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TECHNICAL REPORT

Nanoscale Plasmonic Emitters for Ultra-High Intensity Thermal Pulse Generation

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14. ABSTRACT The objective of this project is to address the key challenges in simulating thermal pulses of nuclear outputs at small scales by developing ultra-high intensity plasmonic thermal emitters. Although plasmonic or nanophotonic structures, such as plasmonic antennas, resonant metamaterials, and photonic crystal cavities, have been identified as a common paradigm for realizing spectrally tunable and high-intensity thermal radiation, there is no general theoretical framework for designing and maximizing thermal radiation from an arbitrary nanophotonic structure.					
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Final Report**Grant/Award #:** HDTRA1-19-1-0028**PI Name:** Sheng Shen**Organization/Institution:** Carnegie Mellon University**Project Title:** Nanoscale Plasmonic Emitters for Ultra-High Intensity Thermal Pulse Generation**What are the major goals of the project?**

List the major goals of the project as stated in the approved application or as approved by the agency. If the application lists milestones/target dates for important activities or phases of the project, identify these dates and show actual completion dates or the percentage of completion. Generally, the goals will not change from one reporting period to the next. However, if the awarding agency approved changes to the goals during the reporting period, list the revised goals and objectives. Also explain any significant changes in approach or methods from the agency approved application or plan.

The objective of this project is to address the key challenges in simulating thermal pulses of nuclear outputs at small scales by developing ultra-high intensity plasmonic thermal emitters. Although plasmonic or nanophotonic structures, such as plasmonic antennas, resonant metamaterials, and photonic crystal cavities, have been identified as a common paradigm for realizing spectrally tunable and high-intensity thermal radiation, there is no general theoretical framework for designing and maximizing thermal radiation from an arbitrary nanophotonic structure. As a result, the limit of using plasmonic or nanophotonic structures for generating thermal pulses still remains unclear. In this project, we propose a combined theoretical and experimental effort to develop ultra-high intensity plasmonic thermal emitters that can potentially simulate thermal pulses generated in a nuclear explosion but at extremely small scales.

In this project, we will design and fabricate nanoscale plasmonic emitters for generating ultra-high intensity thermal signals, which can be utilized to simulate thermal pulses of nuclear outputs at small scales. Specifically, we will develop a new theory to study the limit of thermal output power from an arbitrary plasmonic emitter, and to design novel plasmonic thermal emitters to achieve ultra-high intensity. We will also fabricate the as-designed plasmonic emitters using refractory metals and graphene, and demonstrate their high intensity in both near- and far-fields. In the 3-year base period, we will focus on the theory, design, fabrication and characterization of the proposed ultra-high intensity plasmonic thermal emitters. In the 2-year optional period, we will test the dynamic behaviors of the plasmonic thermal emitters, and measure their overall near-field thermal responses.

What was accomplished under these goals?

For this reporting period describe: 1) major activities; 2) specific objectives; 3) significant results, including major findings, developments, or conclusions (both positive and negative); and 4) key outcomes or other achievements. Include a discussion of stated goals not met. As the project progresses, the emphasis in reporting in this section should shift from reporting activities to reporting accomplishments.

1. Scale law of far-field thermal radiation from plasmonic metasurfaces [Physical Review Letters, 124, 137401, 2020.]

We developed a new scale law to predict and design the thermal emission from a metasurface based on quasi-normal mode (QNM) theory and the tight-binding (TB) method. In this new theory, the thermal emission from one individual sub-wavelength emitter in a metasurface was described by QNM theory, whereas the TB method was employed to quantify the red shift of the thermal emission spectrum due to the interactions between the emitters. Based on this new scale law, we can quantitatively describe the suppression of thermal emission from a single plasmonic emitter when coupled with adjacent emitters. We found that depending on the QNM properties of a single emitter, the overall far-field thermal radiation from the metasurface can be either positively or negatively correlated with the packing density of the emitters. We also experimentally verified the new scale law by fabricating and measuring plasmonic metasurfaces made by metal nanorods. Hence, this new scale law could perform as a general theoretical framework for designing metasurfaces with desired thermal emission properties.

QNMs, which are the eigen solutions to the source free Maxwell equation in the non-Hermitian system, were introduced to quantitatively describe the thermal radiation from a single plasmonic thermal emitter. The TB method was first introduced to calculate the electronic band structure based upon the superposition of wave functions for isolated atoms. Here we adopted the same methodology to account for the modification of the single emitter's QNM inside the array, provided that the electric field of a plasmonic emitter is highly confined around its surface. Due to the translational symmetry of the plasmonic emitter array in a metasurface, the collective QNM of the entire emitter array should follow the form of Bloch's wave. Therefore, the QNM is a periodic function of the lattice constant of the emitter array $\mathbf{R}_l = l_x P_x \hat{\mathbf{e}}_x + l_y P_y \hat{\mathbf{e}}_y$, which are $\mathbf{E}_n(\omega(\mathbf{k}), \mathbf{r}) = \frac{1}{\sqrt{N}} \sum_l \mathbf{a}_n(\mathbf{R}_l, \mathbf{r}) e^{i\mathbf{k} \cdot \mathbf{R}_l}$, $\mathbf{H}_n(\omega(\mathbf{k}), \mathbf{r}) = \frac{1}{\sqrt{N}} \sum_l \mathbf{b}_n(\mathbf{R}_l, \mathbf{r}) e^{i\mathbf{k} \cdot \mathbf{R}_l}$. l_x and l_y here are the integers. N is the number of emitters considered. The coefficients $\mathbf{a}_n(\mathbf{R}_l, \mathbf{r})$ and $\mathbf{b}_n(\mathbf{R}_l, \mathbf{r})$ are two sets of Wannier functions which only depend on $\mathbf{r} - \mathbf{R}_l$.

Since the QNM of a plasmonic emitter is highly confined around its surface, we assumed that the QNM of the entire emitter array behaves similarly in the vicinity of each emitter as it does in the case of a standalone single emitter. Therefore we can use the QNM of a single emitter $\{\mathbf{e}_n(\omega_n, \mathbf{r}), \mathbf{h}_n(\omega_n, \mathbf{r})\}$ as the approximation of $\mathbf{a}_n(\mathbf{r} - \mathbf{R}_l)$ and $\mathbf{b}_n(\mathbf{r} - \mathbf{R}_l)$, namely: $\mathbf{E}_n(\omega(\mathbf{k}), \mathbf{r}) = \frac{1}{\sqrt{N}} \sum_l \mathbf{e}_n(\omega_n, \mathbf{r} - \mathbf{R}_l) e^{i\mathbf{k} \cdot \mathbf{R}_l}$, $\mathbf{H}_n(\omega(\mathbf{k}), \mathbf{r}) = \frac{1}{\sqrt{N}} \sum_l \mathbf{h}_n(\omega_n, \mathbf{r} - \mathbf{R}_l) e^{i\mathbf{k} \cdot \mathbf{R}_l}$. Here, $\{\mathbf{e}_n(\omega_n, \mathbf{r}), \mathbf{h}_n(\omega_n, \mathbf{r})\}$ is the solution to the source free Maxwell equation for a stand-alone emitter:

$$\begin{aligned} \nabla \times \mathbf{e}_n(\omega_n, \mathbf{r}) &= i\omega_n \mu_0 \mathbf{h}_n(\mathbf{r}) \\ \nabla \times \mathbf{h}_n(\omega_n, \mathbf{r}) &= -i\omega_n \varepsilon_s(\omega_n, \mathbf{r}) \mathbf{e}_n(\mathbf{r}) \end{aligned} \quad (1.1)$$

where $\varepsilon_s(\omega_n, \mathbf{r})$ is the permittivity function for the emitter. In order to satisfy the boundary conditions of Maxwell's equation, the coefficients of the Bloch-form QNM of the metasurface should be modified as the following form: $\tilde{\mathbf{e}}_n(\omega_n, \mathbf{r} - \mathbf{R}_l) = \mathbf{e}_n(\omega_n, \mathbf{r} - \mathbf{R}_l)\gamma(\omega_n, \mathbf{r} - \mathbf{R}_l)$, $\tilde{\mathbf{h}}_n(\omega_n, \mathbf{r} - \mathbf{R}_l) = \mathbf{h}_n(\omega_n, \mathbf{r} - \mathbf{R}_l)\gamma(\omega_n, \mathbf{r} - \mathbf{R}_l)$, where $\gamma_{\omega_n, l} = \gamma(\omega_n, \mathbf{r} - \mathbf{R}_l) = 1/\sum_l \varepsilon_s(\omega_n, \mathbf{r} - \mathbf{R}_l)$ and ε_s here is defined inside the repeating unit cell indexed by the lattice constant (l_x, l_y) . Thus, the new Bloch-form QNM satisfies the continuous boundary conditions at the interface of the emitter and the background material.

