

Response to comments from the Reviewer #2

General Response: We sincerely thank the Reviewer for the careful evaluation of our manuscript and for the thoughtful and constructive comments. We greatly appreciate the time and expertise devoted to this review. The Reviewer's suggestions have helped us improve the clarity, rigor, and presentation of the manuscript. We have carefully considered each comment, and our detailed responses are provided below.

Comment 1: In the work, the authors present experiments of near-field radiative heat transfer between doped GaAs surfaces. Experiments are conducted for three doping levels, including undoped, moderately doped, and highly doped. The heat transfer rate is shown to increase when the doping level rises. Fluctuational-electrodynamics calculations agree with the experimental measurements. Raman spectroscopy is conducted. A key concern is novelty. Near-field heat transfer between doped semiconductors has been well studied experimentally. It is known that by introducing doping, the surface plasmon polaritons can enhance the radiative heat transfer. Due to that, the study may not meet the stringent requirement of Light: Science & Applications. The study could be better suited for a specialized journal.

Response 1: We thank the Reviewer for raising this important concern regarding the novelty and broader significance of our work. We agree that free-carrier surface plasmon polaritons have previously been used to enhance NFRHT in doped semiconductors. **However, the central advance of our work is not the use of doping itself, nor is it simply the introduction of an additional plasmonic channel. Rather, we uncover a fundamentally different mechanism of carrier-controlled NFRHT in a polar semiconductor: doping modifies the optical-lattice response of GaAs and induces plasmon-phonon hybridization, thereby reorganizing the surface polaritonic modes that govern near-field energy transfer.** This mechanism is qualitatively distinct from the conventional picture of doping-controlled NFRHT in nonpolar semiconductors, where carrier injection primarily modifies the free-carrier Drude response. Importantly, it also provides intrinsic, material-level control of polaritonic hybridization without relying on artificially patterned micro/nanostructures, which are commonly required to engineer hybrid polaritonic modes. Therefore, the novelty of our study lies in revealing and experimentally establishing a distinct carrier-lattice route for regulating NFRHT in polar semiconductors. Below, we clarify this distinction and its broader significance in detail.

1. Why doping polar GaAs is physically different from the conventional doping of Si

We agree that previous experiments have established that doping Si introduces a free-carrier Drude response, changes the plasma frequency, and regulates SPP-mediated NFRHT [R1, R2]. However, this conventional mechanism does not fully describe the physical response of doped GaAs.

Crystalline Si is nonpolar and does not possess an infrared-active polar optical-phonon response that supports conventional surface phonon polaritons. Its doping-dependent NFRHT is therefore dominated by changes in the free-carrier response. GaAs, in contrast, is a polar semiconductor. Its transverse and longitudinal optical lattice vibrations generate a strong infrared dielectric response and support phonon-related surface-polariton modes before the free-carrier population is introduced.

When GaAs is doped, the collective oscillations of the free carriers interact with the polarization associated with the polar optical lattice vibrations through the long-range Coulomb interaction [R3]. Consequently, doping in GaAs has two interconnected effects: it introduces a free-carrier response and modifies the longitudinal optical-lattice response. The resulting NFRHT is determined by the complete surface plasmon-phonon hybridization effect, rather than by the contribution of independently added plasmonic heat transfer, as in previous studies [R1, R2]. This distinction is supported by the complementary measurements and calculations in our study. Hall measurements determine the carrier concentrations of the GaAs wafers. Raman spectroscopy shows that the position and linewidth of the LO-related longitudinal response in the actual samples change with doping, providing experimental evidence that the lattice response is not unchanged after carriers are introduced.

As experts, the Reviewer keenly observed that we, like previous studies, also utilized doping as a technique. However, we believe that doping is merely a means of control; innovation depends on what new physical responses it triggers in different materials. We think it might not be appropriate to rely solely on the use of doping to determine whether we are suitable for *Light: Science & Applications*. Xie et al. previously reported a conceptually related innovation in *Light: Science & Applications* [R4]. As is well known, using doping to modulate surface plasmonic polaritons has become quite common. However, in that study, the researchers used Mo-doping to modulate the intrinsic optical topological phase transition of plasmonic polaritons in native WTe₂—thereby avoiding the artificial nanostructures traditionally required to realize and control hyperbolic polaritons. Although the observables and underlying modes differ between the two studies, both illustrate how an established material-control method can reveal a new polaritonic regulation mechanism and open a distinct research direction.

