Optimized Colossal Near-Field Thermal Radiation Enabled by Manipulating Coupled Plasmon Polariton Geometry

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Supporting Information

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TABLE OF CONTENTS
Supporting Information Section 1: Sample fabrication
Supporting Information Section 2: Sample characterization
Supporting Information Section 3: Temperature calibration
Supporting Information Section 4: Experimental system and measurement
Supporting Information Section 5: Gap distance and SU8 thermal conductivity
Supporting Information Section 6: Dielectric function of SU8 spacer
Supporting Information Section 7: Effect of heat conduction of SU8 nano pillars
Supporting Information Section 8: Effect of number of layers of Gr/SU8 heterostructures for NFTR
1. Sample fabrication

(a) For receiver samples

Samples are fabricated using a standard UV lithography technique and wetting transfer method (for graphene sheets) as schematically illustrated in Figure S1. **Step 1**: A clean ~300-nm-thick SiO\(_2\) film on a Si wafer was dealt with Hexamethyl Disilazane (HMDS) for 20 min to obtain a hydrophobic surface which promotes photoresist adhesion. **Step 2**: A photoresist (AZ5214, Resemi) was spin coated onto the wafer surface at 500 rpm for 6 s followed by 2000 rpm for 50 s and then baked for 5 min at 90°C, serving as protective layer with thickness of ~2 μm. **Step 3**: UV lithography produced nine 25 × 25 mm\(^2\) blocks containing square patterns with size of 16 × 16 mm\(^2\) which were then immersed in a developing solution (300MIF) and cleaned several times with deionized water (DW) **Step 4**: The wafer was dried using a nitrogen gas gun and cut into nine identical samples. **Step 5**: Inductively coupled plasma (ICP) thoroughly etched the SiO\(_2\) except for the region protected by the photoresist (with 16 × 16 mm\(^2\) area). **Step 6**: To clean the photoresist, the samples were ultrasonicated in NMP solution for 1 h, followed by isopropyl alcohol (IPA) for 10 min and DW for 10 min, and soaked in Piranha solution (volume ratio, concentrated sulfuric acid (H\(_2\)SO\(_4\)) : hydrogen peroxide (H\(_2\)O\(_2\)) = 7 : 3) to eliminate organic residues, and rinsed several times with DW. **Step 7**: The samples were spin coated with dilute SU8-3005 photoresist (mass ratio, SU8 : solvent cyclopentanone = 1 : 13) at 500 rpm for 6 s followed by 3300 rpm for 50 s, and then baked for 30 s at 95°C. **Step 8**: 10 × 10 mm\(^2\) SU8 spacers (marked as the fifth SU8 layer) with thickness of ~90 nm were fabricated after the UV lithography with an exposure dose of 250 mJ and post baked at 95 °C twice for each with 30 s, followed by immersion in a developing solution (PGMEA) and standard cleaning procedures. **Step 9**: A sheet of graphene (on copper foil) covered by a paraffin protective layer with a rectangular pattern (with size of \(L_1 > 10\) mm and \(L_2 \geq 10\) mm for the fifth graphene layer) was transferred to the sample using a wetting transfer method\(^1\) (WTM, see details in **Step 9.1** to **Step 9.9** below). **Step 10**: The Paraffin protective layer was then removed by soaking several times in trichlороethylene (C\(_2\)HCl\(_3\)). The samples were then baked in an oven at 95°C for 3 h (optional procedure to melt large paraffin particles and enhance the contact between graphene and substrate) and soaked in C\(_2\)HCl\(_3\) again, followed by very careful cleaning with IPA and DW to avoid graphene breakage. **Step 11**: For better spin coating of the fourth SU8 layer, the samples were baked at 95 °C for 25 min to reduce the moisture on the surface. The same parameters were employed as in Step 7 except that the samples were dried naturally for 30 min after spin coating instead of baking for 30 s at 95°C. **Step 12**: The UV lithography procedures from Step 8 were
repeated, and the $9 \times 9 \, \text{mm}^2$ SU8 spacers were well-prepared for transferring the fourth layer of graphene (with size of $L_1 > 10 \, \text{mm}$ and $L_2 = 9 \, \text{mm}$). **Steps 13-17:** Fabrication of the third (shown in Figure S1), second, and first layer of both SU8 spacers and graphene sheets, with procedures repeated from Steps 9-12. The areas of SU8 spacers for the third, second, and first layer were $8 \times 8 \, \text{mm}^2$, $7 \times 7 \, \text{mm}^2$, and $6 \times 6 \, \text{mm}^2$. Hence, lengths of graphene sheet in Step 9 were correspondingly change to $L_2 = 8 \, \text{mm}$, $7 \, \text{mm}$, and $6 \, \text{mm}$, respectively. **Step 18:** To prepare the Au electrodes, AZ5214 photoresist was spin coated on the samples at 500 rpm for 6 s followed by 2000 rpm for 50 s and then baked for 2 min at 90 °C. **Step 19:** Patterns were obtained after a standard UV lithography and very careful cleaning. **Step 20:** A 5-nm-thick titanium adhesion layer was deposited on the samples, followed by the deposition of a 95-nm-thick Au film, using an E-beam evaporation method. **Step 21:** Metal films were removed using a lift-off method in acetone solution. **Step 22:** To prepare the SU8 nano pillars. Dilute SU8-3005 (mass ratio, SU8 : solvent cyclopentanone = 1 : 21) was spin coated on the samples at 500 rpm for 6 s followed by 3000 rpm for 50 s, leading to a homogeneous thickness of ~ 58 nm to ~ 63 nm. The same procedures as in step 8 were used to obtain eight SU8 nano pillars with diameter of 20 μm. Emitter samples were fabricated with identical methods without the SU8 nano pillars.

