field radiative heat transfer studies. For example, Song et al. first established quantitative near-field radiative heat-transfer measurements between nanoscale-separated dielectric and metallic plates at room temperature [R5]. Members of the same research team subsequently developed a dedicated near-field thermophotovoltaic platform operating at emitter temperatures of 525–655 K (approximately 252–382 °C) [R6]. The same broader research team later constructed a cryogenic platform to investigate surface-phonon-polariton-mediated NFRHT over 77–300 K [R7]. Importantly, these high- and low-temperature studies did not merely increase or decrease the temperature set point of the original room-temperature apparatus. They employed purpose-built device geometries, thermal calibration procedures, and gap-control strategies appropriate to the corresponding temperature range. These studies demonstrate that first establishing a reliable near-room-temperature baseline and subsequently extending the measurement to an extreme-temperature regime using an upgraded platform is a well-founded experimental progression. We follow the same staged approach: the present work establishes the carrier-dependent NFRHT response under accurately controlled moderate-temperature conditions, while an upgraded platform will be required to investigate the additional temperature-dependent behavior above 300 °C.

Following the Reviewer’s suggestion, we have added the following perspective to the Conclusion:

“The present measurements establish carrier-dependent NFRHT enhancement in a near-room-temperature to moderate-temperature regime. Extending the emitter temperature above 600 K represents an important direction for future investigation. At elevated temperatures, changes in optical-phonon frequency and damping, carrier relaxation, and the thermally excited spectral range may further reshape the plasmonic–phononic response and the resulting near-field heat transfer.”

References cited in this response

- [R1] R. S. Ottens et al., “Near-Field Radiative Heat Transfer between Macroscopic Planar Surfaces,” *Physical Review Letters* 107, 014301 (2011).
- [R2] J. I. Watjen, B. Zhao, and Z. M. Zhang, “Near-field radiative heat transfer between doped-Si parallel plates separated by a spacing down to 200 nm,” *Applied Physics Letters* 109, 203112 (2016).

- [R3] M. Ghashami et al., “Precision Measurement of Phonon-Polaritonic Near-Field Energy Transfer between Macroscale Planar Structures under Large Thermal Gradients,” *Physical Review Letters* 120, 175901 (2018).
- [R4] C. Lucchesi, R. Vaillon, and P.-O. Chapuis, “Temperature Dependence of Near-Field Radiative Heat Transfer above Room Temperature,” *Materials Today Physics* 21, 100562 (2021).
- [R5] B. Song, D. Thompson, A. Fiorino, Y. Ganjeh, P. Reddy, and E. Meyhofer, “Radiative Heat Conductances between Dielectric and Metallic Parallel Plates with Nanoscale Gaps,” *Nature Nanotechnology* 11, 509–514 (2016).
- [R6] A. Fiorino, L. Zhu, D. Thompson, R. Mittapally, P. Reddy, and E. Meyhofer, “Nanogap Near-Field Thermophotovoltaics,” *Nature Nanotechnology* 13, 806–811 (2018).
- [R7] S. Yan, Y. Luan, J. W. Lim, et al., “Surface Phonon Polariton-Mediated Near-Field Radiative Heat Transfer at Cryogenic Temperatures,” *Physical Review Letters* 131, 196302 (2023).
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Comment 4: Fig. 1a is confusing. It shows lateral temperature gradients which is not aligned with the assertion of "negligible lateral heat leakage". It's also not clear how exactly Fig. 1a corresponds to Fig. 2b.

Response 4: We thank the Reviewer for pointing out this ambiguity. Figure 1(a) was intended as a conceptual illustration of a nanoscale interfacial void in a stacked-chip architecture. This color variation is merely an artistic embellishment in the schematic and is not intended to reflect the lateral temperature distribution in the experimental sample. The statement of negligible lateral heat leakage refers exclusively to the apparatus in Fig. 1(b): agreement between the heat-flux sensors on the emitter and receiver sides confirms approximately one-dimensional heat transport through the experimental stack.