2. Why previous studies using doping do not preclude the novelty of the present work

We respectfully consider that the novelty of a study should be evaluated according to the new physical question, mechanism, or capability revealed by a control parameter, rather than according to whether that control parameter has previously been used. Doping is a general method of modifying matter, and its physical consequences depend strongly on the electronic structure, lattice polarity, dimensionality, and carrier–phonon interactions of the material.

The development of NFRHT itself illustrates that previous investigation of a material or control parameter does not exhaust its scientific potential. Shen et al. first experimentally established surface-phonon-polariton-mediated energy transfer across nanoscale gaps between polar dielectrics [R5]. Subsequently, by treating film thickness as a control parameter, Song et al. showed that reducing a polar dielectric to a thin film reorganizes its supported polaritonic modes and produces near-field heat-transfer behavior distinct from that of the corresponding bulk material [R6]. More recently, Tang et al. revisited the well-established SiC system in a finite-size configuration and discovered that electromagnetic corner and edge modes can dominate NFRHT between subwavelength SiC membranes [R7]. These studies remained scientifically important not because the materials or surface phonon polaritons were newly introduced, but because continued investigation of familiar systems under different dimensional and geometric conditions revealed previously unexplored modes and heat-transfer regimes. Similarly, although semiconductor doping is an established material-control method, using carrier concentration to regulate intrinsic plasmon–phonon self-hybridization and the resulting NFRHT in a polar semiconductor constitutes a distinct physical question.

Related studies published in *Light: Science & Applications* provide useful context for the type of advance pursued here. Xie et al. used Mo doping to tune the intrinsic optical topological transition of naturally hyperbolic plasmon polaritons in WTe₂ [R4]. Zhang et al. combined excitation-dependent photoluminescence with Raman measurements of LO-phonon–plasmon coupled modes to determine key carrier and recombination parameters in GaAs that had conventionally required electrical characterization [R8]. In both studies, the methods themselves were established, but their application enabled capabilities that had not previously been demonstrated: intrinsic control of a polariton transition in one case and all-optical determination of semiconductor recombination parameters in the other. For the same reason, the previous experiments on doped semiconductors did not imply that all subsequent studies involving doping were physically equivalent. In this research, the control methods are familiar, but the material responses being regulated, the carrier-lattice physical mechanisms involved, and the resulting near-field spectral behaviors are all different.

3. Why carrier-controlled NFRHT in polar semiconductors is important for future semiconductor technologies

At intact solid–solid interfaces, heat conduction mediated by phonons and, where applicable, electrons is generally much more efficient than radiative heat transfer. This disparity has historically limited the practical role of NFRHT and raises an important question for the field: **under what conditions can its non-contact nature provide a unique advantage for thermal management?** Three-dimensional integration and wafer-level stacking offer a potentially important application scenario. Although these technologies increase device density and system performance, they also introduce multiple active layers and complex bonding interfaces, making interlayer heat dissipation a major challenge for next-generation integrated systems. Practical wafer-bonding interfaces may contain interfacial voids, micro-delamination, and locally unbonded regions caused by surface roughness, residual reaction products, particles, or insufficient planarity [R9].

Direct solid-state conduction is interrupted within these non-contact regions. When the local separation is sufficiently small, near-field radiative heat transfer may provide an additional energy-transfer pathway across the gap, partially compensating for the locally disrupted conductive transport. Meanwhile, semiconductor technology is expanding beyond conventional Si-dominated platforms toward heterogeneous systems incorporating III–V compounds, wide-bandgap semiconductors, and other functional materials [R10, R11], many of which are polar and possess strong infrared lattice responses. These developments lead to a fundamental and technologically relevant question: **how can NFRHT across nanoscale non-contact regions between doped polar-semiconductor layers be effectively controlled?** Addressing this question could establish a new role for NFRHT in the thermal management of heterogeneous 3D integrated systems and provide an important direction for the future development of near-field thermal-radiation technology.

For polar materials, a widely used route to controlling NFRHT is to engineer polaritonic modes through heterojunction [R12], multilayers [R13], or metasurfaces [R14]. Although effective, these approaches introduce additional micro/nanofabrication and can be difficult to integrate into compact, large-area stacked interfaces while maintaining planar surfaces and uniform nanoscale gaps. This raises a distinct question: **can carriers themselves be used to control NFRHT in polar semiconductors directly at the material level?**

Our work provides an initial experimental answer by demonstrating carrier-controlled NFRHT between GaAs wafers without patterning the heat-exchanging surfaces. Although the present experiment is not a complete chip-level thermal-management solution, it establishes a material-level approach for regulating radiative energy transfer across nanoscale gaps. More broadly, it connects NFRHT with the emerging thermal challenges of heterogeneous three-dimensional

integration and opens an application-driven research direction involving other doped polar semiconductors, bonded interfaces, and vertically integrated devices.