Samples close to the center of the SiO$_2$/Si wafer were chosen as the emitter and receiver unless the complicated fabrication procedures produced unusable units. SU8 spacers, Au electrodes, SU8 nano pillars were aligned centrally with alignment marks within an estimated deviation of ~ 2 μm, which was negligible for millimeter-scale plates. **[Caution:]** Piranha solution is highly corrosive and explosive. Trichloroethylene (C$_2$HCl$_3$) is volatile poisonous organic vapour. Make sure these solutions are used inside a fume cupboard and wear a gas mask]
**Figure S1.** Schematic illustration of fabrication of the Gr/SU8/5L sample. Figure in Step 3 is a photo of the SiO₂/Si wafer with patterns of AZ5214 photoresist after UV lithography. Step 4 to step 22 show the side views of the sample with size of 25 × 25 mm². The schematic illustrations are not drawn to scale. 

(b) Wetting transfer method for graphene sheets: **Steps 9.1-9.9**

Based on Ref. [1] and our experimental conditions, paraffin (Sigma-Aldrich, 18634) was chosen as a protective layer when transferring graphene sheets. **Step 9.1:** Graphene on copper foil (CVD commercial product, graphene/Cu/backside graphene) was cut down and put onto a quartz or Si substrate and carefully fixed by adhesive tape on the edges. **Step 9.2:** After heating in an oven for 60 min at 80 °C, paraffin liquid was dropped onto the graphene sample on the substrate and solidified. **Step 9.3:** Paraffin was spin coated onto the graphene after melting by a heat gun.[1] **Step 9.4:** Graphene coated by paraffin film was cut into the required size. The copper foil was etched by 1 (mol L⁻¹) FeCl₃ solution for 8 min and the back side of the copper foil was washed with DW to remove extra graphene (on the back side of the copper foil). **Step 9.5:** The paraffin/graphene/Cu sample was then transferred to a new FeCl₃ solution for 70 min to sufficiently etch the copper foil. **Step 9.6:** A clean hydrophilic substrate (e.g., a SiO₂/Si substrate) was employed to transfer the floating paraffin/graphene into clean DW twice and allowed to sit for 10 min for each time to remove the FeCl₃ residues. **Step 9.7:** To obtain a relatively flat graphene sheet,[1] the floating paraffin/graphene was transferred to a new clean DW and heated in a water bath at ~ 35°C to 38 °C for more than 1 h. **Step 9.8:** Paraffin/graphene was finally transferred onto the target sample very carefully. To avoid tearing of the fragile paraffin/graphene (at ~ 35 °C to 38 °C), this operation was done after the DW naturally cooled down to ~ 25 °C. **Step 9.9:** The sample was then placed in an oven at 40 °C for 24 h to remove the residual water.
Figure S2. Schematic illustration and photos of transferring a graphene sheet to a SU8 spacer. a) Step 9.1 to Step 9.9 show the details of the wetting transfer method for graphene sheet. b-f) Photos of Step 9.1, Step 9.2, Step 9.5, Step 9.6, and Step 9.7, respectively. The schematic illustrations in Figure S2a are not drawn to scale.

(c) Wires for bias voltage device.

The sample after Step 22 was cut to a required size (~25 × 3 mm²). As depicted in Figure S3, two wires (schematic diagram) were attached to the sample as positive (connected to Au electrode on Si substrate) and negative electrodes (connected to Au electrode on SiO₂). The sample was placed on a hot plate and the wires were bonded to the end of each Au electrode with conducting resin (H20E, EPO-TEK, Epoxy), followed by a heating at 110°C for 20 min to solidify the contact areas between the wires and the Au electrodes. All fabrication processes in (a) (b) (c) were preformed in a 100-class modular cleanroom.
Figure S3. Photograph of the well-prepared Gr/SU8/5L sample with size of $\sim 25 \times 3$ mm$^2$. Two independent wires are bonded at the end of the Au electrodes for bias voltage as schematically depicted.

