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Nonlocal Near-field Radiative Heat Transfer by Transdimensional Plasmonics

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Using transdimensional plasmonic materials (TDPM) within the framework of fluctuational electrodynamics, we demonstrate nonlocality in dielectric response alters near-field heat transfer at gap sizes on the order of hundreds of nanometers. Our theoretical study reveals that, opposite to the local model prediction, propagating waves can transport energy through the TDPM. However, energy transport by polaritons at shorter separations is reduced due to the metallic response of TDPM stronger than that predicted by the local model. Our experiments conducted for a configuration with a silica sphere and a doped silicon plate coated with an ultrathin layer of platinum as the TDPM show good agreement with the nonlocal near-field radiation theory. Our experimental work in conjunction with the nonlocal theory has important implications in thermophotovoltaic energy conversion, thermal management applications with metal coatings, and quantum-optical structures.

Near-field radiative heat transfer has attracted enormous interest for energy conversion [1, 2], solid-state cooling [3, 4], and thermal management [5, 6] owing to the substantial radiation enhancement beyond Planck’s law at nanoscale gaps. Dielectric response of materials essentially governs near-field thermal radiation due to its relationship with absorption (and emission based on Kirchoff’s law). Based on fluctuational electrodynamics, near-field heat transfer theory generally relies on local dielectric properties and predicts continuously enhancing heat flux with a decreasing gap, d [3, 7–12]. In the extreme nanometer-sized gaps ($d < 10$ nm), however, necessity of a nonlocal description arises to obtain non-divergent heat fluxes [13]. Thus, so far, theoretical attempts [14–17] to explain nonlocality have focused on the gap sizes in the extreme near-field. Also, recent experimental studies [18–20] reveal inconclusive results for nonlocality. However, when considering thin plasmonic films there is another nonlocal effect which may alter the heat flux even at separation gaps of tens or hundreds of nanometers. Here, we provide an experimental proof of this nonlocality arising from plasmonic thin films with a thickness, t , of extreme nanometer-sized dimensions ($t < 10$ nm).

To control near-field thermal radiation, experimental studies [21–23] have so far mainly examined conventional bulk materials and thin films. Recent advancements in the fields of nanophotonics and plasmonics uncover that atomically thin plasmonic layers [24, 25], also known as transdimensional plasmonic materials (TDPMs) [26], show unusual tunability in optical effects [27], and light-matter interactions [28]. In theory [29, 30], out-of-plane confinement in an ultrathin metal film yields strong spatial dispersion and nonlocality [31–33] of dielectric response. While the thickness-dependent dielectric response due to the nonlocal effect provides a new degree of freedom for tunability [34–37], TDPMs are largely unexplored for controlling nanoscale thermal radiation.

Here, we examine both local and nonlocal dielectric responses of TDPMs within the framework of fluctuational electrodynamics. Based on a sensitive experimental platform capable of probing near-field signals as small as 0.1-1 nW at a gap of ~ 10 nm, we measure near-field radiative heat transfer between a silica sphere and a p-doped silicon substrate coated with an atomically thin layer of platinum (Pt) as TDPM, as shown in Fig. 1(a). Our experimental results show unprecedented dependence to nonlocal effects in the near-field heat flux at separations over a large range from tens of nanometers to micrometer gap sizes.

As shown in Fig. 1(a), we consider structure 1 as a bare planar semi-infinite medium (large sphere) and structure 2 as a multilayered planar structure consisting of a semi-infinite medium and a coated TDPM with a thickness t . Heat flux between the structures at temperatures T_1 and T_2 with a separation gap, d , is expressed by

$$q = \int_0^\infty \frac{d\omega}{2\pi} [\Theta(\omega, T_1) - \Theta(\omega, T_2)] \int_0^\infty \frac{d\kappa}{2\pi} \kappa \mathcal{T}(\omega, \kappa). \quad (1)$$