Substituting the QNM of the entire emitter array into the source-free Maxwell equation $\nabla \times \mathbf{H}_n(\mathbf{r}) = -i\omega(\mathbf{k})\varepsilon_t(\omega(\mathbf{k}), \mathbf{r})\mathbf{E}_n(\mathbf{r})$, we have

$$\begin{aligned} & \sum_l \frac{1}{\sqrt{N}} \nabla \times \mathbf{h}_n(\omega_n, \mathbf{r} - \mathbf{R}_l) e^{i\mathbf{k} \cdot \mathbf{R}_l} \gamma_{\omega_n, l} \\ &= \frac{1}{\sqrt{N}} \sum_l e^{i\mathbf{k} \cdot \mathbf{R}_l} \gamma_{\omega_n, l} (-i\omega_n \varepsilon_s(\omega_n, \mathbf{r} - \mathbf{R}_l) \mathbf{e}_n(\mathbf{r} - \mathbf{R}_l)) \\ &= \frac{1}{\sqrt{N}} \sum_l -i\omega(\mathbf{k}) \varepsilon_t(\omega(\mathbf{k}), \mathbf{r}) \mathbf{e}_n(\omega_n, \mathbf{r} - \mathbf{R}_l) e^{i\mathbf{k} \cdot \mathbf{R}_l} \gamma_{\omega_n, l} . \end{aligned} \tag{1.2}$$

Note that $\varepsilon_t(\omega(\mathbf{k}), \mathbf{r})$ is the permittivity function of the emitter array which is related to the single emitter permittivity function by $\varepsilon_t(\omega, \mathbf{r}) = \sum_l \varepsilon_s(\omega, \mathbf{r} - \mathbf{R}_l)$. Now we obtain the new equation between the modified resonant frequency $\omega(\mathbf{k})$ and the resonant frequency of the single emitter ω_n :

$$\sum_l e^{i\mathbf{k} \cdot \mathbf{R}_l} \gamma_{\omega_n, l} \omega_n \varepsilon_s(\omega_n, \mathbf{r} - \mathbf{R}_l) \mathbf{e}_n(\omega_n, \mathbf{r} - \mathbf{R}_l) = \sum_l e^{i\mathbf{k} \cdot \mathbf{R}_l} \gamma_{\omega_n, l} \omega(\mathbf{k}) \varepsilon_t(\omega(\mathbf{k}), \mathbf{r}) \mathbf{e}_n(\omega_n, \mathbf{r} - \mathbf{R}_l). \tag{1.3}$$

Multiplying both sides of Equation (1.3) with $\mathbf{e}_n(\omega_n, \mathbf{r} - \mathbf{R}_{l'})$ and integrating over the whole emitter array volume, we get:

$$\sum_l \int d\mathbf{r}^3 e^{i\mathbf{k} \cdot \mathbf{R}_l} \gamma_{\omega_n, l} \omega_n \varepsilon_s(\omega_n, \mathbf{r} - \mathbf{R}_l) \mathbf{e}_{n, l} \mathbf{e}_{n, l'} = \sum_l \int d\mathbf{r}^3 e^{i\mathbf{k} \cdot \mathbf{R}_l} \omega(\mathbf{k}) \gamma_{\omega_n, l} \varepsilon_t(\omega(\mathbf{k}), \mathbf{r}) \mathbf{e}_{n, l} \mathbf{e}_{n, l'} , \tag{1.4}$$

where $\mathbf{e}_{n, l} = \mathbf{e}_n(\omega_n, \mathbf{r} - \mathbf{R}_l)$. If the distance between adjacent emitters is not significantly smaller than the lateral dimension of the emitters and the index l' is set to be 0, then the left-hand side (LHS) of Equation (1.4) is reduced to $\sum_l \int d\mathbf{r}^3 e^{i\mathbf{k} \cdot \mathbf{R}_l} \gamma_{\omega_n, l} \omega_n \varepsilon_s(\omega_n, \mathbf{r} - \mathbf{R}_l) \mathbf{e}_{n, l} \mathbf{e}_{n, 0} = \int d\mathbf{r}^3 \gamma_{\omega_n, 0} \omega_n \varepsilon_s(\omega_n, \mathbf{r}) \mathbf{e}_{n, 0} \mathbf{e}_{n, 0}$. Finally, the modified resonant QNM frequency of the emitter array can be calculated by solving the equation:

$$\sum_l \int dr^3 e^{i\mathbf{k} \cdot \mathbf{R}_l} \gamma_{\omega_n, l} \omega(\mathbf{k}) \varepsilon_t(\omega(\mathbf{k}), \mathbf{r}) \mathbf{e}_{n, l} \mathbf{e}_{n, l'} = \int dr^3 \gamma_{\omega_n, 0} \omega_n \varepsilon_s(\omega_n, \mathbf{r}) \mathbf{e}_{n, 0} \mathbf{e}_{n, 0} . \quad (1.5)$$

The index l only needs to run over the nearby emitters to obtain the accurate result. If we only consider the coupling effect from the nearest emitters, the modified resonant frequency for the 2D array of emitters in a metasurface can be calculated as follows:

$$\omega(\mathbf{k}) = \frac{I_0^s}{I_0^t + 2\cos(\mathbf{k} \cdot \mathbf{R}_0) I_{1,x}^t + 2\cos(\mathbf{k} \cdot \mathbf{R}_0) I_{1,y}^t} \omega_n . \quad (1.6)$$

The integral $I_l^s = \int dr^3 e^{i\mathbf{k} \cdot \mathbf{R}_l} \gamma_{\omega_n, l} \varepsilon_s(\omega_n, \mathbf{r}) \mathbf{e}_{n, 0} \mathbf{e}_{n, l}$, $I_l^t = \int dr^3 e^{i\mathbf{k} \cdot \mathbf{R}_l} \gamma_{\omega_n, l} \varepsilon_t(\omega(\mathbf{k}), \mathbf{r}) \mathbf{e}_{n, 0} \mathbf{e}_{n, l}$. The integral $I_{1,x}^t$ and $I_{1,y}^t$ are mode overlapping terms from the adjacent emitters in the x-direction (shoulder to shoulder) and y-direction (head to head) respectively. Equation (1.6) could be solved iteratively. If the mode overlapping terms $I_{1,x}^t$ and $I_{1,y}^t$ are small as compared to the zero-order coupling term I_0^t , then $\omega(\mathbf{k}) = \omega = \omega_n I_0^s / I_0^t$. In this case, the resonant frequency has no dispersion with respect to the lattice momentum $\mathbf{k}$. For the nontrivial scenario where the coupling integral I_l^s is no longer negligible, the energy of the resonant photons forms a band, and the far-field thermal radiation from the emitter array needs to be calculated within the first Brillion zone, together with the density of states and the Bose-Einstein distribution term.

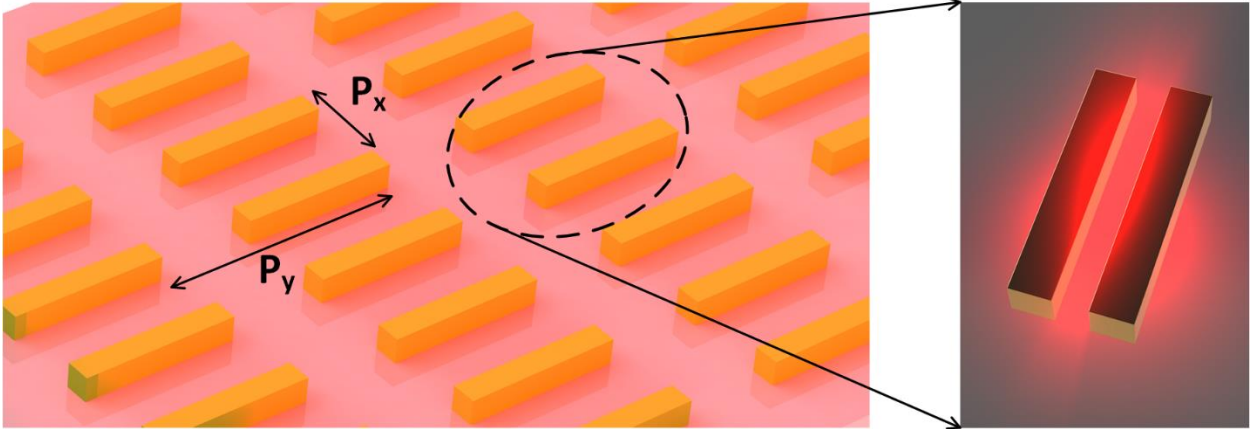


Figure 1.1 (a) Schematic of an array of resonant thermal emitters in a plasmonic metasurface, and (b) Zoomed-in image of two coupled adjacent emitters.