2. Sample characterization

Sample characterization involves three parts: (a) Surface curvature and surface residue of the sample. (b) Characterization of SU8 spacers and nano pillars. (c) Raman spectra of graphene sheets.

(a) Surface curvature and surface residue of the sample

A relatively flat surface could be obtained near the center of the SiO$_2$/Si wafer.$^{[2,3]}$ As shown in Figure S4a, the natural bow of a SiO$_2$/Si substrate was measured by a laser interferometer (ZYGO OMP-035/M). The maximum surface curvature was $\sim 7$ nm within an area of $3 \times 3$ mm$^2$, which is comparable to the flatness reported elsewhere.$^{[4,5]}$ The relative heights along four scanning directions of the SiO$_2$/Si substrate are shown in Figure S4b, indicating an adequately flat surface compared to the gap distance. Hence, the samples were treated as a flat plate-to-plate model in our calculation.$^{[2-5]}$
Figure S4. a) Surface curvature of the $3 \times 3$ mm$^2$ active region of a SiO$_2$/Si substrate, measured using the laser interferometer. b) Relative heights from four directions of the sample extracted from Figure S4a.

Paraffin is easier to remove from graphene than PMMA.$^{[1]}$ However, great efforts were still made to obtain a relatively clean graphene sheet as the thickness of the next SU8 spacer was only $\sim 90$ nm. After Step 10 in Section 1, the surface residues (mainly paraffin) of graphene sheet were characterized by AFM scanning as shown in Figure S5a. Graphene features a relatively flat surface with roughness less than $\sim 1.5$ nm. Some residues may have thickness of $\sim 34$ nm, $\sim 40$ nm, $\sim 36$ nm or $\sim 55$ nm (white-dashed circles) but they should not affect the
flatness and roughness of the next SU8 spacer. Figure S5b shows the ‘topography’ of Figure S5a where shading is accounted for which gives a more intuitive understanding of the height. Similar methods were employed in Figure S5d, Figure S5f, and Figure S5h.

We also characterized the surface roughness of SU8 spacer after spin coating onto the graphene sheet by AFM scanning. As illustrated in Figure S5c-S5d, the roughness of SU8 spacer is less than ~ 1 nm. Figure S5d shows that little particles surrounded by SU8 have thickness of ~ 10 nm (leveling to SU8 ‘substrate’), showing a good flatness of the SU8 spacer as expected. For comparison, AFM scanning of SU8 coated on a smooth SiO$_2$/Si was also presented, showing a similar roughness with that in Figure S5c. AFM scanning image of the first (top) graphene layer of a Gr/SU8/5L sample was shown in Figure S5g-S5h. The average roughness is ~ 1.5 nm with maximum thickness of the residues of ~ 54 nm. The areas of the residues are quite small compared with the scanning area of 8 × 8 μm$^2$. It is reasonable to assume that the heat conduction of the residues between the emitter and receiver is negligible during the NFTR experiments, which was also demonstrated by the good agreement of our computed and experimental data.
**Figure S5.** $8 \times 8 \ \mu m^2$ AFM scanning images of: a) Graphene on SU8. c) SU8 spacer after spin coating onto the graphene sheet. e) SU8 spin coated on a SiO$_2$/Si substrate. g) the top
graphene layer of our Gr/SU8/5L. (b), (d), (f) and (h) are the corresponding ‘topography’
maps with shading effects.

(b) Characterization of SU8 spacers and nano pillars
The thickness of the SU8 spacers (without graphene sheets) was confirmed by a film
thickness measuring instrument (Filmetrics, F40-UV). As illustrated in Figure S6, twenty-five
points were taken as samples within the active area (3 × 3 mm²) for each of the layers. Data
for each point was obtained by measuring the reflectance with wavelength from 400 nm to
1100 nm. The average standard deviation within such an area is less than 3 nm, indicating a
quite flat surface could be obtained with the spin coating procedures. Figure S7 give one of
the measurements in Figure S6 for the five layers of SU8 spacer, respectively, showing the
agreement between the fitting and measured data. The thickness of each SU8 spacer could be
confirmed as ~ 90 nm. Hence, considering the cleanliness of graphene sheets and the same
experimental procedures, it was reasonable to assume that the thickness of each SU8 spacer of
the Gr/SU8/5L sample was ~ 90 nm.