Here, $\Theta(\omega, T)$ is the mean energy of oscillators with frequency ω at temperature T and is expressed by $\hbar\omega / (e^{\hbar\omega/k_B T} - 1)$, where \hbar and k_B represent the reduced Planck constant and the Boltzmann constant, respectively. κ denotes the wave vector component parallel to the planar surface. $\mathcal{T}(\omega, \kappa)$ represents the transmission probability of propagating ($\kappa < k_o = \omega/c$) and evanescent ($\kappa > k_o$) waves across the gap, and $\mathcal{T}(\omega, \kappa) \in [0, 1]$. In the near-field, the probability reads [38–40] (see Section 5 in [41] in the far-field)

$$\mathcal{T}(\omega, \kappa) = \frac{4\text{Im}(r_1)\text{Im}(r_2)}{|1 - r_1 r_2 e^{2ik_z d}|^2} e^{-2\text{Im}(k_z) d}. \quad (2)$$

Equation (2) shows that energy carried by the evanescent waves depends on reflection coefficient, r_i with the imaginary component $\text{Im}(r_i)$, for the planar structure i (see

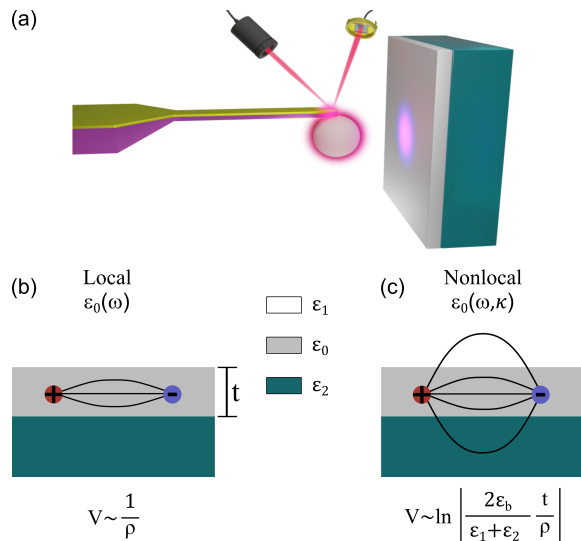


FIG. 1. (a) Schematic of the heat-exchanging structures (a large sphere medium, modelled as a plate for theoretical consideration, and a semi-infinite medium, dark teal, coated with an atomically thin metal film as the TDPM, light gray). Modelling of charge interactions based on the local (b) and nonlocal (c) approaches in a thin metal film, ϵ_0 , sandwiched between vacuum, ϵ_1 , and substrate, ϵ_2 . The local response assumes potential V is inversely proportional to the charge separations, $\sim 1/\rho$, as in a bulk whereas, based on the nonlocal model, $V \sim \ln \left| \frac{2\epsilon_b}{\epsilon_1 + \epsilon_2} \frac{t}{\rho} \right|$. Here, ϵ_b denotes background response of the film.

Section 4 in [41]) and the separation gap. These waves can be frustrated or surface waves. Energy of frustrated waves decays in proportion to $e^{-2\text{Im}(k_z)d}$ and completely attenuates for $\text{Im}(k_z) \gg (2d)^{-1}$. For surface waves, i.e. surface phonon and plasmon polaritons, the coupling of resonant waves between the interfaces across the gap, accounted by $|1 - r_1 r_2 e^{2ik_z d}|^2$, compensates the exponential decay in a larger wavevector range so that $\mathcal{T}(\omega, \kappa)$ can have values even close to 1 for large κ .

Our understanding of dielectric response relies on local and nonlocal approaches. In this study, dielectric responses of both semi-infinite media are considered as local. Nonlocal response is only examined for TDPMs. The local model of TDPMs is derived based on the Drude model with classical equations of motion, where electromagnetic force acting on free electron clouds is uniform in the entire bulk medium (Fig. 1(b)). Figure 2(a) shows the real component of the dielectric permittivity, ϵ'_0 , of a Pt ultrathin film based on the local model as a function of ω and κ (see the Supplemental Material in [41] for local dielectric properties). Frequency dependent ϵ'_0 varies from $\sim -1 \times 10^3$ at the high frequencies to $\sim -4 \times 10^3$ at the low frequencies. Figure 2(a) also reveals that the local response is independent of κ . Thus, dielectric response to a wave at a given frequency is independent of wave condition (propagating or evanescent in free space).