For a metasurface (Fig. 1.1) made from an array of plasmonic emitters with P_x and P_y as the periodicities in the x- and y-directions, we assume that there exists a unit cell including $N+1$ emitters, where when the center emitter is thermally excited, the rest of N emitters are coupled to it, and the coupling of the emitters outside this unit cell can be neglected. We follow the same mathematical derivation in our previous work to rewrite the far-field thermal radiation from the unit cell. Note that the QNM here is essentially different from that of a single emitter. As an approximation, we use the QNM of a single emitter to calculate the coupling strength in the emitter array. The modification of the resonant frequency is addressed by the TB method. We can express

the far-field thermal radiation from one emitter inside the array in terms of the fractional losses D_E , D_A and D_∞ from the single emitter's QNM profile yet at the modified resonant frequency ω .

$$\phi_\infty(\omega) = 4 \left(\frac{D_E}{D_\infty + D_E + \sum_{i=1}^N D_{A_i}} F \right) \left(P - \frac{D_E + \sum_{i=1}^N D_{A_i}}{D_\infty + D_E + \sum_{i=1}^N D_{A_i}} F \right), \quad (1.7)$$

where $D_E = \int_{V_E} dr^3 \frac{1}{2} \sigma_E \text{Re}(\omega) |E(r)|^2$, $D_\infty = \int_{\partial V} dr^2 \frac{1}{2} \text{Re}(E(r) \times H^\dagger(r))$, and

$D_{A_i} = \int_{V_{A_i}} dr^3 \frac{1}{2} \sigma_{A_i} \text{Re}(\omega) |E_{A_i}(r)|^2$. P and F are two imperfection factors that each has a value between 0 and 1. Here E_{A_i} is the QNM excited inside the i th absorber through the mode overlapping from the center emitter. $\phi_\infty(\omega_0)$ is the reduced heat flow, and the overall radiative heat flux can be estimated as $\Phi_\infty = \int \frac{d\omega}{2\pi} \Theta \left[\frac{1}{A} L(\omega) \phi_\infty(\omega_0) \right]$, where the $L(\omega)$ is the Lorentzian lineshape function with the peak at the resonant frequency ω_0 and its linewidth is related to the quality factor of the mode $Q = |\text{Re}(\omega_0)/2\text{Im}(\omega_0)|$. The relative magnitude of D_E and D_{A_i} is determined by the coupling strength of the center emitter and the i th absorber. When thermally excited, the electric field of the plasmonic thermal emitter is strongly confined around its surface but exponentially decays to the far field. For two emitters to be coupled, the excitation of the QNM in one emitter needs to effectively invoke the same QNM in the other emitter. To quantitatively investigate the coupling strength between two adjacent emitters placed in the near field, we can integrate the QNM field overlapping as

$f_n(r) = \int dr'^3 E_0(r')^\dagger E_n(r' - r_n) / \int dr'^3 E_0(r')^\dagger E_0(r')$, where E_0 and E_n are the electrical fields of the QNMs of the center emitter and the n th emitter respectively, and r_n is the location of the n th emitter center. Thus, the relative intensity of the QNM field incurred inside the neighboring emitters can be evaluated. We further introduce the effective number of emitters

$$N_{eff} = \sum_{n=1}^N f_n(r) \quad (1.8)$$

to calculate the far-field thermal radiation from one emitter in the array.

Under the electric quasi-static approximation for a plasmonic emitter, the imperfection factors P and F approach unity. By utilizing the effective number of emitters coupled, we can get $\sum_i D_{A_i} = N_{eff} D_E$. Finally, the far-field thermal radiation from one emitter inside the array becomes

$$\phi_{\infty}(\omega) = \frac{4D_E D_{\infty}}{\left((N_{eff} + 1)D_E + D_{\infty}\right)^2} = \frac{4\beta}{\left((N_{eff} + 1)\beta + 1\right)^2}, \quad (1.9)$$

where $\beta = \frac{D_E}{D_{\infty}}$. Depending on the coupling strength of the plasmonic emitters, there exist three types of scaling behaviors for the far-field thermal radiation as follows. (i) When the plasmonic emitters are in the overcoupling regime, the QNMs of individual emitters are in the same phase and intensity. Hence N_{eff} approaches N as indicated by Equation (1.8), and the total thermal radiation from $(N+1)$ such plasmonic emitters inside the unit cell is $\phi_{\infty, total}(\omega) = \frac{4(N+1)\beta}{((N+1)\beta+1)^2} \propto \frac{1}{N+1}$. Thus, it indicates that the total thermal radiation from overcoupled plasmonic emitter array will be inversely proportional to the packing density. (ii) When the plasmonic emitters are in the non-coupling regime, the QNMs of individual emitters are in uncorrelated phase, hence N_{eff} becomes 0. The total thermal radiation from such an uncoupled plasmonic emitter array is $\phi_{\infty, total}(\omega) = \frac{4(N+1)\beta}{(\beta+1)^2} \propto (N+1)$. This implies that the total thermal radiation is now proportional to the packing density of the plasmonic emitter array. (iii) When the coupling between adjacent plasmonic emitters leads to the opposite phase for their QNMs, N_{eff} could become a negative value, and the total thermal radiation from such a plasmonic emitter array is able to deliver superior power as compared to the conventional uncoupled plasmonic emitter array: $\phi_{\infty, total}(\omega) = \frac{4(N+1)\beta}{((N_{eff}+1)\beta+1)^2} > \frac{4(N+1)\beta}{(\beta+1)^2} = (N+1)\phi_{\infty, uncoupled}$. It should be noted that the cases in (i) and (iii) arrive at the same scaling behaviors as reported in the previous work, which are referred to as the “superradiance” and “subradiance” effects, respectively. For the cases other than the aforementioned three scenarios, the thermal radiation from one emitter inside the array is always suppressed, compared with the thermal radiation from the stand-alone single emitter, $\frac{4\beta}{(\beta+1)^2}$. As the distance between adjacent emitters decreases, the thermal emission from one emitter inside the array ϕ_{∞} is continuously suppressed, while the packing density increases simultaneously. If the decreasing rate of ϕ_{∞} is larger than the increasing rate of the packing density, the total thermal radiation from the unit cell will be positively correlated with the packing density. Otherwise, the total thermal radiation will be negatively correlated with the packing density.

To verify this new scale law, we theoretically and experimentally investigated the far-field thermal radiation from metasurfaces consisting of gold nanorod arrays. Each nanorod emitter has a rectangular shape with 2.5 microns in length, and 70 nm by 80 nm in cross-section area. In the experiment, the nanorod emitter array was placed on a substrate which was designed to reflect all the infrared radiation to the upper space for the measurement purpose. The substrate was composed of a 150 nm thick alumina layer and a 50nm thick aluminum layer deposited on a SiO₂ thermal oxide wafer. The spatial distribution function of the electrical field intensity around a single nanorod emitter with and without the substrate was plotted in Fig. 1.2(a) for the first resonance frequency, and the corresponding QNM field profile was shown in the inset of Fig. 1.2(a). The exponential decay of electric field intensity leads to a fast-descending coupling strength and effective number of emitters coupled to the center one with respect to the distance between adjacent emitters, as shown in Fig. 1.2(b). The coupling strength can vary when emitters are

aligned in different directions. In this case, the coupling strength in the x-direction (shoulder-to-shoulder) is stronger than that in the y-direction (head-to-head) because of the dipole-like QNM profile. Also, the coupling strength is weakened when the emitter is placed on the high-index substrate as shown by the blue curve in Fig. 1.2(a). The weak coupling makes it difficult to observe the subradiance effect or reach the superradiance limit. For this QNM, the value of β is calculated to be 0.11 by the frequency-domain finite element method, and the reduced radiative heat flow ϕ_∞ at the resonant frequency from the stand-alone nanorod emitter is 0.36. By normalizing to the blackbody limit $\Phi_{BB} = \int \frac{d\omega}{2\pi} \Theta\left(\frac{2\pi}{\lambda^2}\right)$, the emissivity of the metasurface at the resonant frequency can be calculated.