Figure S6. Thickness of the five layers of SU8 spacers within the 3 × 3 mm² active area
measured at twenty-five data points.
**Figure S7.** Thickness of each SU8 spacer obtained by the reflectance by Filmetrics (F40-UV). Red lines are the fitting data calculated by Cauchy model. Blue lines are the measured data.
The thicknesses of the SU8 nano pillars were equivalently confirmed by that of the SU8 film after UV treatments. Using the same experimental parameters in Step 22 in Section 1, the SU8 film was fabricated on SiO₂/Si substrate with thickness measured by Filmetrics (F40-UV) as well as an ellipsometer (not shown here). Twenty-five points for each sample were measured by Filmetrics within the active area. As shown in Figure S8, the maximum and minimum thickness of the SU8 films for different samples are ~63 nm and ~58 nm, respectively. The size of the top (first) SU8 spacer was 6 × 6 mm², which is larger than that of the active area and should offer an adequately flat ‘substrate’ for the nano pillars fabrication (as confirmed by Figure S5c and Figure S6). Hence, the thickness of the SU8 nano pillars was estimated within this range (i.e., ~58 nm to ~63 nm). Inset in Figure S8 is the optical image of a SU8 nano pillar with diameter of 20 μm. Note that during the NFTR experiment in our case, a total 95 g mass was applied to the emitter for mechanical stability which led to a small decrease of the thickness of the SU8 nano pillars (on the receiver), see SI Section 5 below.

**Figure S8.** Thickness of the SU8 nano pillars are equivalently characterized by the SU8 films with same experimental parameters in Step 22 in Section 1. Sample X and sample Y give the minimum and maximum thicknesses. Inset shows the optical image of a SU8 nano pillar.

(c) Raman spectra of graphene sheets

Graphene sheets (of the Gr/SU8/5L) on different layers were in conductive contact through the Au/Ti electrodes and should have a similar Fermi level.\(^6\) Hence, for simplicity, the Fermi level of the fifth graphene layer of a well-prepared Gr/SU8/5L sample was determined from the Raman spectra shown in Figure S9 without (Figure S9a) or with bias voltages (Figure...
S9b). The higher intensity of the 2D peak (compared to the G peak) is indicative of single-layer graphene.\cite{2,5,7,8} The Fermi level could be determined from the wavenumber $w$ of the G peak according to the established equation: \( w_G = 1580 \text{ cm}^{-1} = (42 \text{ cm}^{-1}/\text{eV}) |E_F| \).\cite{8,10} For simplicity, the fifth graphene layer was then characterized by Raman spectra when applying different bias voltages, such as 5 V, 20 V, and 40 V. The measured G peaks of graphene with different bias voltages were shown in Figure S9b. The Fermi levels of the graphene sheets could be found as: \(-0.105\) eV at 0 V, \(-0.13\) eV at 5 V, \(-0.125\) eV at 20 V, and \(-0.127\) eV at 40 V. \( w_G \) shifted to a smaller absolute value when more electron carriers flowed to the graphene sheets (with positive bias voltage), indicating a hole doping of the original graphene sheets. These measured values are only used for reference and the Fermi levels of the Gr/SU8/5L for the NFTR measurement in the main text were fitting parameters. The Fermi levels for different samples should feature a relatively small deviation due to the same sample fabrication procedures (i.e., samples came from a same batch).

The Fermi level for each sheet should be similar because they are in conductive contact through the gold electrode. The influence of fluctuations of the Fermi level on the NFTR was investigated to understand the sensitivity to sheets with mismatching Fermi levels. As shown in Figure S10, the NFTR between the Gr/SU8/5L heterostructures with Fermi level fluctuation versus different \( AF \) were plotted. The gap distance \( d \) is 54.7 nm and \( T_2 \) is 303.15 K. The five Fermi levels are set to:

| $d=$54.7nm | Case A (main text) | Case B | Case C | Case D | Case E | Case F |
|-------------|---------------------|--------|--------|--------|--------|--------|
| $E_{F1}$ (eV) | -0.11 | -0.20 | -0.11 | -0.11 | -0.13 | -0.09 |
| $E_{F2}$ (eV) | -0.11 | -0.17 | -0.115 | -0.105 | -0.125 | -0.095 |
| $E_{F3}$ (eV) | -0.11 | -0.15 | -0.12 | -0.10 | -0.12 | -0.10 |
| $E_{F4}$ (eV) | -0.11 | -0.13 | -0.125 | -0.095 | -0.115 | -0.105 |
| $E_{F5}$ (eV) | -0.11 | -0.11 | -0.13 | -0.09 | -0.11 | -0.11 |

Table S1. List of the Fermi level fluctuations. Case A is assumed in the main text.

Firstly, with a larger fluctuation (e.g., Case B, the graphene Fermi level $E_{F1}$ is almost 2-fold of $E_{F5}$), the error of the NFTR heat flux is \( \sim 24\% \) compared to Case A, which does not match well with the experiment results. While with a smaller fluctuation, e.g., from Case C to Case F, the error of the NFTR heat flux compared to Case A is less than \( \sim 3.1\% \). Small or moderate local fluctuations do not meaningfully disrupt the NFTR. Hence, it is justified to assume a
Fermi level of -0.11 eV for each layer, which is also verified by the good agreement of the experiment data and theoretical calculation.