On the other hand, the quasi-classical approach [42]

based on the Keldysh-Rytova (KR) potential suggests a nonlocal model, and differs from the Coulomb potential in that the out-of-plane confinement in an atomically thin film is considered when dielectric constants of substrate ϵ_1 and superstrate ϵ_2 surrounding the film, as shown in Fig. 1(c), are smaller than background dielectric response of the film, ϵ_b . Then, the Coulomb potential loses the out-of-plane dependence and becomes a purely in-plane 2D potential with the only remnant of the out-of-plane coordinate being the layer thickness to represent the size of the vertical confinement (Fig. 1(c)). Consequently, Fourier transform of the KR potential results in wavevector, κ , dependence. Applying the κ -dependent KR potential to the equation of motion of electrons returns a κ -dependent plasma frequency, resulting in a confinement-induced nonlocal Drude dielectric response. The KR potential varies with thickness due to change in the electrostatic interaction strength of electrons across the boundaries, and the plasma frequency of TDPMs becomes thickness dependent [27]. (see the Supplemental Material, Section 1 in [41] for nonlocal dielectric properties).

To demonstrate the effect of nonlocality in dielectric function, we select a thin Pt film as the TDPM sandwiched between vacuum and a p-doped silicon substrate and plot real component, ϵ'_0 , of dielectric function in terms of ω and κ for thicknesses of 2, and 10 nm in Figs. 2(b) and (c), respectively (see Fig. S1 in [41] for the imaginary component). Based on the nonlocal model, dielectric responses exhibit both frequency and wavevector dependencies. The major frequency dependence arises from the comparable condition of ϵ_b , ϵ_1 , and ϵ_2 at around $\omega \sim 1.5 \times 10^{14}$ where $\epsilon'_0 \sim 0$. The contour lines at the center, i.e. the line pointed in Fig. 2(b), represent $\epsilon'_0 = 0$ and divide Figs. 2(b) and (c) into two regions. Across the lines, ϵ'_0 changes its sign from negative at the high frequencies to positive at the low frequencies, meaning that the optical response transitions from a metallic (metallic region) to dielectric response (dielectric region), respectively. Red dashed lines indicate the light line ($\kappa = k_o$). As κ varies from propagating ($\kappa < k_o$) to evanescent ($\kappa > k_o$) wave vectors for a given thickness, the strength of the response, related to absolute magnitude of the dielectric function, $|\epsilon'_0|$, increases. We also recognize that with the increasing thickness, the strength of optical response increases for a given frequency and wavevector. Still, the responses for evanescent waves are stronger as compared to those of propagating waves. The comparison of Fig. 2(a) with Figs. 2(b) and (c) reveals that both the local and nonlocal models predict approximately the same strength of response for large wavevector ranges (evanescent waves). Conversely, for all thicknesses, dielectric response of atomically thin Pt over the wavevector range of propagating waves ($\kappa < k_o$) is weaker than that for bulk Pt.