There is no one-to-one geometrical correspondence between Fig. 1(a) and Fig. 2(b). Figure 1(a) shows the application motivation, Fig. 1(b) shows the actual measurement configuration, and Fig. 2(b) reports the heat-flux data for the GaAs pair. We have removed the lateral color gradient from Fig. 1(a).

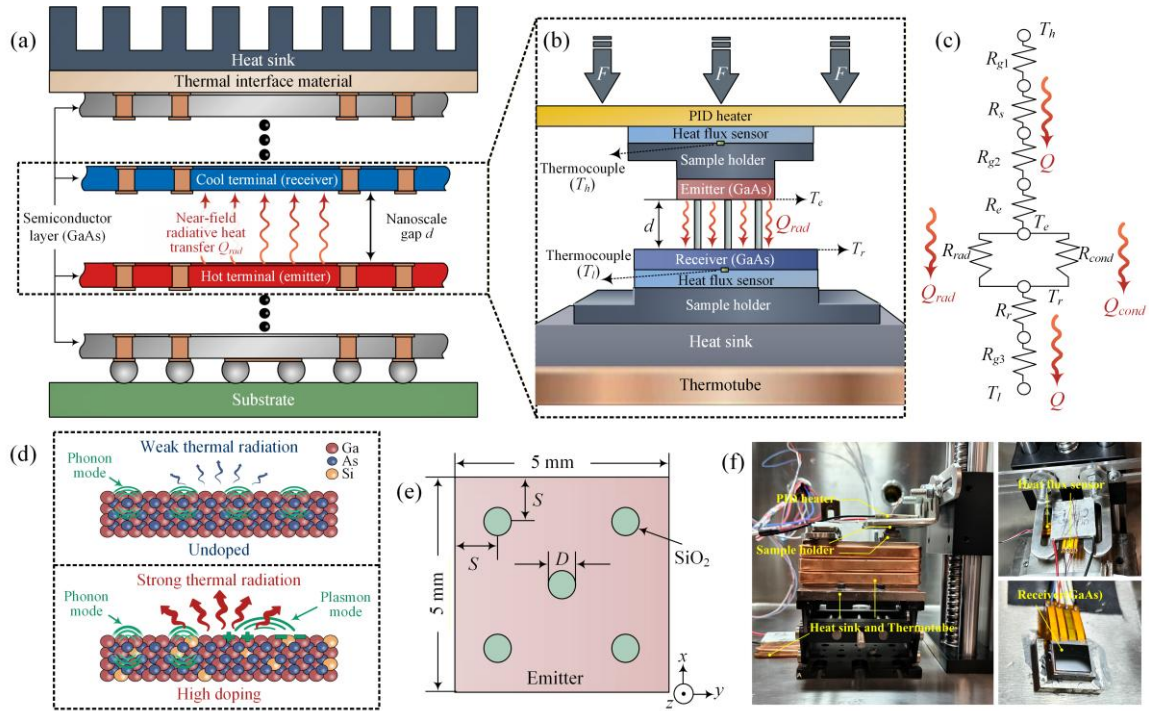


Fig 1 in Main text

Comment 5: This sentence should be revised for clarity: "A high emitter temperature is established using a PID-controlled heater."

Response 5: We agree and have revised the sentence for clarity. Revision in the manuscript: **"The emitter temperature is controlled and maintained at the prescribed value using a PID-regulated heater."**

Comment 6: "strengthening behavior" is used incorrectly to describe heat flux.

Response 6: We agree. The phrase "strengthening behavior" has been replaced with the more precise expression "heat-flux enhancement." Revision in the manuscript: **"The calculated spectral heat flux further elucidates the origin of this heat-flux enhancement [Figs. 3(c-e)]."**

Comment 7: Regarding "theoretical uncertainty bands (shaded regions) explicitly account for deviations in receiver temperature and structural variations (actual nanopillar height and macroscopic sample deflection)." I'm curious how these variations are measured and accounted for. Please provide detailed information.

Response 7: We thank the Reviewer for requesting these details. The shaded regions are not arbitrary fitting intervals. **Their inputs are derived from independent temperature and structural**

measurements, and the fluctuational-electrodynamics calculation is then proceeded at the corresponding conditions.