**Figure S9.** Raman spectra for the Gr/SU8/5L sample with different bias voltages. a) Raman spectra of the fifth graphene sheet was displayed without bias voltage (i.e., 0 V). b) Raman peaks from G resonance modes of the fifth graphene sheet were illustrated with bias voltages of 0 V, 5 V, 20 V, 40 V, respectively.
Figure S10. NFTR heat flux between the Gr/SU8/5L heterostructures with different amounts of Fermi level fluctuation. The gap distance $d$ is 54.7 nm and $T_2$ is 303.15 K. Case A has the Fermi level of each graphene layer equal to -0.11 eV, which is used in the main text. Graphene Fermi level in other cases with a fluctuation around -0.11 eV are listed in Table R1.

3. Temperature calibration

The surface temperatures of the emitter and receiver were estimated by the relationship: $T_2 (T_1) = T_{2b} (T_{1b}) \pm P_{total} R_1$. The measured value $R_1$ (unit: K/W) was the sum of the thermal resistances of the emitter (receiver), thermal conductive adhesive and the copper carrier (between the temperature detector and the thermal conductive adhesive). Hence $R_1$ could be obtained by:

$$R_1 = \frac{1}{2} \frac{(T_{1b} - T_{2b})}{P_{total}}$$

(S1)

As shown in Figure S11, two platinum resistance temperature detectors (thermistor, M222 class 1/3B, Heraeus, Germany) coated with a little thermal grease were buried into a hole beneath the top surface of the copper carriers to characterize the backside temperature $T_{1b}$ of the emitter and $T_{2b}$ of the receiver. In this case, the emitter and receiver were two identical SiO$_2$/Si samples with wires connected to the Au/Ti electrodes (not shown here), as the thermal resistance of the graphene-covered SU8 films could be ignored due to the huge width-depth ratio. The thin thermal grease was glued onto the interface between the emitter and receiver to minimize the contact thermal resistance, and a total 95 g mass (including copper carrier A) was added onto copper carrier A to strengthen the contact and mechanical stability of the
system (not shown here). The measurement was done in a high vacuum environment ($\sim 2.8 \times 10^{-5}$ Pa). $R_1$ from three independent measurements were shown in Figure S11, with range from $\sim 7.49$ K/W to $\sim 11.5$ K/W. Hence, $R_1$ was set at an average value of $\sim 9.2$ K/W during the NFTR experiments.

**Figure S11.** Measured value of $R_1$ with different $\Delta T$. Inset shows the key experimental set-up. The sum thermal resistance of the emitter, thermal conductive adhesive and the copper carrier (between the temperature detector and the thermal conductive adhesive) was assumed to be same as that of the receiver.

### 4. Experimental system and measurement

Measurement of the NFTR was done in a vacuum chamber with a high vacuum environment ($\sim 2.8 \times 10^{-5}$ Pa). The parallel alignment of the emitter and receiver was guaranteed by the SU8 nano pillars between them. Such a method for a fixed gap distance is widely employed. Each point of the curves in Figure 3 in the main text was an average value from three possible active areas (i.e., $2.9 \times 2.9$ mm$^2$, $3 \times 3$ mm$^2$, and $3.1 \times 3.1$ mm$^2$) after four readings from the external heat flux meter (connected to the heat flux sensor below copper carrier B). Error bars were plotted due to the uncertainty of the active area. Temperature $T_2$ was maintained by a temperature electric controller (TEC) with an external source (Keithley, 2231A-30-3, Keithley instruments, USA) according to the thermal resistance $R_1$ and the total heat power $P_{\text{total}}$. Temperature $T_{1b}$ and $T_1$ were then recorded after holding for at least 10 min to ensure thermal equilibrium. Temperature $T_{1b}$ and $T_{2b}$ were obtained by reading the values of the platinum resistance temperature detectors via an external digital multimeter.
(Keithley, DMM6500, Keithley instruments, USA). Figure S12 shows the whole experimental system. The emitter and receiver were controlled at the same bias voltages by external sources. The experimental data at other bias voltages with different $\Delta T$ were obtained with the same operations mentioned above. Before the NFTR experiment, some Gr/SU8/5L samples were used for Raman spectra measurement at different bias voltages. Since we found that under the bias voltage of ~ 40 V, the graphene Fermi level of our samples could reach ~ -0.1 eV to ~ -0.13 eV, which meets the needs of our theoretical prediction, we used the bias voltage only up to 40V.