Nonlocal and local transmission probabilities demon-

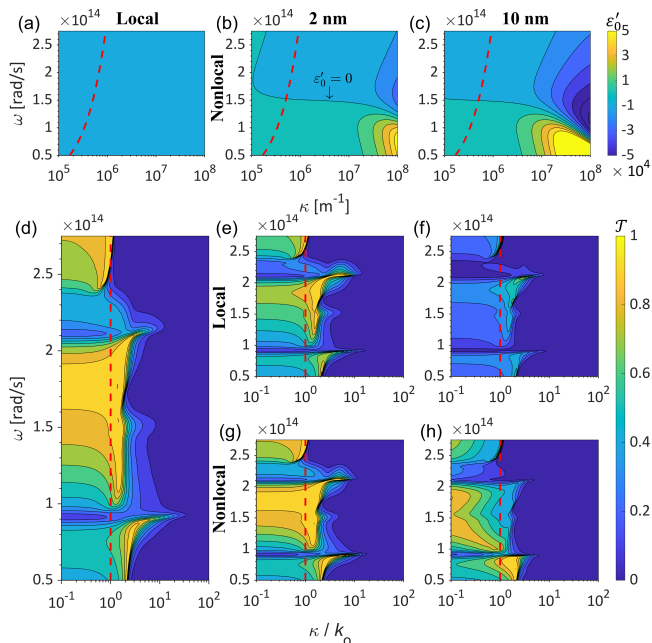


FIG. 2. Dielectric functions of Pt based on the local and nonlocal models. Real component of dielectric function, ε'_0 , for Pt based on the local (a), $\varepsilon_0(\omega)$, and nonlocal (b)-(c), $\varepsilon_0(\omega, \kappa)$, model. The permittivity based on the nonlocal model exhibits strong κ -dependence for a thickness of 2 (b), and 10 (c) nm. In (b) and (c), contour lines are separated by 1×10^4 and the contour line at the very center of each panel corresponds to $\varepsilon'_0 = 0$, also pointed in (b). The colormap scales ε'_0 . Transmission probability, $\mathcal{T}(\omega, \kappa)$, across a 100-nm gap between a silica and a bare p-doped silicon (d), a silica and a p-doped silicon coated with a thin layer of Pt based on the local (e)-(f) and nonlocal (g)-(h) dielectric responses with respect to ω and κ/k_o .

strate strong contrast in energy transport capability of propagating and evanescent waves. Figures 2(d)-(h) show the transmission probability as a function of ω and κ over both propagating and evanescent ranges for p-polarized waves at a separation gap of 100 nm between a silica plate and a p-doped silicon plate with and without Pt TDPM coating (see Fig. S7 in [41] for the transmission probability of s-polarized waves). We first consider the case without TDPM coating and plot the probability in Fig. 2(d). Within the evanescent wavevector range ($\kappa/k_o > 1$), modes with large wavevectors ($\kappa/k_o \sim 2$) excited at two narrow frequency bands around $\omega \sim 2 \times 10^{14}$ and $\omega \sim 1 \times 10^{14}$ rad.s $^{-1}$ carry most energy. These sharp peaks arise from SPhPs supported by silica and coupled across the separation gap. Also, frustrated waves ($\kappa/k_o \sim 1$) contribute to energy transport over a broad frequency range.

Now, we compare the transmission probabilities when a layer of Pt TDPM is considered on the silicon substrate. First, the transmission probabilities are plotted based on the local dielectric responses of TDPM with 2-, 5-, and 10-nm thicknesses in Figs. 2(e) and (f), respec-

tively. From the comparison of Fig. 2(e) with Fig. 2(d), we point out that the presence of 2-nm-thick Pt diminishes the energy carried by propagating waves. Energy transport by SPhPs is mostly preserved. Increasing the film thickness to 10 nm almost prevents propagating and frustrated waves from contributing to energy transport. The local model predicts that, for propagating waves, optical response of a thin Pt layer is same with that of a perfect mirror, i.e, strong reflection (see Fig. S3 in [41] for plots of reflection coefficients of propagating waves). Similarly, the coupling strength of SPhPs weakens with the presence of 10-nm Pt. With the thickness, $Im(r_2)$ (see Fig. S4 in [41]) approaches zero for the assembled structure, resulting in fast decaying $\mathcal{T}(\omega, \kappa)$.

The nonlocal transmission probability differs from the local one due to its wave condition (propagating or evanescent) dependence. We compare the bottom panels (Figs. 2(g)-(h)) with the top panels (Figs. 2(e)-(f)) for a given thickness. Propagating waves carry a higher fraction of energy across the gap based on the nonlocal model because the nonlocal dielectric permittivity is weaker than that of the local permittivity in the corresponding wavevector range (see Figs. 2(a)-(c)). This indicates that based on the nonlocal response, the Pt film is more transparent to propagating waves and has minimal interference with energy transport capability of these waves across silica and silicon plates. In addition, we observe that the difference in energy transported by SPhPs (two major peaks) depends on the frequency ranges. The wavevector range of SPhPs at $\omega \sim 2 \times 10^{14}$ shrinks with a greater rate with thicker TDPM based on the nonlocal model because the strength of metallic response gets stronger over broader wavevector range (see the metallic region in Figs. 2(b)-(c)) and exceeds that of the local model. In contrast, SPhPs at $\omega \sim 1 \times 10^{14}$ mainly preserve their energy transport capability compared to that of the local model. The reason is that the nonlocal TDPM film treats as a dielectric medium in the corresponding frequency and wavevector ranges (dielectric region as defined for Figs. 2(b)-(c)), and interferes less with the coupling of SPhP.

