Near-Field Radiative Heat Transfer Modulation with an Ultrahigh Dynamic Range through Mode Mismatching

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ABSTRACT: Modulating near-field radiative heat transfer (NFRHT) with a high dynamic range is challenging in nanoscale thermal science and engineering. Modulation depths [(maximum value − minimum value)/(maximum value + minimum value) × 100%] of ≈2% to ≈15.7% have been reported with matched modes, but breaking the constraint of mode matching theoretically allows for higher modulation depth. We demonstrate a modulation depth of ≈32.2% by a pair of graphene-covered SU8 heterostructures at a gap distance of ≈80 nm. Dissimilar Fermi levels tuned by bias voltages enable mismatched surface plasmon polaritons which improves the modulation. The modulation depth when switching from a matched mode to a mismatched mode is ≈4.4-fold compared to that when switching between matched modes. This work shows the importance of symmetry in polariton-mediated NFRHT and represents the largest modulation depth to date in a two-body system with fixed gap distance and temperature.

KEYWORDS: modulation of near-field radiative heat transfer, graphene, surface plasmon polaritons coupling, mode mismatching, fluctuational electrodynamics, radiative heat transfer measurement

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hermal radiation, with spectral and power density properties described by Planck’s Law and the Stefan–Boltzmann Law, is one of the known noncontact heat transfer modes in vacuum.\(^1,2\) Classical physics predicts that a perfect thermal emitter operates at the blackbody (BB) limit. Fluctuational electrodynamics has demonstrated that evanescent modes, including plasmon and phonon polaritons, allow large near-field radiative heat transfer (NFRHT) far beyond the limit.\(^3\)–\(^14\) Compared to the general far-field broadband radiative spectrum, NFRHT is primarily dominated by the resonance coupling modes between two close objects, allowing for active control of the spectrum\(^11,15\) and energy transfer.\(^6,18\)

Dynamic modulation of NFRHT requires changing the optical responses of the emitter and receiver to modulate the radiative heat flux. Previous works\(^17\)–\(^20\) have shown that a phase-change material (VO\(_2\)) allows for thermal radiation modulation due to its insulator-to-metal transition. However, the modulation requires temperature variation. Graphene has highly tunable surface plasmon polaritons (SPPs) related to its Fermi level in a linear Dirac band, which makes it an ideal thermal modulator with external stimuli at a fixed operating temperature.\(^21\)–\(^23\) Altering the free carrier states of a van der Waals’ heterostructure also allows for exotic nanoscale phenomena such as tunable Mott insulator,\(^24\) nanoimaging in an infrared-waveguide,\(^25\) and efficient Fizeau drag from Dirac electrons,\(^26\) etc. Recent experimental works have demonstrated that graphene plasmons enable giant radiative heat transfer at the nanoscale.\(^5,6,12\) Graphene with different Fermi levels supports tunable SPPs and accounts for different radiative heat flux. A high dynamic range modulation of NFRHT allows for a high signal-to-noise ratio (SNR) enabling potential applications in thermal switches and communication. Thomas et al. have shown an electronic modulation depth of ≈2% [defined by (maximum value−minimum value)/(maximum value + minimum value) × 100%] with a pair of graphene sheets on Al\(_2\)O\(_3\)/SiO\(_2\) substrate.\(^21\) The Fermi levels of the two graphene sheets are assumed to be equal since the samples are in conductive contact. Our recent work has shown that the modulation depth could reach ≈15.7% with stacked graphene layers at similar Fermi levels.\(^6\) However, the modulation effect is limited by the matched resonance modes between the identical graphene sheets, as the graphene on the emitter and receiver have similar Fermi levels.

Here, we present a significant modulation improvement by a pair of graphene/SU8 heterostructures at a gap distance of ≈80 nm. Back-gated tuning was employed for the graphene
Fermi level control. The mismatched SPPs due to different Fermi levels of the two graphene sheets allow for a much smaller heat flux compared to the matched SPPs case. The measured maximum modulation depth could reach ≈32.2% and is ≈4.4-fold compared to that of the matched case. This experimental work represents the largest modulation depth ever reported for the radiative heat transfer in a two-body planar system with a fixed gap distance and operating temperature. The mode-mismatch-induced high-efficiency NFRHT modulation should inspire potential applications of thermal switches,\textsuperscript{25,27,28} thermal communication, and is suitable for tunable plasmon- or phonon-induced thermal or photonic regulation.