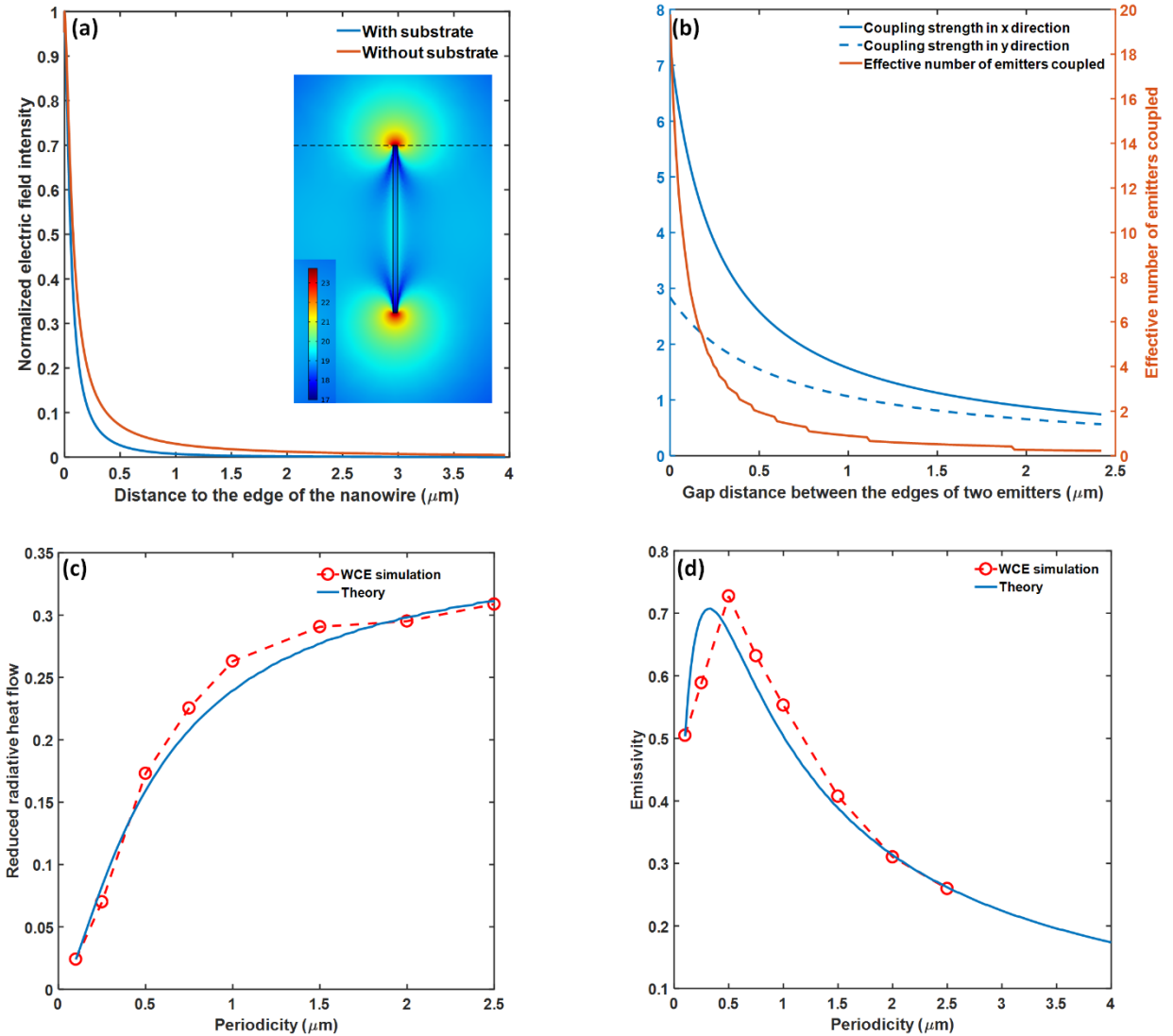


Figure 1.2 (a) Electric field intensity around a single nanowire emitter with and without the substrate that decays exponentially from the nanowire. Inset: corresponding QNM field profile (b) Coupling strengths (blue curves) between two emitters in the x- and y-directions, and corresponding effective number of emitters (red curve) coupled to the emitter due to the x-direction coupling. (c) Thermal emission from a single emitter when placed in the array at different periodicity in the x-direction. The periodicity in the y-direction is fixed to be 3.5 μm . The blue line

represents the prediction from our theory, and the dotted red curve is from the direct calculation using the WCE method. (d) Corresponding emissivity of the metasurface (gold nanorod array), where the total thermal emission from the metasurface is normalized to blackbody radiation.

Here we first investigated the suspended nanorod emitter array in vacuum where the periodicity in the y-direction is fixed to be $3.5\ \mu\text{m}$ but the periodicity in the x-direction varies. In Fig. 1.2(c), the predicted reduced radiative heat flow from one emitter inside the array was plotted in the blue curve. The corresponding emissivity was plotted in Fig. 1.2(d). The maximum emissivity of 0.72 is achieved when the periodicity in the y-direction is around 500 nm. To validate our theory, we directly calculated the far-field thermal radiation from the metasurface using the Wiener chaos expansion (WCE) method. The direct calculation and the prediction from the new scale law show good agreement, as shown in Figs. 1.2(c) and (d).

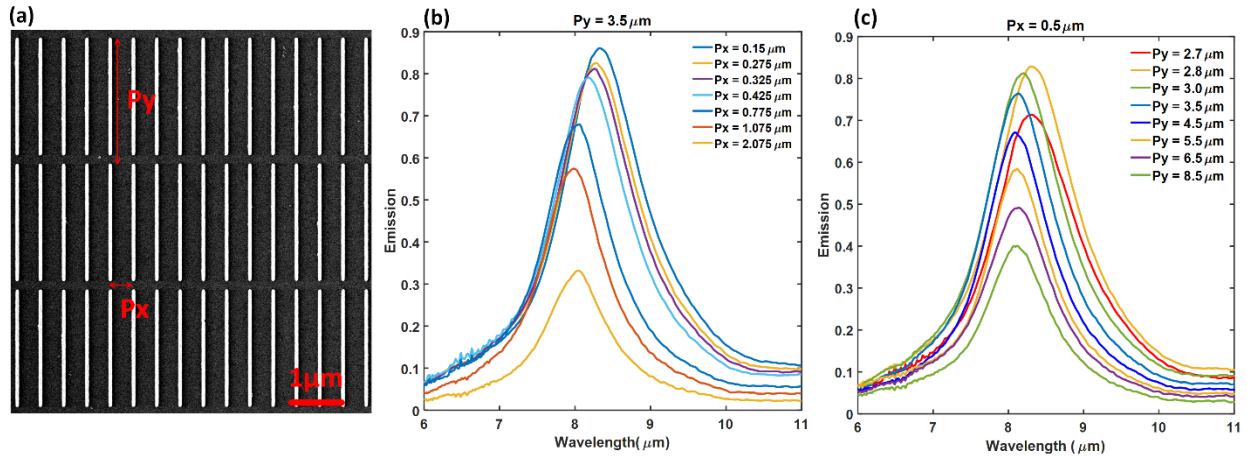


Figure 1.3 (a) SEM image of the nanorod emitter array when $P_y = 2.7\ \mu\text{m}$ and $P_x = 0.5\ \mu\text{m}$. (b) Emission spectra of a nanorod emitter array when P_y is fixed to be $3.5\ \mu\text{m}$ but P_x varies, and (c) when P_x is fixed to be $0.5\ \mu\text{m}$ but P_y varies.

We experimentally measured the emissivity of metasurfaces made from the nanorod emitter arrays, where the emissivity was indirectly evaluated as the absorptivity according to the Kirchhoff's law. This law is fully applicable in our experiment because the size of the arrays ($> 200\ \mu\text{m} \times 200\ \mu\text{m}$) is much larger than the characteristic wavelength of thermal radiation. Since our samples are opaque, their reflectivity is directly measured, and the emissivity can then be calculated by one minus the reflectivity. The y-polarized reflectivity spectra of the samples were measured by a Fourier-transform infrared (FTIR) spectrometer combined with an infrared microscope. An infrared linear polarizer was used to only allow the y-polarized reflected light to pass through. The reflection spectra of the samples were normalized to that of a gold standard mirror. The scanning electron microscopy (SEM) image of the fabricated nanorod emitter arrays was shown in Fig. 1.3(a). For $P_y = 3.5\ \mu\text{m}$, the measured emission spectra with varied P_x were plotted in Fig. 1.3(b). Indeed, the emissivity increases monotonically with the packing density as shown by both the experiment results and the theoretical prediction in Fig. 1.4(a), and the maximum emissivity point is pushed to be around $P_x = 100\ \text{nm}$. The overall emissivity is also larger than that without the substrate. The decrease of the mode coupling strength in the existence of a high-index substrate leads to larger thermal emission from the individual emitter at the same packing density, thereby resulting in a smaller optimized periodicity value and a larger maximum emissivity as compared to the case in vacuum (Fig. 1.2(d)). There is a red shift of the resonant frequency of the nanorod

emitter inside the array when the periodicity decreases in the x-direction as plotted in Fig. 1.4(c). The new theory developed in this work was utilized to calculate the frequency shifting, as shown in Fig. 1.4(d). Our experimental results agree well with our new theory.

To reach the “turning point” of the correlation between the metasurface emissivity and the nanorod packing density, we can further increase the coupling strength between the emitters by shrinking the periodicities. Here we used the other nanorod emitter array pattern where P_x was fixed to be $0.5 \mu\text{m}$ such that the coupling in the x-direction is still strong, but P_y varied. The corresponding emission spectra were shown in Fig. 1.3(c). In Fig. 1.4(b), an optimized periodicity was found to be around $P_y = 2.75 \mu\text{m}$ as predicted by the new scale law. The emissivity of the metasurface keeps increasing with the increased packing density until P_y reaches $2.75 \mu\text{m}$. After passing the optimized point, the overall emissivity decreases with increasing the packing density. An optimized point reaches at $P_y = 2.8 \mu\text{m}$ which gives the emissivity of 0.83. When P_y is further reduced, the emissivity quickly decreases. The discrepancies between theory and experiment in Fig. 1.4 are attributed to the following reasons. First, in QNM theory of thermal radiation, we only considered the contribution of dominant resonant modes. The non-resonant component of the radiation field was treated as background noise in the expansion of dyadic Green’s function, therefore its contribution to thermal radiation is omitted in the aforementioned theory. Second, the nanorod roughness and the small variations of the nanorod sizes ($\sim 20 \text{ nm}$ in the length direction and $\sim 5 \text{ nm}$ in the width direction) also contributed to the deviation of the theoretical prediction from experimental results.