To examine the effect of nonlocality to near-field thermal radiation, we compute near-field heat transfer coefficient, $h \sim q/\Delta T$. Figure 3 shows the coefficient between silica and p-doped silicon coated with 2-, 5-, 10-, and 100-nm-thick TDPM films as a function of gap size. Both the local and nonlocal models estimate increasing heat transfer coefficient with respect to separation gap due to the evanescent wave contribution. The enhancement for a given separation gap decreases with the increase in film thickness based on both models. Comparison of nonlocal estimations with local ones indicates that the nonlocal near-field radiation predicts higher heat transfer for larger separation gaps because of the higher transmission probability for propagating and frustrated waves as discussed for Fig. 2. On the other hand, the nonlocal pre-

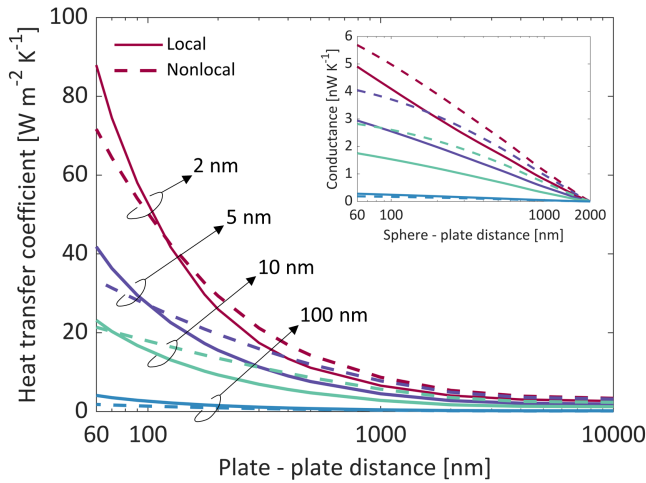


FIG. 3. Theoretical predictions of near-field radiation based on both the local and nonlocal models. Inset shows computed near-field conductance between the silica sphere and the silicon plate obtained using the Derjaguin approximation for the corresponding configurations. For later comparison with experiment, the numerical results are shifted such that the conductance is zero at a distance of 2 μm .

diction is lower at separation gaps below a certain gap distance. This observation is consistent with the theoretical results in literature [43]. The lower heat flux arises because at shorter separations, waves with large κ values dominate the heat transfer and for the corresponding κ range, the corresponding dielectric response is even stronger than that of the local model. Thus, the coupling of polaritons between silica and p-doped silicon surfaces is weaker for large κ values. We also plot the theoretical near-field conductance between a silica sphere and a p-doped silicon plate based on the two models using the Derjaguin approximation. The inset of Fig. 3 reveals that the difference between the estimations of the two models is prominent for the sphere-plate configuration. We note that the nonlocal model is not only showing higher conductances than the local model as expected, but it also has the tendency of a small “saturation effect” for very small distances. The reason is that the heat flux for small gaps is smaller in the nonlocal model than in the local one as observed in Fig. 3.

To demonstrate the nonlocal effect from TDPMs, we conduct experiments for the near-field thermal radiation between a silica sphere and a p-doped silicon uncoated or coated with atomically thin Pt films of 2-, 5-, and 100-nm thickness. The schematic and the picture of our experimental platform are shown in Figs. 1(a) and 4(a), respectively. This platform was also employed in our previous work [44, 45]. The inset and Fig. 4(b) show the real-time and the corresponding position-dependent deflection signal for the case with the uncoated substrate. The deflection signal monitors the near-field induced change (see Section 11 in [41] for details of the experiment). The Pt