We study the NFRHT between a pair of graphene-covered SU8 heterostructures on SiO\textsubscript{2}/Si substrates (marked as Gr/SU8, \textbf{Figure 1}). The thickness of the SU8 is chosen to be 90 nm to reduce the influence of phonon polaritons from the substrate. Based on the fluctuational electrodynamics, the net radiative heat flux between the emitter and receiver (with temperatures \(T_1\) and \(T_2\), respectively) at a gap distance \(d\) is calculated by\textsuperscript{29–33}

\[
\begin{align*}
Q &= \frac{1}{4\pi^2} \int_0^\infty \left\{ \Theta(T_1, \omega) - \Theta(T_2, \omega) \right\} \\
&\times \left[ \int_{k_0}^{k_s} d\beta \sum_{j=s,p} \xi_j(\omega, \beta, d) \right] d\omega \\
&\times d\beta \\
&= \frac{1}{4\pi^2} \int_0^\infty \left\{ \Theta(T_1, \omega) - \Theta(T_2, \omega) \right\} \\
&\times \left[ \int_{k_0}^{k_s} d\beta \sum_{j=s,p} \xi_j(\omega, \beta, d) \right] d\omega \\
&\times d\beta \\
&\times d\beta.
\end{align*}
\]

where \(\Theta(T_1, \omega) = \frac{\hbar\omega}{\exp(\hbar\omega/k_B T) - 1}\) is the mean energy of the Planck thermal harmonic oscillators without zero point energy. \(\hbar\) is the reduced Planck constant and \(k_B\) is the Boltzmann constant. \(\xi_j\) represents the energy transmission coefficient between the emitter and receiver (considering both \(s\)- and \(p\)-polarization modes):

\[
\xi_j(\omega, \beta) = \begin{cases} 
\frac{(1 - |r_{j,s}|^2)(1 - |r_{j,p}|^2)}{1 - r_{j,s}r_{j,p}e^{2ik_d\beta}}, & \beta < k_0 \\
4\text{Im}(r_{j,s})\text{Im}(r_{j,p})e^{2ik_d\beta}, & \beta > k_0 
\end{cases}
\]

(2)

where \(k_0\) is the z-component of the wave vector in vacuum \((k_0)\). \(r_{j,s}\) and \(r_{j,p}\) are the Fresnel reflection coefficients of the emitter and receiver, respectively. \(\xi_{sp}\) with \(\beta > k_0\) represents the photon tunneling probability of the \(p\)-polarized evanescent modes.

For a homogeneous medium with finite thickness, the reflection coefficients become\textsuperscript{39,32}

\[
\begin{align*}
&\begin{cases} 
&\begin{align*}
&1 + r_{j,12}(1 + r_{j,01} + r_{j,10})e^{2ik_1h_j} \\
&1 - r_{j,10}r_{j,12}e^{2ik_1h_j}
\end{align*}
\end{cases}, \ j = s \\
&\begin{cases} 
&\begin{align*}
&1 + r_{j,12}(1 - r_{j,01} - r_{j,10})e^{2ik_1h_j} \\
&1 - r_{j,10}r_{j,12}e^{2ik_1h_j}
\end{align*}
\end{cases}, \ j = p
\end{align*}
\]

(3)

where \(h_j\) is the thickness of layer one and \(r_{j,01}\) is the Fresnel reflection coefficient at the interface between layer 0 and layer 1 for \(s\)- or \(p\)-polarization modes:

\[
\begin{align*}
&\begin{cases} 
&\begin{align*}
&k_z^{(0)} - k_z^{(1)} - \sigma p,\omega \ \ \ \ \ \ \ j = s \\
&k_z^{(0)} + k_z^{(1)} + \sigma p,\omega
\end{align*}
\end{cases} \\
&\begin{cases} 
&\begin{align*}
&k_z^{(1)} - k_z^{(0)} + \alpha p,\omega k_z^{(0)} + \frac{\alpha p,\omega k_z^{(0)}}{\varepsilon_{0,\omega}} \ \ \ \ \ \ \ j = p
\end{align*}
\end{cases}
\end{align*}
\]