The NFTR experiments with different bias voltages were challenging and highly dependent on the quality and cleanliness of the graphene sheets. To have a better chance of success of the NFTR experiments, one may employ polymer-free method,$^{[13]}$ or dry transfer method$^{[14]}$ for a better surface cleanliness, and may use a smaller size of the active area (e.g., 0.5 × 0.5 mm$^2$) to obtain a more flat surface. To fabricate such a small and complex system (intergrated with temperature measurement and Fermi level manipulation), one may refer to Ref. $^{[15-19]}$.

![Figure S12. Schematic illustration of the whole measurement system. The equivalent thermal circuit was also displayed.](image-url)
5. Gap distance and SU8 thermal conductivity

The gap distance between the emitter and the receiver was a calculated parameter according to the thickness and mechanical properties\cite{4,20} of the SU8 nano pillars. As mentioned in the main text, a total 95 g mass was applied to the emitter for mechanical stability but led to a small decrease of the thickness of the SU8 nano pillars (as shown in Figure S13a). Young’s modulus of SU8 is in the range of $\sim 3.6$ GPa to $\sim 4.1$ GPa at room temperature.\cite{20} Hence, an average value of 3.85 GPa was considered in our case. A one-dimensional linear elastic analysis was employed according to Hooke’s law. Displacement $\Delta x$ could be calculated by:

$$\Delta x = \frac{F_x}{E_{SU8}}$$

where $t$ is the thickness of the SU8 nano pillar, $F_x$ is the stress along the axial direction of the SU8 nano pillar, and $E_{SU8}$ is the corresponding Young’s modulus. When the 95 g mass was applied to the SU8 nano pillars with thickness in a range of $\sim 58$ nm to $\sim 63$ nm (SI Section 2), displacements of $\sim 5.6$ nm to $\sim 6.1$ nm would occur. Similar analyses (not shown here) could verify the results in Ref. [4] computed by Comsol Multiphysics. Thus, the gap distance between the emitter and receiver was within a range from $\sim 52.4$ nm to $\sim 57$ nm (i.e., $d \sim 55$ nm in the main text), which was also verified by the agreement between the computed and experimental data of the NFTR.

We also measured the NFTR between two identical $\sim 305$-nm-thick SiO$_2$/Si samples. SU8 nano pillars with thickness in a range of $\sim 88$ nm to $\sim 92$ nm (using the same experimental parameters as the SU8 spacer, see Step 7 in Section 1) were fabricated onto the receiver. According to elastic analysis, the gap distance between the emitter and receiver should be in a range of $\sim 79.5$ nm to $\sim 83.1$ nm. The computed and experimental data displayed in Figure S13b were consistent with this analysis. For comparison, heat flux within a gap distance range from 88 nm to 92 nm versus different $\Delta T$ were also calculated, showing a clear difference between the computed and experimental data. Temperature $T_2$ was maintained at 301.65 K $\pm$ 0.5 K. The dielectric functions of SiO$_2$ and Si were taken from Ref. [21].

The thermal conductivity has been verified by the NFTR experiment via a pair of 305-nm-thick SiO$_2$/Si sample (Figure S13b), as the experimental data of the NFTR ($= \text{measured total heat flux}$ - $\text{heat conduction portion}$) agrees well with the prediction. In this case (305-nm-thick SiO$_2$/Si sample at a gap distance of 81.3 nm), the ratio of the heat conduction to the total heat flux is quite high ($\sim 58\%$; due to the large gap distance and consequently the near-field coupling is relatively small). We have obtained the heat conduction by subtracting the well-known NFTR of the 305-nm-thick SiO$_2$/Si sample\cite{16,19} from the total heat flux, and plotted
the measured thermal conductivity (Figure S13c), which matches well with the value given by the SU8-3005 producer and Ref. [4]. The average value of the measured thermal conductivity is $0.207 \pm 0.01 \text{ W m}^{-1} \text{ K}^{-1}$ (red-dotted line). Hence, the thermal conductivity of $0.2 \text{ W m}^{-1} \text{ K}^{-1}$ used in our work is reliable and reasonable.

**Figure S13.** a) Schematic illustration of the condition of a SU8 nano pillar before (left) and after (right) adding the mass. b) Measured NFTR heat flux between two ~ 305-nm-thick SiO$_2$/Si samples with different $\Delta T$ within a gap distance range from 79.5 nm to 83.1 nm (i.e., $d \sim 81.3$ nm). The computed data for $d = 79.5$ nm to 83.1 nm (blue) and $d = 88$ nm to 92 nm (red) were given for comparison. Temperature $T_2$ was maintained at 301.65 K $\pm$ 0.5 K. c)
Verification of the thermal conductivity of the SU8 nano pillars. The red-dotted line is the average value of the measured thermal conductivity. Points are the experimental data of the thermal conductivity obtained from the heat conduction. The black-solid line represents the thermal conductivity of 0.2 W m\(^{-1}\) K\(^{-1}\) used in our work. Error bars were plotted due to the uncertainty of the active area (from 2.9 \(\times\) 2.9 mm\(^2\) to 3.1 \(\times\) 3.1 mm\(^2\)).