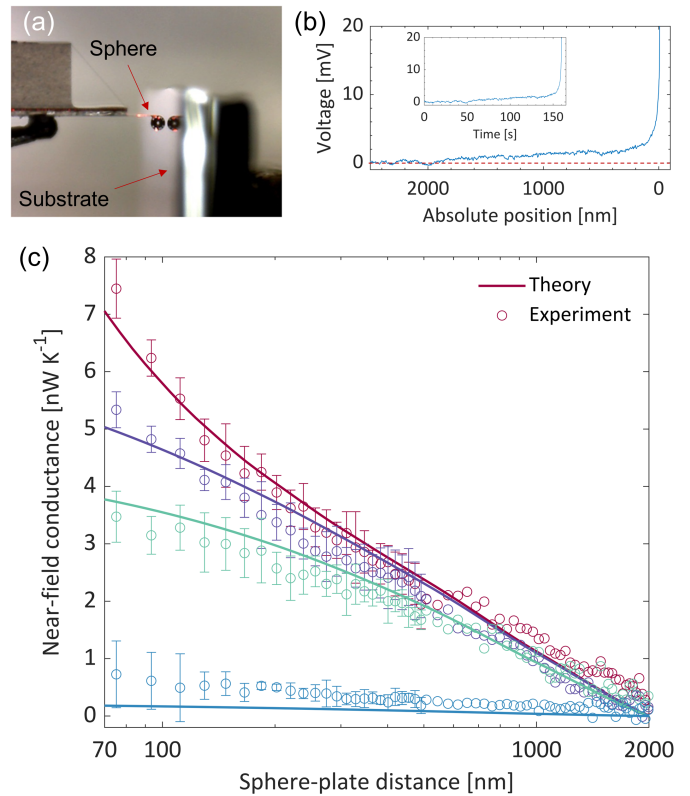


FIG. 4. Near-field experimental results. (a) Picture of the silicon substrate and the silica microsphere. A red laser shines on the bi-material cantilever. (b) Position-dependent deflection signals converted from real-time signals acquired using the optical measurement system and shown in the inset. (c) Measured near-field conductance as a function of gap for the cases without (red) and with ~ 2 nm (purple), ~ 5 nm (light green) and ~ 100 nm (cyan) ultrathin films, compared to non-local theoretical predictions obtained using the Derjaguin approximation.

thin films are coated on the silicon substrates by sputtering. By lowering the plasma power, the deposition rate of Pt can be adjusted to be as low as $\sim 1\text{\AA}$ per second. We perform AFM measurements to characterize the roughness (see Section 7 in [41]) and find RMS roughnesses of ~ 0.1 nm, ~ 0.2 nm, and ~ 0.4 nm for silicon without coating and with ~ 2 nm and ~ 5 nm coatings, respectively. The near-field measurements are carried out under a vacuum level of 1×10^{-6} Torr.

We plot experimental results of the near-field conductance measurements for the cases with the bare silicon plate and the silicon plates with atomically thin layers of ~ 2 nm, ~ 5 nm, and ~ 100 nm in Fig. 4(c), showing that experimental results agree very well with the nonlocal near-field radiation predictions (see Fig. S9(b) in [41] for the estimation based on the local model and Figs. S10(a) and (b) in [41] for the sensitivity analysis of the local and nonlocal models when the film thickness is shifted by ± 0.5 nm). As the highest conductance is

obtained in the silica-bare silicon case, the conductance decreases with increasing thickness at a given separation gap. Theoretical predictions based on the local model consistently underestimate the experimental results for the cases with ~ 2 - and ~ 5 -nm-thick TDPMs. We expect with the increasing thickness, dielectric response of thin films approaches bulk response. For ~ 100 -nm-thick film, the signal is buried in noise, as predicted by both models.

In addition to the quantum nanophotonics applications already reported [31–37], we propose two possible methods to make use of TDPMs in near-field based applications. First, the metallic and dielectric response over distinct frequency ranges obtained with TDPMs can be exploited for filtering near-field thermal radiation. This filtering may control the emission peak frequency to match with the bandgap of the photovoltaic cells, reducing waste heat and increasing device energy conversion efficiency. Second, temperature variation may change thickness of TDPMs. Relying on temperature dependent dielectric properties, near-field thermal diodes may utilize thickness sensitivity of dielectric response from TDPMs to temperature variation for adjusting the contrast between reverse and forward biases.

In summary, we have shown that the nonlocal near-field radiative heat transfer alters thermal radiation at submicron separation sizes. Our theoretical calculations indicate that the thickness and wavevector dependent dielectric response of TDPMs modifies near-field thermal radiation between silica and doped silicon dominated by surface phonon polariton contributions. Our experiments have found that the local prediction underestimates the measured results, and verified the nonlocal near-field thermal radiation. We expect that our nonlocal consideration will change the focal point of studies about the nonlocal near-field thermal radiation from extreme near-field regions to tens and even hundreds of nanometer gap sizes. The nonlocal effect based on TDPMs provides a new degree of freedom for controlling near-field radiation, which has important implications in energy conversion and thermal management. **We also foresee that material dimensions much smaller than the characteristic wavelength in both lateral and normal directions will give rise to new phenomena and applications on near-field thermal radiation.**

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