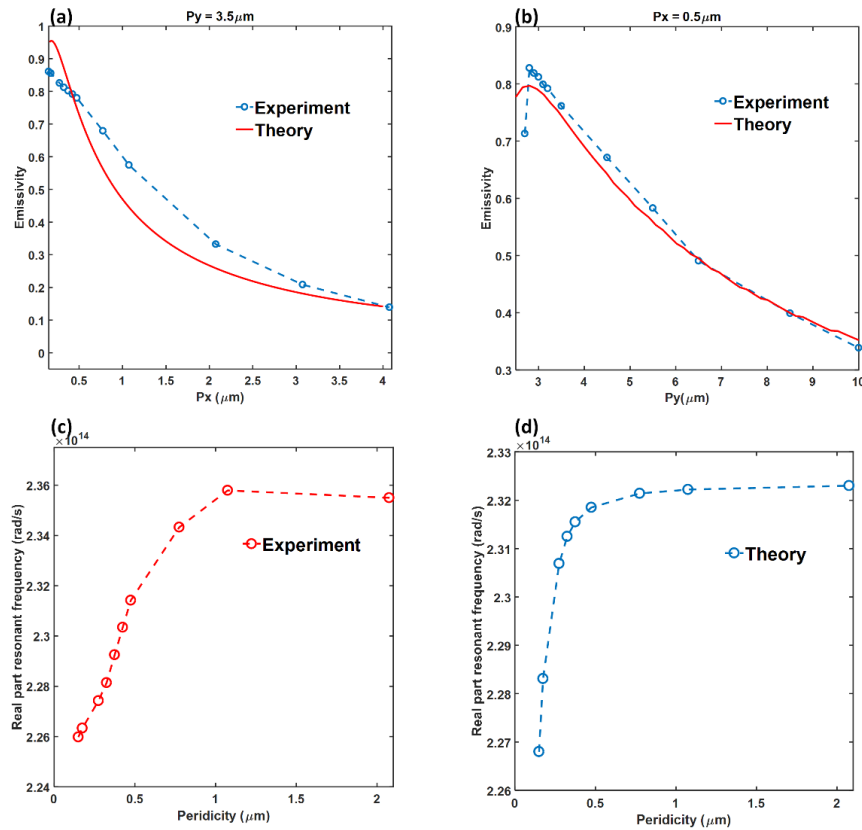


Figure 1.4 Emissivity of the nanorod emitter array for (a) $P_y = 3.5 \mu\text{m}$ and (b) $P_x = 0.5 \mu\text{m}$. The red shifts of the resonance frequency observed in experiment (c) and predicted by theory (d).

In summary, we developed a new scale law of far-field thermal emission by applying QNM theory to the metasurface consisting of densely packed plasmonic emitters. The TB method was used to approximate the collective resonant mode of the emitter array. The thermal radiation from a single emitter inside the array was observed to be suppressed by its adjacent emitters due to their in-phase near-field coupling, where such the suppression of thermal radiation can be quantitatively predicted by the new scale law. Depending on properties of the single emitter's QNM, the overall thermal radiation can be either positively or negatively correlated with the packing density. This new scale law of thermal radiation can thus serve as a general guideline for designing metasurfaces with desired thermal emission properties.

2. Design and fabrication of nanoplasmonic emitter based metasurfaces using graphene [Physical Review B, 106, 115416, 2022]

We designed and fabricated plasmonic metasurfaces based on graphene. Precise control of a material's emissivity is critical for thermal-engineering applications such as thermal imaging and reconfigurable thermal camouflage. Metamaterials, which derive their optical properties from subwavelength structures, have emerged as a promising way to actively modulate emissivity both spectrally and spatially. As a building block, single nanowire made of noble metals like gold can be used to construct metasurface or metamaterials with extraordinary emission properties, such as narrow-band and perfect emission spectrum at the desired wavelength range. Thermal radiation from 2D materials like graphene can be strongly mediated by the surface plasmon mode when appropriately excited. On the other hand, optical properties of graphene can be easily tuned by changing its electrical properties via gating or doping. Combining the nanowire-based metasurface with graphene layer, it is feasible to gain active control of free-space thermal radiation from such coupled systems.

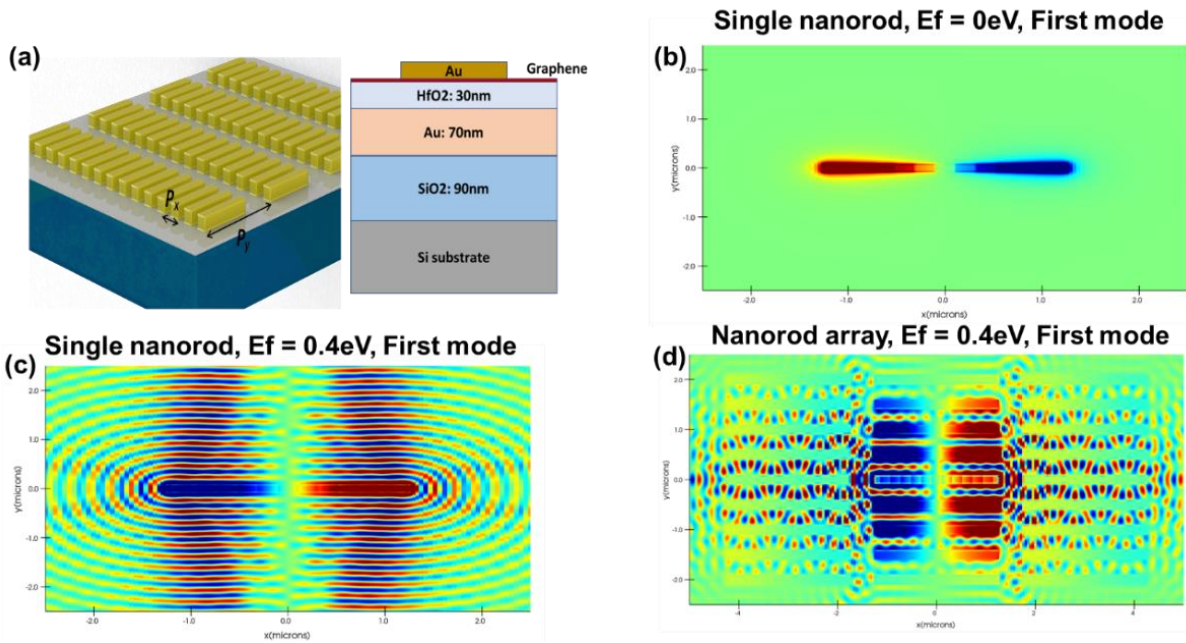


Figure 2.1 (a) Schematic of graphene-nanowire metasurface. (b) Fundamental resonant mode excited within the single nanowire on the intrinsic graphene layer. (c) Hybridized surface plasmon mode excited within the single nanowire on the gated graphene layer, Fermi level is 0.4eV. (d) Hybridized surface plasmon mode excited within the nanowire array on the gated graphene layer, fermi level is 0.4eV.

As shown in Fig. 2.1(a), the nanowires on top of the graphene layer will function as the near-field scattering tip to efficiently excite the surface plasmon modes inside the graphene. The gold nanowire will support multiple resonant plasmonic modes at different wavelengths. In Fig. 2.1(b), we used the WCE method to excite the first resonant mode of single nanowire placed on the intrinsic graphene layer. The corresponding nanowire resonant mode manifests a typical Fabry-Perot resonant mode characteristic with charges accumulated at two ends of the nanowire. However, for the intrinsic graphene layer without any gating, surface plasmon inside the infrared range is not supported. By gating the graphene layer, more free electrons can be injected in the graphene. Thus, graphene behaves like a metallic material where surface plasmon could be excited at the metal-dielectric interface. As we thermally heated up the graphene-nanowire array system, not only the nanowire resonant mode could be excited, but also the graphene surface plasmon come into existence. In Fig. 2.1(c), we plotted the electric field profile of the surface plasmon and nanowire resonant mode excited inside the coupled system. Such hybrid plasmon modes formed by the localized resonant mode inside the nanowire and the surface plasmon mode supported inside the graphene is the key to the modulation of the metasurface optical properties. The modulation speed can be much faster than those induced by thermo-optical effect. When there is only single nanowire deposited on the graphene layer functioning as the scattering tip, the surface plasmon mode supported by the graphene layer will continuously extend to the entire graphene layer until its energy completely decays. Once the nanowire array is introduced and a metasurface is formed above the graphene layer, the continuous surface plasmon supported by the graphene will then be transformed into the localized surface plasmon. In Fig. 2.1(d), we plotted the electric field profile of the corresponding localized surface plasmon mode inside the graphene and nanowire array system. The first resonant mode of the center gold nanowire is excited by placing a set of electric dipoles to mimic the heating process in the experiment. Consequently, the graphene surface plasmon mode is excited and trapped by the contact points between the nanowire and graphene layer. The emissivity of such metasurfaces could be controlled by either changing the packing density of the nanowire passively or gating the graphene layer actively.

We performed optical simulation (as shown in Fig. 2.2(a)) to prove the active control of nanowire-graphene metasurface emissivity in the mid-infrared range via electrical gating. Gold nanowires with the length of $2.5\mu\text{m}$ and cross-section area of 150nm by 80nm are patterned directly above a graphene layer of which the fermi level is tunable via electrical gating. A 30nm thick layer of HfO_2 is subsequently deposited with atomic layer deposition (ALD) below the graphene acting as the dielectric spacer for electrical gating. Another 70nm thick gold layer is deposited under the HfO_2 to serve as a mirror as well as the electrical gating electrode. The emissivity of the system is calculated by simulating its absorptivity to the incoming plane wave light. The electric field of the plane wave is parallel with the principal axis of the nanowire. As we set the pattern periodicity of gold nanowire array to $0.5\mu\text{m}$ in the x-direction and $3\mu\text{m}$ in the y-direction, the corresponding metasurface emissivity dramatically changes from 0.05 to 0.8 at wavelength of $10.5\mu\text{m}$ when graphene fermi level is tuned from 0.4 eV to 0.1 eV as shown in Fig. 2.2(b). The mechanism of controlling emissivity at a certain wavelength is the shifting and split of the nanowire resonant modes. As mentioned in the previous section, the hybrid plasmonic mode as shown in Fig. 2.2(d) is composed of two parts. One the intrinsic resonant mode of the gold nanowire itself. The other one is the localized surface plasmon modes existing inside the cavity created by the adjacent nanowires and graphene layer. When the graphene layer is not gated, no surface plasmon mode is supported, and the original resonant peak of the gold nanowire is preserved at $10.5\mu\text{m}$. When the graphene layer is electrically gated, it becomes metallic and starts to support surface plasmons.