Revision to the original manuscript

Following the Reviewer's comment, we have revised the manuscript to state this novelty more precisely and to avoid presenting generic carrier-induced SPPs enhancement as our principal contribution. The relevant passages in the 3rd paragraph on the 3rd page as follows:

“Carrier doping has been widely employed to regulate NFRHT in nonpolar semiconductors such as Si, where its principal role is commonly understood as introducing or tuning a Drude-type free-carrier response and the associated surface plasmon polaritons. Polar semiconductors present a different physical situation. In GaAs, mobile carriers coexist and interact with infrared-active polar optical phonons. Consequently, changing the carrier concentration modifies not only the plasmonic response but also the optical-phonon-related dielectric response, thereby reshaping the coupled surface-polariton spectrum governing NFRHT. The scientific question addressed here is therefore not whether conventional doping-induced SPPs can enhance NFRHT, but whether carrier concentration can serve as an intrinsic material-level parameter for controlling the plasmon–phonon response of a polar semiconductor without relying on artificial photonic structures.”

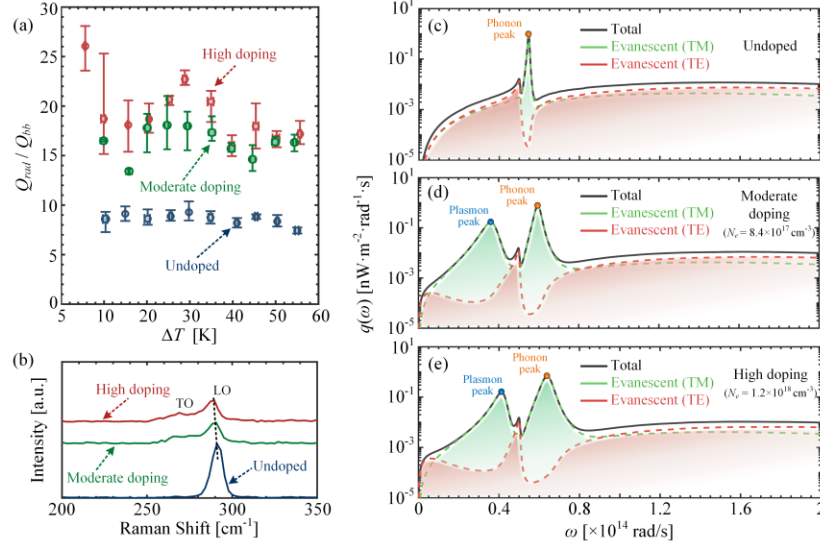
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- [R2] J. DeSutter, L. Tang, and M. Francoeur, “A near-field radiative heat transfer device,” *Nature Nanotechnology* **14**, 751–755 (2019).
- [R3] J.-M. Lihm and C.-H. Park, “Plasmon–Phonon Hybridization in Doped Semiconductors from First Principles,” *Physical Review Letters* **133**, 116402 (2024).
- [R4] Y. Xie, C. Wang, F. Fei, *et al.*, “Tunable optical topological transitions of plasmon polaritons in WTe₂ van der Waals films,” *Light: Science & Applications* **12**, 193 (2023).
- [R5] S. Shen, A. Narayanaswamy, and G. Chen, “Surface phonon polaritons mediated energy transfer between nanoscale gaps,” *Nano Letters* **9**, 2909–2913 (2009).
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- [R13] M. Lim, J. Song, S. S. Lee, and B. J. Lee, “Tailoring near-field thermal radiation between metallo-dielectric multilayers using coupled surface plasmon polaritons,” *Nature Communications* **9**, 4302 (2018).
- [R14] E. J. C. Dias, R. Yu, and F. J. García de Abajo, “Thermal manipulation of plasmons in atomically thin films,” *Light: Science & Applications* **9**, 87 (2020).
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Comment 2: For Fig. 3a, Q_{rad}/Q_{bb} as a function of ΔT has a peak at ~ 30 K, and another peak at small ΔT . Are these peaks due to measurement uncertainty, or are physical feature that agrees with modeling?

Response 2: We thank the Reviewer for drawing attention to these variations. The apparent local maxima in Fig. 3(a), including the point near $\Delta T \sim 30$ K and the variation at small ΔT , are not interpreted as distinct physical features. These variations primarily arise from experimental uncertainty and its amplification by the normalization Q_{rad}/Q_{bb} . When the temperature difference is small, the blackbody reference value is relatively small. Therefore, slight fluctuations in uncertainty can result in significant fluctuations in the ratio, even though the actual fluctuations in heat flux are not that large. The dashed lines connecting the experimental points in the original figure may also have inadvertently suggested that these point-to-point variations represent continuous physical peaks. Therefore, we revised Fig. 3(a) to present the measurement results as discrete data points.