6. Dielectric function of SU8 spacer

The dielectric function of SU8 (\(\varepsilon_{SU8}\)) was retrieved from the measured reflectance spectra using a Fourier transform infrared (FTIR) spectrometer. SU8 film was spin coated on a 95-nm-thick Au film (with 5-nm-thick Ti as adhesion layer on the Si substrate). The same UV lithography procedures in Step 8 in Section 1 were employed. SU8 films with thickness of 595 nm and 819 nm (which were both average values measured by the Filmetrics) were used for the reflectance spectra experiments with normal incidence for better signal noise ratio (SNR). The Multiple Lorentz-Drude oscillator model\(^{[22,23]}\) given in the methods in the main text was employed for the dielectric function of SU8. Table S2 gives the parameters of the SU8 dielectric function. The error of the measured value and fitted value of each frequency point was put into a column vector, and the average value of the norm of the two column vectors (with data come from 595-nm and 819-nm SU8 thin film) was taken through MATLAB as the error function. The fitting and measured reflectance spectra in a wavelength range from 1.42 \(\mu\)m to 20 \(\mu\)m were illustrated in Figure S14a. The average relative error was less than 0.3% indicating a good agreement. The reflectance spectra within the color bands (covered a wavelength range from 9 \(\mu\)m to 20 \(\mu\)m) indicate negligible absorption (i.e., small loss) of the SU8 films. Figure S14b gives the real and imaginary portions of the dielectric function of SU8 described by the multiple Lorentz-Drude oscillator models.
Figure S14. a) Measured (colored lines) and fitting data (dash-dotted lines) for the reflectance of the SU8 films with thickness of 819 nm and 595 nm, respectively. b) Real and imaginary parts of the dielectric function of the SU8 retrieved by the reflectance spectra.

| $\varepsilon_{\text{inf}}$ | $A_{1x}$ (a. u.) | $A_{2x}$ (cm$^{-1}$) | $A_{3x}$ (a. u.) |
|--------------------------|-------------------|----------------------|------------------|
| 1.39999916               |                   |                      |                  |
| x = 1                    | 0.00146075124671989 | 3434.00449603104    | 0.0550829714785812 |
| x = 2                    | 0.00339324045214859 | 2929.00010065020    | 0.0342293909903620 |
| x = 3                    | 0.000254076611781735 | 1726.95250654918   | 0.000238835928456674 |
| x = 4                    | 0.00164505234500289 | 1607.32938462883    | 0.0123386061018492 |
| x = 5 | 0.00766820735287619 | 1506.00012268947 | 0.0166791825465552 |
| x = 6 | 0.000799976495587904 | 1454.55691091618 | 0.00408971338164520 |
| x = 7 | 0.00199995712070057 | 1299.97538186534 | 0.0243073273473874 |
| x = 8 | 0.0119999546282229 | 1243.54009888685 | 0.0230847195383060 |
| x = 9 | 0.00299993349067481 | 1180.01066179183 | 0.0339948737217765 |
| x = 10 | 0.0119994935888491 | 1107.00214537184 | 0.0362603031297629 |
| x = 11 | 0.0116815750547869 | 830.973779283478 | 0.0366756925269940 |
| x = 12 | 0.00242967416538911 | 755.762192815132 | 0.037814204042466 |
| x = 13 | 0.00499559061930623 | 668.504481832154 | 0.0353934972498788 |
| x = 14 | 2.09166652558700e-05 | 520.673151122782 | 2.61640166279757e-05 |

Table S2. List of the parameters of the Lorentz-Drude model for the SU8 dielectric function.

7. Effect of heat conduction of SU8 nano pillars

The heat conduction of the SU8 nano pillars was calculated using Fourier’s Law (here the temperature difference was taken as absolute value):

\[ P_c = \frac{S\kappa |\Delta T|}{d}, \]

(S3)