The first resonant mode of the gold nanowire hybridizes with the graphene surface plasmons of different orders and gets further split into multiple peaks around the original peak wavelength. In Fig. 2.2(b), we plotted the emissivity spectrum of the graphene and gold nanowire array at two graphene Fermi level. When the fermi level is 0.1 eV which is relatively low, the original nanowire resonant mode peak is still preserved at 10.5 μm . As we increased the fermi level from 0.1 eV to 0.4 eV, the major resonant peak at 10.5 μm splits into three peaks at 8.7 μm , 9.7 μm and 11.7 μm . The corresponding electric field profiles (x-z plane) of these three peaks are plotted in Figs. 2.2(c)-(e). The peak #1 is the surface plasmon mode of the lowest order, and peak #3 is the highest order mode in the current wavelength range. For other smaller peaks that show up outside the 8-12 μm range, they might result from the hybridization of the graphene surface plasmon and higher order nanowire resonant modes. The number of the hybridized peaks around the original nanowire resonant modes and the spectrum distance between them could be controlled by the gap distance between the adjacent nanowires. In general, the smaller the gap distance between adjacent nanowires, the smaller the number of the hybridized mode is and the larger the spectrum distance between them is. Carefully designing the pattern periodicity and the gold nanowire parameters could shift the desired wavelength to arbitrary value in the mid-infrared spectrum. We could further divide the entire metasurface into multiple unit cells and obtain full control of their emissivity properties individually.

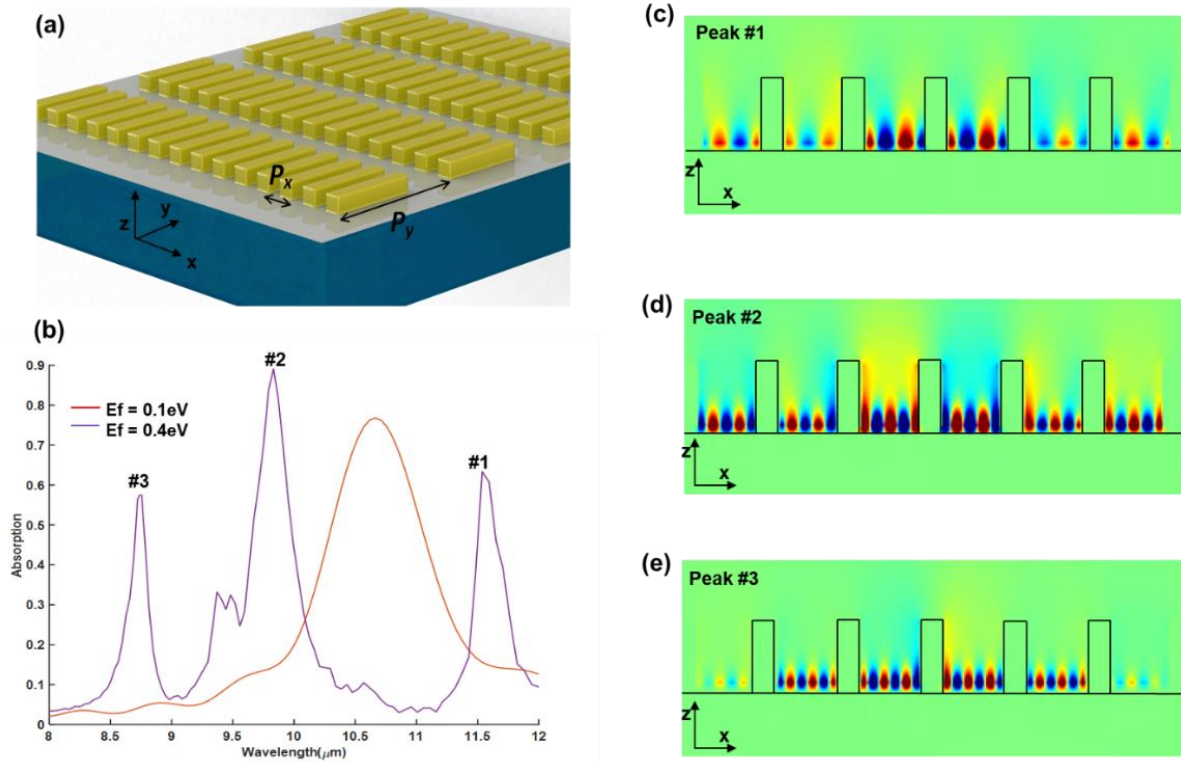


Figure 2.2 The modulation of the absorption(emission) spectrum for the graphene-nanowire metasurface. Nearly 90% of the change in the absorption spectrum around 10.5 microns wavelength is observed when the Fermi level of the graphene is changed from 0.1eV to 0.4eV.

As the building block of the metasurface, nanowire requires the polarization of the incident light to be in alignment with its principal axis such that the corresponding resonant mode could be

excited most effectively. By reciprocity, the emission from metasurface built upon nanowires is highly polarized. The emissivity of the metasurface is significantly reduced when the incident electric field polarization is switched to be perpendicular to the principal axis of nanowires (Fig. 2.3(a)). As shown in Fig. 2.3(b), the emissivity of the metasurface drops nearly to zero as the polarization of the incident light is changed compared to the result in Fig. 2.3(c). With the help of the coupling between the nanowire and graphene, the strict dependence on the polarization of the incident light could be relieved to some extent. In Figs. 2.3(c) and (d), we plotted the angular emissivity spectrum of the metasurface when the graphene Fermi level is tuned to be 0 eV and 0.6 eV. For the case of 0 eV, the emissivity is still very low. As the Fermi level is increased to 0.6 eV, there is a significant increase in the emissivity when the incident angle of the light is increased. Such improvement of the emissivity is attributed to the coupling between nanowire resonant mode and graphene surface plasmon. Even though the incident light is unable to effectively excite the resonant mode inside the nanowire, it is possible for the scattered light from the nanowire to excite the surface plasmons inside graphene layer. As a result, the hybridized plasmonic mode is invoked and contributes to the absorption of the incident light.

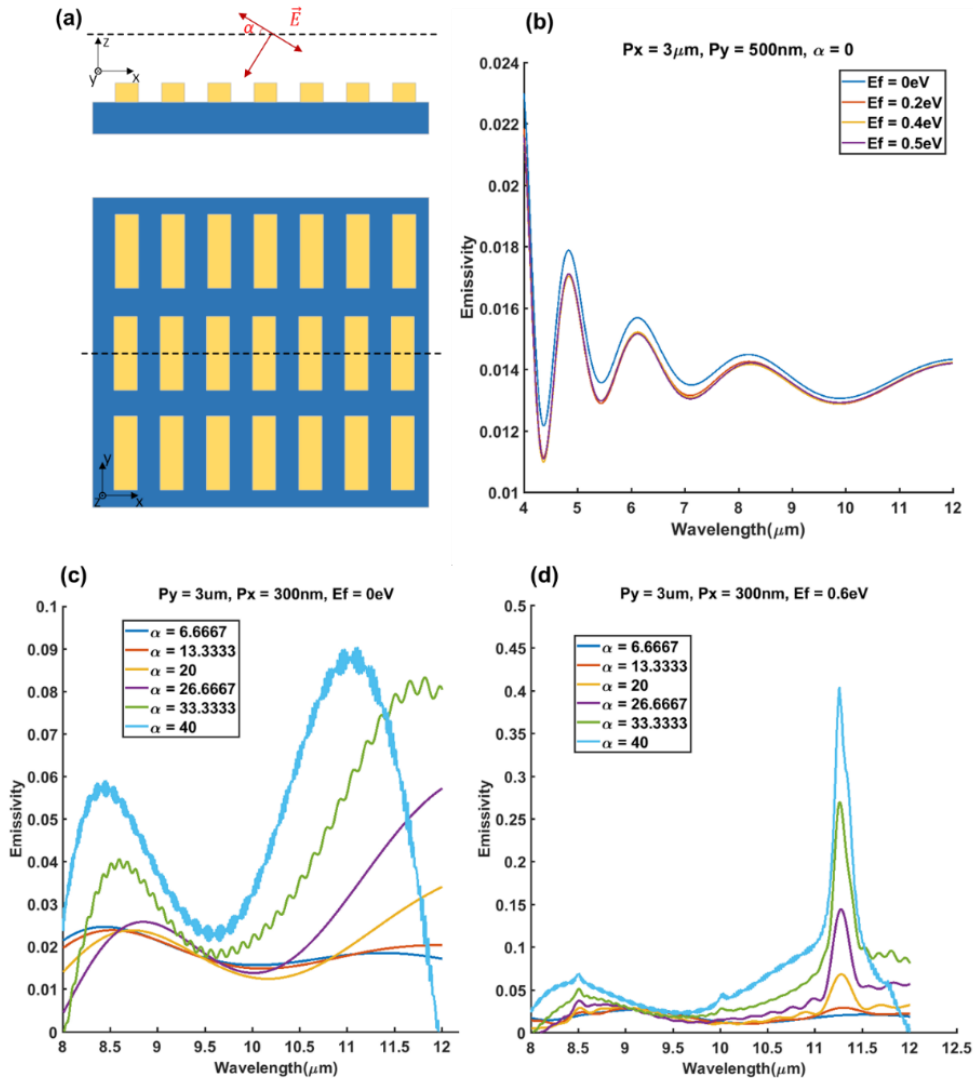


Figure 2.3 Angular dependence of the emissivity for the graphene nanowire array system when the incident electric field is perpendicular to the principal axis of the gold nanowire. (a) Schematic of the oblique incidence of the light onto the metasurface at angle α , and the periodicity of the nanowire is $3\mu\text{m}$ in the x-direction and 300nm in the y-direction. The angular dependent spectral emissivity of the metasurface when the graphene fermi level is tuned to be (b) 0 eV and (c) 0.6 eV.