First, the unloaded SiO₂ nanopillar geometry was measured by atomic force microscopy before and after the NFRHT experiments. The nanopillars have an average height of 159.50 nm and an average diameter of 2.72 μm . No damage or plastic deformation was observed after measurement. The maximum elastic compression under the applied load was estimated from the measured geometry and a room-temperature SiO₂ Young's modulus of 76.6 GPa, giving a conservative maximum compression of 6.44 nm [R1].

Second, the macroscopic surface profiles of the GaAs wafers were measured over a $5 \times 5 \text{ mm}^2$ area using an RTEC UP-LAMBDA-U-3000 white-light interferometer. Profiles along two orthogonal lines through the center of each wafer were used to determine the upper and lower deflection limits. Combining these limits with the nanopillar height yields the sample-specific gap estimates reported in the manuscript: 159 nm (+38/-19 nm), 159 nm (+30/-25 nm), and 159 nm (+32/-30 nm) for the undoped, moderately doped, and highly doped configurations, respectively [R1].

Third, the temperature recorded during the measurements remained between 23 and 25 °C, i.e., $297 \pm 1 \text{ K}$. For each experimental temperature difference, the upper theoretical heat-flux boundary was calculated using the smaller limiting gap and a receiver temperature of 25 °C, whereas the lower boundary was calculated using the larger limiting gap and a receiver temperature of 23 °C [R2]. Based on these measured data, we can roughly determine the upper and lower limits for temperature and spacing; using these parameters, we can then calculate the upper and lower limits for theoretical near-field radiative heat transfer.

We have also clarified the distinction between the shaded theoretical bands and the experimental error bars. The bands reflect uncertainty in the model inputs described above. The error bars reflect the repeatability of the measured heat flux, including the heat-flux sensor and thermocouple uncertainties and the variation among repeated steady-state measurements. Our previous description may not have been entirely clear, so we have added a summary in S11 page of the supplementary material to help readers understand the source of the uncertainty more clearly and intuitively:

“In conclusion, the unloaded nanopillar height and diameter were measured by AFM to be 159.50 nm and 2.72 μm on average, respectively. The maximum elastic compression under the applied load was calculated to be 6.44 nm. Macroscopic wafer profiles were independently measured over a $5 \times 5 \text{ mm}^2$ area using white-light interferometry, and the measured deflection limits were combined with the nanopillar height to determine the minimum and maximum possible vacuum

gaps for each sample pair. The receiver temperature varied between 23 and 25 °C during measurement. The upper and lower theoretical heat-flux curves were obtained by evaluating the fluctuational-electrodynamics model using the corresponding limiting combinations of gap distance and receiver temperature. Thus, the shaded bands in Fig. 2 represent the prediction range arising from experimentally determined gap and temperature variations, whereas the experimental error bars describe the repeatability and instrumental uncertainty of the heat-flux measurements.”

References cited in this response

[R1] L. Tang, J. DeSutter, and M. Francoeur, “Near-field radiative heat transfer between dissimilar materials mediated by coupled surface phonon- and plasmon-polaritons,” ACS Photonics 7, 1304–1311 (2020).

[R2] J. DeSutter, L. Tang, and M. Francoeur, “A near-field radiative heat transfer device,” Nat. Nanotechnol. 14, 751–755 (2019).

Comment 8: Thermophotonic is a type of conversion device. The term should not be used interchangeably with "thermal photons".

Response 8: We agree with the Reviewer. In the original manuscript, "thermophotonic tunneling" and "thermophotonic tunneling probability" were used to describe the transmission of thermally excited electromagnetic modes. This terminology can be confused with thermophotonic energy-conversion devices and is inappropriate in the present context.

We have systematically replaced the terminology throughout the Abstract, main text, figure captions, and Supplementary Material. We now use "**thermal photons**" as a qualitative physical description.

We would like to thank the Reviewer’s review of the article and careful reading of our manuscript and many constructive comments. We think that the implementation of these suggestions has resulted in great improvements to the present paper.