The revised version of Figure 3 in the main text

In addition, we explicitly stated in the figure caption that the local fluctuations in the experimental ratios are attributable to measurement uncertainties. The revised caption reads:

“It is noticed that the local fluctuations in the experimental ratios are attributable to measurement uncertainties.”

Comment 3: The term “intrinsic” before “carrier-lattice interactions” is unclear. Discussion should be provided to explain why it is intrinsic.

Response 3: We thank the Reviewer for identifying this ambiguity. Here, “intrinsic” does not mean that the GaAs is electrically intrinsic or undoped. **It refers to the material-level origin of the interaction.**

In doped GaAs, collective free-carrier oscillations couple through the long-range Coulomb interaction to the polarization associated with polar optical lattice vibrations [R1, R2]. Both degrees of freedom reside within the same homogeneous semiconductor. Their interaction therefore does not require an externally added heterostructure, photonic crystal, multilayer, or resonant nanostructure. Changing the carrier concentration modifies the plasma frequency and its spectral relationship with the optical phonons, thereby generating interaction between plasmons and phonon polaritons.

To prevent confusion, we have replaced “intrinsic” with the more specific term “material-intrinsic” at its first occurrence and added the following explanation in 2nd paragraph on the 7th page:

“It is noticed that, here, ‘material-intrinsic’ denotes that the interacting free-carrier and polar-lattice excitations coexist within uniformly doped GaAs and couple through their internal long-range

Coulomb fields. Carrier concentration therefore regulates the coupled dielectric response at the material level. This differs from hybridization implemented by combining distinct materials or fabricating resonant structures, as exemplified by graphene/h-BN heterostructures³⁹, metal–dielectric multilayers⁵¹, and patterned resonators⁵². Here, different resonance modes reside in the same semiconductor, allowing the polaritonic spectrum to be tailored without additional heterostructure assembly or resonator patterning. This structural simplicity, together with compatibility with semiconductor doping and planar processing, offers a complementary route to broadband NFRHT enhancement.”

We have also revised the subsection title to “Material-intrinsic plasmon–phonon coupling mechanism of NFRHT enhancement.”

References cited in this response

- [R1] B. B. Varga, “Coupling of Plasmons to Polar Phonons in Degenerate Semiconductors,” *Physical Review* 137, A1896–A1902 (1965).
- [R2] 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 4: How the results of Raman spectroscopy can be used to support the claim of study is unclear. The Raman spectroscopy results seem tangential to near-field heat transfer. More explanation is needed.

Response 4: We thank the Reviewer for this important comment. We agree that the original manuscript did not clearly distinguish the evidence provided by Raman spectroscopy from that provided by the radiative-transfer calculations and heat-flux measurements. We have therefore clarified the role of Raman spectroscopy and its connection to NFRHT.

Raman spectroscopy is a method for detecting carrier-induced changes in the longitudinal optical response of GaAs. In this polar semiconductor, the macroscopic electric field associated with longitudinal optical lattice vibrations interacts with collective free-carrier oscillations through the long-range Coulomb interaction. This interaction can modify the frequency, linewidth, intensity, and line shape of the Raman spectroscopy [R1–R3]. This further supports the core physical phenomenon discussed in this paper—namely, that carriers alter the properties of optical phonons, giving rise to plasma-phonon hybrid polaritons—which differs from traditional research that introduces plasmonic polaritons solely through carriers.

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.

We also emphasize, however, that Raman spectroscopy is not a direct measurement of either the evanescent surface modes or the NFRHT spectrum. Raman spectroscopy predominantly probes near-zone-center longitudinal excitations, whereas NFRHT is governed by surface electromagnetic modes spanning broad ranges of frequency and in-plane wavevector. The measured Raman peak positions were also not inserted into or used to fit the radiative calculations. Instead, the dielectric function employed in the fluctuational-electrodynamics model was constructed using independently reported carrier-concentration-dependent optical parameters for n-type GaAs. These include the TO and LO phonon frequencies, phonon damping constant, plasma frequency, and carrier relaxation rate, as summarized in Table S1 of the Supplementary Material. These parameters were interpolated to the carrier concentrations of our samples, which were independently determined by Hall measurements. The Raman measurements and radiative calculations therefore provide independent and complementary evidence rather than constituting a parameter-fitting procedure.

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.

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 (SM^{33} , 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).
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- [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).
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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.