3. Theory and design of inhomogeneous plasmonic thermal emitters [*Physical Review B*, 104 (19), 195426, 2021]

We explicitly formulated the principle of the WCE method in an inhomogeneous manner and implemented it in finite-different time-domain (FDTD) solvers via the discrete dipole approximation (DDA). The WCE method was validated by a case with the material inhomogeneity, but can also work in cases with temperature inhomogeneity, since the two types of cases share the same mathematical formulations.

According to fluctuational electrodynamics, the thermal radiation originates from the random current source induced by the Brownian motion of charges inside an emitter. Consider an emitter V with a current source $\vec{j}(\vec{r}, \omega)$ inside it, the emitted electrical field and magnetic field can be expressed as:

$$\vec{E}(\vec{r}, \omega) = i\mu_0\omega \int_V G^E(\vec{r}, \vec{r}_1, \omega) \vec{j}(\vec{r}_1, \omega) d^3\vec{r}_1 \quad (3.1)$$

and

$$\vec{H}(\vec{r}, \omega) = \int_V G^H(\vec{r}, \vec{r}_1) \vec{j}(\vec{r}_1, \omega) d\vec{r}_1 \quad (3.2)$$

respectively, where G^E and G^H are the Dyadic Green's functions of the system. The Poynting vector of the emitted field is:

$$\begin{aligned} \langle P_k(\vec{r}, \omega) \rangle &= \varepsilon_{ijk} E_i H_j = \varepsilon_{ijk} \cdot i\mu_0\omega \int_V d\vec{r}_1 \int_V d\vec{r}_2 \text{Tr}[G_i^{E*} G_j^H \langle \vec{j}(\vec{r}_1, \omega) \vec{j}^*(\vec{r}_2, \omega) \rangle] \\ &\equiv L_p[V; \langle \vec{j}(\vec{r}_1, \omega) \vec{j}^*(\vec{r}_2, \omega) \rangle] \end{aligned} \quad (3.3)$$

In equation (3.3), $i, j, k \in \{x, y, z\}$, P_k represents the k component of the Poynting vector $\vec{P}$ and ε_{ijk} is the Levi-Civita symbol. Here we define a linear operator $L_p[\Omega; X]$ associated with the Poynting vector: Ω specifies the domain where the integrations are carried over, and X is a 3 by 3 correlation matrix of the current source. The brackets $\langle \dots \rangle$ represent the ensemble average.

The current source $\vec{j}$ is induced by the Brownian motion of the free charges inside the emitter and thus can be quantified by the fluctuation dissipation theorem (FDT):

$$\langle \vec{j}(\vec{r}, \omega) \rangle = 0 \quad (3.4)$$

$$\langle \vec{j}(\vec{r}_1, \omega) \vec{j}^*(\vec{r}_2, \omega) \rangle = V_E^2(\vec{r}, \omega) \delta(\vec{r}_1 - \vec{r}_2) \mathbf{I} \quad (3.5)$$

In equation (3.5), $\mathbf{I}$ is the 3 by 3 identity and V_E^2 is a non-zero (if the emitter is lossy) deterministic term that relates to the imaginary part of the permittivity and the temperature of the emitter V :

$$V_E^2(\vec{r}, \omega) = \frac{4}{\pi} \omega \text{Im}[\varepsilon(\vec{r})] \Theta(\omega, T(\vec{r})) \quad (3.6)$$

where $\Theta(\omega, T(\vec{r}))$ is the Planck term defined as:

$$\Theta(\omega, T(\vec{r})) = \frac{\hbar \omega}{\exp\left(\frac{\hbar \omega}{k_B T(\vec{r})}\right) - 1} \quad (3.7)$$

with $\hbar$ and k_B representing the reduced Planck constant and the Boltzmann constant, respectively. In equation (3.6), we explicitly write V_E as a function of the position $\vec{r}$ so that the derivation can be directly applied to cases where the material inhomogeneity and the temperature gradient cannot be neglected.

Since V_E^2 is nonzero, it can be divided to the left-hand side of equation (3.5) and be moved into the bracket because it is deterministic. By defining a new vector $\vec{\xi} = \vec{j}(\vec{r}, \omega)/V_E$, equation (3.5) can be re-written as:

$$\langle \vec{\xi}(\vec{r}_1) \vec{\xi}^*(\vec{r}_2) \rangle = \delta(\vec{r}_1 - \vec{r}_2) \mathbf{I} \quad (3.8)$$

The delta function on the right-hand side of equation (3.8) implies $\vec{\xi}$ can be expressed by a linear superposition of the white noise function $dW(\vec{r})$:

$$\vec{\xi}(\vec{r}) = \sum_{m \in \{x, y, z\}} dW_m(\vec{r}) \vec{e}_m \quad (3.9)$$

where $\vec{e}_m$ is the directional vector in the m direction.

According to the Karhunen-Loève theorem, the white noise function dW can be expanded onto a set of pre-determined complete and orthogonal base functions defined within the emitter V :

$$dW_m(\vec{r}) = \sum_{\alpha=1}^{\infty} c_{m\alpha} f_{\alpha}(\vec{r}) \quad (3.10)$$

where f_{α} is the set of base functions that follow:

$$\int_V f_i(\vec{r}) f_j^*(\vec{r}) d\vec{r} = \delta_{ij} \quad (3.11)$$

and $c_{m\alpha}$ are the expansion coefficients that obey:

$$\langle c_{m\alpha} \rangle = 0 \quad \langle c_{m\alpha} c_{n\beta} \rangle = \delta_{nm} \delta_{\alpha\beta} \quad (3.12)$$

Substituting equation (3.10) into equation (3.9), the current source $\vec{j}$ can be expressed as:

$$\vec{j}(\vec{r}, \omega) = \sum_{m \in \{x,y,z\}} \sum_{\alpha=1}^{\infty} c_{m\alpha} V_E(\vec{r}, \omega) f_{\alpha}(\vec{r}) \vec{e}_m \quad (3.13)$$

Hence,

$$\begin{aligned} \langle \vec{j}(\vec{r}_1, \omega) \vec{j}^*(\vec{r}_2, \omega) \rangle_{mn} &= V_E^2 \langle dW_m(\vec{r}_1) dW_m(\vec{r}_2) \rangle \\ &= \sum_{\alpha, \beta} V_E^2 \langle c_{m\alpha} c_{n\beta} \rangle f_{\alpha}(\vec{r}_1) f_{\beta}^*(\vec{r}_2) \\ &= \sum_{\alpha} V_E^2 f_{\alpha}(\vec{r}_1) f_{\alpha}^*(\vec{r}_2) \delta_{mn} \end{aligned} \quad (3.14)$$

consequently,

$$\langle \vec{j}(\vec{r}_1, \omega) \vec{j}^*(\vec{r}_2, \omega) \rangle = \sum_{\alpha} V_E^2 f_{\alpha}(\vec{r}_1) f_{\alpha}^*(\vec{r}_2) \mathbf{I} \quad (3.15)$$

Plugging equation (3.15) into equation (3.3), the Poynting vector can now be expressed in a summation form:

$$\begin{aligned}
\langle P_k(\vec{r}, \omega) \rangle &= \varepsilon_{ijk} \cdot i\mu_0\omega \int_V d\vec{r}_1 \int_V d\vec{r}_2 \text{Tr} \left[G_i^{E*} G_j^H \sum_{\alpha} V_E^2 f_{\alpha}(r_1) f_{\alpha}^*(r_2) \mathbf{I} \right] \\
&= \sum_{\alpha} \sum_{m \in \{x,y,z\}} \varepsilon_{ijk} \cdot i\mu_0\omega \int_V d\vec{r}_1 \int_V d\vec{r}_2 \text{Tr} [G_i^{E*} G_j^H \overrightarrow{J_{\alpha,m}} \overrightarrow{J_{\alpha,m}^*}] \\
&= \sum_{\alpha} \sum_{m \in \{x,y,z\}} L_P[V; \overrightarrow{J_{\alpha,m}} \overrightarrow{J_{\alpha,m}^*}]
\end{aligned} \tag{3.16}$$

where $\overrightarrow{J_{\alpha,m}}$ can be viewed as a current mode defined by f_{α} as:

$$\overrightarrow{J_{\alpha,m}}(\vec{r}, \omega) = V_E(\vec{r}, \omega) \cdot f_{\alpha}(\vec{r}) \overrightarrow{e_m}, \quad m \in \{x, y, z\} \tag{3.17}$$

According to the definition of the operator $L_p[\Omega; X]$, each term of the summation in equation (3.16) represents the Poynting vector emitted by the emitter V when it is excited by the current mode $\overrightarrow{J_{\alpha,m}}$, defined in equation (3.17). Therefore, in order to evaluate the thermal emission of a considered emitter, one can excite the emitter by a series of current modes and calculate all the corresponding Poynting vectors (usually converged over the first several of modes). The summation of them will be the Poynting vector of the thermal radiation from the excited emitter. Because solving the Poynting vectors from a known current source can be achieved in most Maxwell equations' solvers, the WCE method can be conveniently implemented to achieve a direct simulation of thermal radiation.