(4)

where \(k_z^{(n)} = \left(\frac{\varepsilon_z^{(n)}k_0^2 - \varepsilon_z^{(0)}\beta^2}{\varepsilon_z^{(0)}}\right)^{1/2}\), \(k_z^{(n)} = (\varepsilon_z^{(n)}k_0^2 - \beta^2)^{1/2}\), \(n = 0, 1, 2\) is the number of the layers, \(\varepsilon_z^{(n)}\) and \(\varepsilon_z^{(0)}\) are the perpendicular and parallel components of the relative dielectric tensor. Here, \(\varepsilon_z^{(n)} = \varepsilon_z^{(0)}\) is set for the isotropic medium. Graphene was treated as surface current with complex conductivity \(\sigma\):\textsuperscript{34,35}

\[
\begin{align*}
\sigma &= \frac{2ie^2k_BT\ln[2\cosh(E_F/2k_BT)]}{(\omega + i\tau)n\hbar} + \frac{e^2}{4\hbar} \left[ \frac{\hbar\omega}{2} + i\hbar\omega \right] \\
I &= \int_0^\infty \left[ G(\delta) - G\left(\frac{\hbar\omega}{2}\right) \right]/(|\hbar\omega|^2 - 4\delta^2) d\delta \\
G(\delta) &= \sinh(\delta/k_BT)/[\cosh(\varepsilon_F/k_BT) + \cosh(\delta/k_BT)]
\end{align*}
\]

(5)

(6)

(7)

The left and right terms in eq 5 account for the intraband and interband electron transitions, respectively. \(E_F\) is the Fermi level of graphene. \(\tau = 100\) fs is related to the carrier–carrier intraband collisions and phonon emission is used in our calculations for the collision time.\textsuperscript{34,35,36,37} The dielectric function of the vacuum-like SU8 material was modeled as multiple Lorentz–Drude oscillators,\textsuperscript{38} while the dielectric...
function of SiO$_2$ was taken from ref 39. The Si substrate was omitted due to its negligible contribution to the NFRHT.

The photon tunneling probability $\xi_p$ of the evanescent modes between the emitter and receiver at a gap distance of 80 nm is calculated and shown in Figure 2a,b. Strong coupling modes arising from the matched SPPs could be observed when the graphene Fermi levels of the receiver ($E_{F2}$) and emitter ($E_{F1}$) are both $-0.13$ eV (Figure 2a), corresponding to our experiment with bias voltages ($V_2$, $V_1$) = (35, 35) V. The SPP coupling modes are slightly deviated from the ideal dispersion curves of a pair of suspended graphene sheets (dashed-dotted lines) due to the impact of the SU8 spacers and the SiO$_2$ substrates. Typical phonon polaritons from the SiO$_2$ substrate support the near-unity $\xi_p$ around wavelengths of 8.56 and 19.9 $\mu$m, but have no contribution to the heat flux modulation. The yellow-dashed lines correspond to the occupation factor $\Theta(T_{p},\omega) - \Theta(T_{s},\omega)$ in arbitrary units. The near-unity $\xi_p$ overlaps the dispersion curves of the SPP coupling modes of two Gr/SU8 heterostructures (not shown). The coupled modes split into two branches at a lower $\beta$ but merge at a larger $\beta$, as the larger loss (at large $\beta$) in the vertical direction enables rapid attenuation of the SPP modes and prevents the interaction between the emitter and receiver. These matched SPP coupling modes are supported at the mid- and far-infrared regions with a larger occupation factor and are the dominant contributor to the radiative heat flux. Compared with changing the graphene Fermi levels synchronously (e.g., change both $E_{F2}$ and $E_{F1}$ to $-0.22$ eV), producing dissimilar Fermi levels is a more effective way to pursue an improved modulation depth. When