where \( S \) is the sum area of eight SU8 nano pillars, \( \kappa \) is the thermal conductivity\(^4\) \( (0.2 \text{ W m}^{-1} \text{ K}^{-1}) \), and \( d \) is the thickness of SU8 nano pillars which is set at an average value of \( \sim 54.7 \text{ nm} \) (i.e., \( (52.4+57)/2 \text{ nm} \)) in the calculations. Figure S15 show the ratios of the heat conduction \( P_c \) with respect to (a) the total heat power and (b) the NFTR power \( P_t \) at different \( \Delta T \) for the Gr/SU8/5L samples at \( E_F = -0.11 \text{ eV} \), \(-0.17 \text{ eV} \), \(-0.25 \text{ eV} \), and \(-0.36 \text{ eV} \). For simplicity, the gap distance \( d \) was set at 54.7 nm, and temperature \( T_2 \) was 304.15 K. The ratio with respect to the total heat power reaches a value of \( \sim 0.13 \) at \( E_F = -0.11 \text{ eV} \) and \( \sim 0.20 \) at \( E_F = -0.36 \text{ eV} \), indicating the contribution of the heat conduction to the total heat power is relatively small. Hence, in our case, the results of the NFTR power \( P_t \) (obtained by subtracting the calculated heat conduction \( P_c \), whose thermal conductivity has also been verified by our experiment in a simple case, from the measured total heat power \( P_{\text{total}} \)) should be fairly close to the actual NFTR power \( P_t \). To achieve a smaller heat conduction, one can reduce the area or increase the length of the SU8 nano pillars.\(^4\)
Figure S15. Ratio of the heat conduction $P_c$ with respect to (a) the total heat power and (b) the NFTR power $P_r$ at different $\Delta T$ for the Gr/SU8/5L samples with different graphene Fermi levels. Gap distance $d$ was set at 54.7 nm, and temperature $T_2$ was 304.15 K.

8. Effect of number of layers of Gr/SU8 heterostructures for NFTR

We have theoretically investigated the effect of the number of layers of the Gr/SU8 heterostructures for NFTR. Calculations of the heat flux normalized with the corresponding BB limit at a fixed gap distance $d = 50$ nm were displayed in Figure S16a for Gr/SU8/1L, Gr/SU8/2L, Gr/SU8/3L, Gr/SU8/4L, and Gr/SU8/5L heterostructures with different $E_F$, respectively. Bulk SiO$_2$ was considered as a substrate. Gr/SU8 with infinite layers was also computed for comparison. Thickness $t$ of each SU8 spacer was 90 nm, and $\Delta T$ was 10 K with $T_2$ set at 303.15 K.

As shown in Figure S16a, with the number of layers of Gr/SU8 increased, $\eta$ is always increasing. Better enhancement could be observed with more Gr/SU8 layers and would eventually converge to a limit, due to more graphene SPP coupling modes. Points in black-dashed squares are the theoretical targets in this work. $\eta$ of the Gr/SU8/5L heterostructures at $E_F = -0.11$ eV reaches $\sim 1189$, which is 1.21-fold larger than that of the Gr/SU8/1L heterostructures, and is $\sim 98\%$ of that of the Gr/SU8/infinite heterostructures. Without the contribution of the surface phonon polaritons (SPhPs) from the SiO$_2$ substrate, enhancement of the Gr/SU8/1L heterostructures could only reach $\sim 77\%$ of that of the Gr/SU8/infinite heterostructures. This enhancement difference is more pronounced with larger $E_F$. For example, when $E_F = -0.36$ eV, $\eta$ of the five-layer structure is $\sim 628$, which is 1.51-fold of that of the single-layer structure (on the SiO$_2$/Si substrate) and 1.76-fold of that without SiO$_2$/Si substrate (not shown in the figure). With more Gr/SU8 layers, the contributions of the SPhPs from the substrate will be highly reduced due to the rapid attenuation of the evanescent...
modes. The photon tunneling probability $\xi_p$ of each structure (on the SiO$_2$/Si substrate) at $E_F$ = -0.36 eV were plotted in Figure S16b-S16g to show the SPP coupling modes. For the Gr/SU8/1L heterostructure, near-unity $\xi_p$ enabled by coupled graphene SPPs from two interacting graphene sheets (of emitter and receiver) is restricted in two thin branches, which split at a lower $\beta$ and merge at a larger $\beta$ in a broad frequency range. $\xi_p$ is near-unity in the region where the dispersion curves (not shown here) of the resonance modes overlap with the contour plot, demonstrating the important role of coupled SPPs in enhancing near-field heat transfer. When the number of layers of Gr/SU8 increases, more coupled SPPs from more graphene sheets are excited and enable near-unity $\xi_p$ within more branches, e.g., four branches could be clearly observed in the Gr/SU8/2L heterostructures. Hence, in the Gr/SU8/5L heterostructures, ten branches with near-unity $\xi_p$ enabled by the coupled SPPs of ten layers of graphene sheets form an almost continuous region in the $k$-space, accounting for the greater enhancement of NFTR than that of other graphene-covered SU8 heterostructures with fewer layers. For comparison, Gr/SU8/infinite heterostructures with continuous near-unity $\xi_p$ (within the mid- and far-infrared frequency region) was also calculated.
Figure S16. a) Enhancement normalized with the corresponding BB limit at a fixed gap distance $d = 50$ nm for Gr/SU8/1L, Gr/SU8/2L, Gr/SU8/3L, Gr/SU8/4L, Gr/SU8/5L, and Gr/SU8/infinite heterostructures at $E_F = -0.11$ eV and -0.36 eV, respectively. b-g) show the corresponding contour plots of $\xi_\phi$. 
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