only the graphene Fermi level of the receiver changes to $-0.22$ eV (Figure 2b), the SPPs arising from the emitter and receiver contribute less to the near-unity $\xi_p$. Due to the mismatched SPP modes, the resonance modes are decoupled around $1.0 \times 10^{14}$ and $1.5 \times 10^{14}$ rad/s with large $\beta$. In contrast, the phonon polaritons modes remain unchanged due to the identical SiO$_2$ substrates on both sides. The spectral heat flux (after integration over $\beta$) between the Gr/SU8 heterostructures shows the influence of the graphene SPPs with identical or dissimilar Fermi levels (Figure 2c). When $E_{F2} = E_{F1} = -0.13$ eV (case I), the spectrum attributed to the matched strong SPPs covers a broad frequency region with the highest value. A broader spectrum with lower spectral heat flux (compared to case I) could be observed when $E_{F2} = E_{F1} = -0.22$ eV (case II). However, the spectral heat flux decreases dramatically when $E_{F2} = E_{F1} = -0.22$ and $E_{F1} = -0.13$ eV (case III). The spectrum in case III shares a similar frequency region with case I, but it has smaller intensity due to the mismatched resonance modes. The radiative heat flux reaches $1.34 \times 10^4$, $1.19 \times 10^4$, and $0.57 \times 10^4$ W/m$^2$ for cases I, II, and III, respectively. This leads to a remarkable improvement of the NFRHT modulation depth from case I to case III, where the modulation depth is 40.3% (calculated by (radiative heat flux of case I − radiative heat flux of case III)/(radiative heat flux of case I + radiative heat flux of case III) × 100%) and is ≈6.7-fold compared to that from case I to case II. Figure 2d gives the contour map of the radiative heat flux normalized with corresponding BB limit with variable $E_{F2}$ and $E_{F1}$. For different $E_{F1}$ at a fixed $E_{F2}$ the radiative heat flux arrives at a maximal value with two similar $E_{p}$. This confirms the importance of the symmetry of the polariton-mediated NFRHT system. White-dashed line shows the cases with matched coupling SPP modes. Case I with $\eta = 416$ is quite close to the peak point at $E_{F2} = E_{F1} = -0.14$ eV. The white-dashed-dotted line highlights the cases corresponding to
Figure 3. Schematic illustrations of the NFRHT experimental setup. (a) Schematic diagram of the receiver part. White-dashed square shows the active area of $3 \times 3 \text{mm}^2$. Au/Ti electrodes for both emitter and receiver are conductively contacted with the graphene sheets to control the Fermi level by the external electrostatic field. (b) Side view of the receiver. The receiver (or emitter) was modeled as two capacitors in series, where $C_{\text{SU8}}$ and $C_{\text{SiO}_2}$ are the capacitances per unit area of the SU8 spacer and SiO$_2$ dielectric, respectively. The graphene Fermi level was determined by the equivalent capacitance model. (c) Top view of the emitter and receiver. The conductive wires with positive and negative signs correspond to the emitter and receiver, respectively, estimated by the measured sum $P_{\text{sum}}$ $(\text{SI section 3})$ of the thermal resistances of the sample, thermal conductive adhesive, and the copper carrier. $T_1$ was maintained at 303.15 K, while $T_2$ was 308.15 K unless otherwise specified.

Figure 4. (a) Measurement setup of the NFRHT device. A back-gated device was employed to tune the Fermi level of graphene. $V_g = V_n$ allows an excess-electron surface concentration of $n = \eta_t(V_g - V_n)$, where $V_n$ is the gate-tuning voltage, $V_n$ is the voltage at the charge neutral point, and $\eta_t$ is a coefficient related to the back-gated structure with gate insulators composed of 90 nm thick SU8 and 300 nm thick SiO$_2$ (see SI section 1). The graphene Fermi levels could be obtained by $E_F = \text{sgn}(n) \ h V_g (\pi e h)^{1/2} / e$ (unit: eV), where $\text{sgn}(x)$ is the sign of $x$ and $V_F = 1 \times 10^6 \text{ m/s}$ is the Fermi velocity of graphene.\textsuperscript{30–44} The emitter was pressed on the receiver in a cross shape and separated by the SU8 nanopillars (Figure 3c). A total of 95 g mass above the emitter and two fixed posts were used to strengthen the contact and mechanical stability of the system (Figure 3d).\textsuperscript{5,10,12,33} The gap distance was estimated within a range from $\approx 79$ to $\approx 83$ nm (i.e., at an average value of $\approx 81$ nm) based on the one-dimensional linear elastic analysis\textsuperscript{6} (SI section 4). Figure 3e illustrates the equivalent thermal circuit of the system. $P_{\text{sum}}$, detected by the heat flux sensor (HFS) is the sum heat power of $P_r$ and $P_p$, where $P_r$ is the near-field radiative heat power including the contribution of both propagating waves and evanescent waves. The measured radiative heat flux could be obtained by $Q = (P_{\text{sum}} - P_r) / A$, where $A$ is the active area of $3 \times 3 \text{mm}^2$. $P_r$ is the heat conduction from the eight SU8 nanopillars and is $\approx 81$ nm. When no bias voltages are applied to the receiver within the active area (white-dashed square with $3 \times 3 \text{mm}$ in Figure 3a). A positive (negative) $V_g$ allows an excess-electron surface concentration of $n = \eta_t (V_g - V_n)$. A back-gated device was employed to tune the Fermi level of graphene. $V_g = V_n$ allows an excess-electron surface concentration of $n = \eta_t(V_g - V_n)$, where $V_n$ is the gate-tuning voltage, $V_n$ is the voltage at the charge neutral point, and $\eta_t$ is a coefficient related to the back-gated structure with gate insulators composed of 90 nm thick SU8 and 300 nm thick SiO$_2$ (see SI section 1). The graphene Fermi levels could be obtained by $E_F = \text{sgn}(n) \ h V_g (\pi e h)^{1/2} / e$ (unit: eV), where $\text{sgn}(x)$ is the sign of $x$ and $V_F = 1 \times 10^6 \text{ m/s}$ is the Fermi velocity of graphene.\textsuperscript{30–44} The emitter was pressed on the receiver in a cross shape and separated by the SU8 nanopillars (Figure 3c). A total of 95 g mass above the emitter and two fixed posts were used to strengthen the contact and mechanical stability of the system (Figure 3d).\textsuperscript{5,10,12,33} The gap distance was estimated within a range from $\approx 79$ to $\approx 83$ nm (i.e., at an average value of $\approx 81$ nm) based on the one-dimensional linear elastic analysis\textsuperscript{6} (SI section 4). Figure 3e illustrates the equivalent thermal circuit of the system. $P_{\text{sum}}$, detected by the heat flux sensor (HFS) is the sum heat power of $P_r$ and $P_p$, where $P_r$ is the near-field radiative heat power including the contribution of both propagating waves and evanescent waves. The measured radiative heat flux could be obtained by $Q = (P_{\text{sum}} - P_r) / A$, where $A$ is the active area of $3 \times 3 \text{mm}^2$. $P_r$ is the heat conduction from the eight SU8 nanopillars and is $\approx 81$ nm. When no bias voltages are applied to the emitter and receiver, i.e., $(V_{\text{E}}, V_{\text{R}}) = (0, 0)$, $E_{\text{F2}}$ and $E_{\text{F1}}$ are

\[ P_{\text{sum}} = P_r + P_p \]

\[ Q = \frac{P_{\text{sum}} - P_r}{A} \]

\[ E_F = \text{sgn}(n) \ h V_g (\pi e h)^{1/2} / e \]

\[ V_F = 1 \times 10^6 \text{ m/s} \]
calculated to be \(-0.205\) eV according to the measured radiative heat flux. According to the back-gated method, other bias voltages of (10, 10), (25, 25), (35, 35), and (45, 45) V correspond to the graphene Fermi levels of \(-0.187, -0.155, -0.13,\) and \(-0.098\) eV respectively. The Fermi levels become closer to the Dirac point with higher positive voltages, indicating the hole doping of the graphene sheets. When \(E_{F2} = E_{F1} = -0.13\) eV, the matched SPPs allow for the broadband near-unity \(\xi\) within the desired mid- and far-infrared region, producing the best performance among all measured cases. The radiative heat flux reaches \(1.43 \times 10^4\) W/m² and is \(\approx\)441-fold of the corresponding BB limit. When \(E_{F2} = E_{F1} = -0.205\) eV, the enhancement (\(\approx 381\)-fold with respect to the BB limit) is relatively robust, as the SPP modes of the emitter and receiver are still matched, despite the blue-shift to a higher frequency region with less occupation factor (similar to the spectral heat flux of case II in Figure 2c). For the mismatched cases, the bias voltage of the emitter \(V_1\) is fixed at 35 V while the bias voltage of the receiver \(V_2\) is set to \(-10, 0, 10, 25, 35,\) and 45 V. Remarkable modulation of the radiative heat flux was observed when the bias voltage of the receiver became different from that of the emitter. Here the minimum radiative heat flux (\(\approx 220\)-fold with respect to the BB limit) was observed at \((V_2, V_1) = (35, 35)\) V, where the graphene Fermi levels are dissimilar [i.e., \((E_{F2}, E_{F1}) = (-0.22, -0.13)\) eV]. When \(V_2\) increases to 35 V again, the radiative heat flux can still reach a peak value of \(1.39 \times 10^4\) W/m² with enhancement of \(\approx 429\)-fold of the BB limit, indicating the robustness of the tuning devices. Figure 4b illustrates the modulation depths [(measured maximum value—other measured value)/(measured maximum value + other measured value) \(\times 100\)%] of the matched and mismatched cases, respectively. The maximum modulation depth achievable with only matched cases is \(\approx7.3\)% going from \((E_{F2}, E_{F1}) = (-0.13, -0.13)\) eV to \((E_{F2}, E_{F1}) = (-0.205, -0.205)\) eV, but the modulation depth is \(\approx26.3\)% when only changing \(E_{F2}\) to \(-0.205\) eV. The maximum modulation depth from the matched case to a mismatched case reaches \(\approx32.2\)% when tuning \(E_{F2}\) to \(-0.22\) eV by applying \(-10\) V bias voltage to the receiver. It could potentially be further improved when other larger negative voltages are applied. Considering the breakdown of the capacitor-like samples, only \(-10\) V was investigated in this work. Construction of the mismatched resonance modes plays a significant role in the NFRHT modulation improvement. Switching the symmetry of the polariton-mediated near-field system gives promising high-efficiency modulation in thermal radiation. The modulation depth of \(\approx32.2\)% is \(\approx4.4\)-fold compared to that for the matched cases and is 16.1-fold of the previous report on graphene-based heterostructures. The modulation of the radiative heat flux at another \(\Delta T\) of 3 K was also investigated in Figure 4c. The time-varying heat flux were recorded by the HFS when changing the bias voltages from \((V_2, V_1) = (35, 35)\) V to \((25, 35)\) V repeatedly. The average modulation depth of \(\approx7\)% is similar to that with \(\Delta T = 5\) K in Figure 4b (\(\approx 8.3\)%). Small \(\Delta T\) (like that in this work) is more likely required in potential applications such as thermal communication, since the state of the thermal equilibrium will be easier to achieve after switching the graphene Fermi levels, hence a faster response time. The results show good repeatability and robustness of our devices for the NFRHT.
modulation. The sample in this work consists of a single-layer graphene-covered SU8 heterostructure, which is simpler and easier to fabricate, and allows for larger modulation depth (with mismatched modes) than that of the previous work with multilayer structure (only matched modes are considered). The physical mechanism in ref 6 is that multilayer systems allow many branches in k-space to provide stronger NFRHT and modulation with the tuning of only one Fermi level was used for the optimal radiative heat flux. This work focused on the ultrahigh dynamic modulation and the essential physics is that independent tuning of two different Fermi levels allows a massive improvement in modulation depth. In addition, the determination of the graphene Fermi level is significantly improved by the equivalent capacitance model related to the back-gated tuning devices.

The modulation depth versus variable vacuum gap distance \( d \) within a range from 50 to 500 nm is theoretically investigated (Figure 5). The black-dashed line shows the gap-dependent radiative heat flux between two identical Gr/SU8 heterostructures with graphene Fermi levels of \((E_{F2}, E_{F1}) = (-0.13, -0.13)\) eV and \((E_{F2}, E_{F1}) = (-0.22, -0.13)\) eV, respectively.

**Figure 5.** Calculated modulation depth (red-solid line) versus variable vacuum gap distance \( d \) within a range from 50 to 500 nm. The temperatures of the emitter and receiver are set to 308.15 and 303.15 K, respectively. The black-dashed line and dashed-dotted line show the calculated radiative heat flux between two Gr/SU8 heterostructures with graphene Fermi levels of \((E_{F2}, E_{F1}) = (-0.13, -0.13)\) eV and \((E_{F2}, E_{F1}) = (-0.22, -0.13)\) eV, respectively.

Back-gated tuning of the graphene Fermi level; sample fabrication and characterization; experimental setup and measurement of the thermal resistance \( R_t \); gap distance between the emitter and receiver; heat conduction of the SU8 nanopillars; uncertainty analysis of the measured radiative heat flux (PDF)

### ASSOCIATED CONTENT

**Supporting Information**

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Author Contributions
S.H. conceived and supervised the work. K.S. conceived the work and performed the calculations. K.S., Z.C., J.Y., and X.X. fabricated the samples. K.S., Z.C., and J.Y. performed the sample characterization. K.S. and Z.C. performed the experiments. The manuscript was discussed and written by K.S., J.E., Y.X., and S.H. with comments and input from all authors.

Notes
The authors declare no competing financial interest.

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