In the WCE method, the Poynting vector of the thermal radiation from a given emitter is decomposed into Poynting vectors when the emitter is excited by a series of current mode. The current modes are defined by a set of complete and orthogonal base functions $f_i(\vec{r})$ which are determined by the morphology of the emitter according to equation (3.11). For instance, consider a cuboid emitter located in $x \in [0, L], y \in [0, W], z \in [0, H]$, the base functions can be chosen in the Fourier form:

$$f_{lmn}(x, y, z) = \sqrt{\frac{(1 + \delta_{l,0})(1 + \delta_{m,0})(1 + \delta_{n,0})}{V}} \cos\left(\frac{l\pi x}{L}\right) \cos\left(\frac{m\pi y}{W}\right) \cos\left(\frac{n\pi z}{H}\right) \tag{3.18}$$

where l, m and $n = 0, 1, 2 \dots$ label the order of the base functions and $V = L * W * H$. For emitters of other geometries, the base functions can be generated through performing the Gram-

Schmidt orthonormalization to any set of complete base functions (for example, the polynomial bases $\{1, x, x^2, x^3, x^4 \dots\}$).

In order to excite a given emitter by a certain current mode in the simulation, the DDA can be applied to approximate the volumetric current source: an array of dipoles is distributed within the emitter; the polarization and the amplitude of each dipole are determined by the polarization and the local magnitude of the current mode, respectively. As an example, in order to excite the cuboid emitter mentioned before with the current mode $\vec{J}_{\alpha,m}$, a dipole array containing X, Y and Z dipoles can be assigned to the emitter, resulting in the total amount of dipole $n = XYZ$. The center location of the dipole indexed as ijk ($i \in [0, X - 1], j \in [0, Y - 1]$ and $k \in [0, Z - 1]$) is given by:

$$\vec{r}_{c,ijk} = \left(\left(i + \frac{1}{2} \right) \frac{L}{X}, \left(j + \frac{1}{2} \right) \frac{W}{Y}, \left(k + \frac{1}{2} \right) \frac{H}{Z} \right) \quad (3.19)$$

Furthermore, because an electrical dipole located at $\vec{r}_0$ with amplitude $\vec{J}_0$ is equivalent to a concentrated current source $\vec{J}(\vec{r}) = \vec{J}_0 \delta(\vec{r} - \vec{r}_0)$, by setting the amplitude and polarization of the dipole ijk as:

$$\vec{J}_{ijk,\alpha,m} = V_d \cdot \vec{J}_{\alpha,m}(\vec{r}_{c,ijk}) \quad (3.20)$$

in which $V_d = LWH/n$ represents the volume associated with the dipole ijk , the dipole array will excite the desired current mode inside the emitter.

Solving the emitted Poynting vector from the emitter excited by a certain current mode can be done by most commercially available Maxwell equations' solvers. Here we showed a typical simulation setup in the Lumerical FDTD-solutions. In this case, far field emission from a slab emitter was simulated. The simulation domain is bounded within the surfaces whose edges are marked by the orange lines in Fig. 3.1(a). The boundary conditions of the simulation domain are set to be perfectly matched layers (PMLs) in order to eliminate all the reflected electromagnetic waves to prevent non-physical echoes caused by the limited size of the simulation domain. The slab emitter is excited by a dipole array (shown in Fig. 3.1(b)) whose distribution and amplitude are determined by equations (3.19) and (3.20), respectively. Poynting vectors of the emitted field is captured and integrated by the frequency domain power monitor enclosure (the areas surround by the yellow lines in Fig. 3.1(a)) to calculate the total spectrum. In addition, frequency domain filed monitors can be added to where needed in order to reveal the field profile around the emitter (Fig. 3.1(b)).

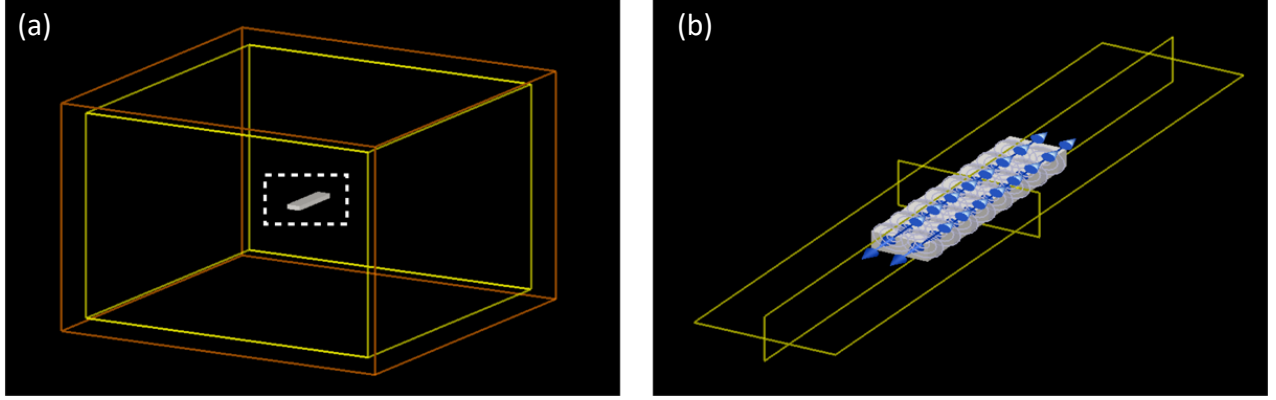


Figure 3.1 (a) Typical simulation setup of the WCE method in Lumerical FDTD-solutions. The simulation domain is bounded within the orange lines, and edges of the frequency domain power monitor enclosure is shown in the yellow lines. (b) Enlarged view of the region marked by the white dashed lines in (a). Blue arrows represent the dipoles to excite the emitter and yellow lines represent the borders of the frequency domain field monitors. The dipoles and the field monitors are not shown in (a).

We considered a simpler case with inhomogeneous material property, where the permittivity of the emitter studied is piece-wisely homogeneous. This problem can be viewed in two aspects in order to validate the WCE method in inhomogeneous cases: On one hand, the WCE method can be applied to the entire emitter in an inhomogeneous manner (i.e., V_E^2 in equation (3.6) is a function of position) so that the emission spectrum of the emitter $P_1(\omega)$ can be acquired; on the other hand, the WCE method can be individually applied to each homogenous part of the emitter, and superpose the contributions from each part results in the total emission spectrum $P_2(\omega)$. If $P_1(\omega)$ matches $P_2(\omega)$, as the additive principle of thermal radiations suggested (Fig. 3.2), the WCE method will then be validated for inhomogeneous cases.

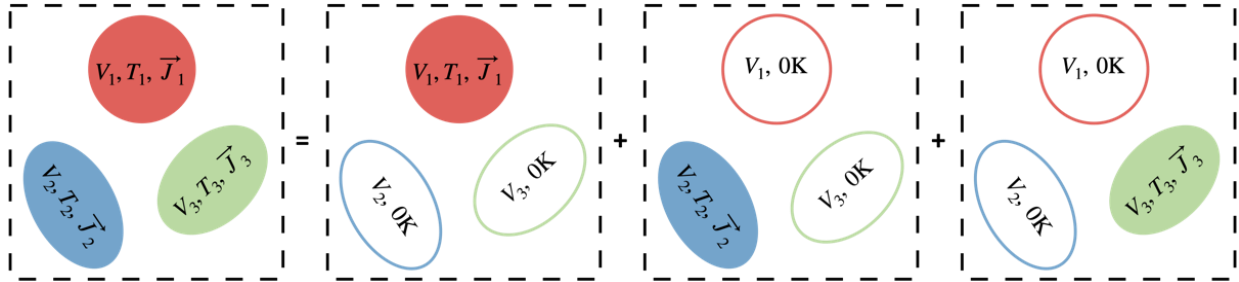


Figure 3.2 Schematic of the additive principle of thermal radiation. The thermal radiation from multi-emitter system is equivalent to the superposition of the thermal radiation when each emitter is excited individually to the same temperature as it is in the system. Excited emitters are marked by solid blocks.

Specifically, we simulated a slab emitter centered at the origin, as shown in Fig. 3.3(b). The relative permittivity of the emitter is defined by the Lorentz model together with a spatial dependent factor $\Xi(z)$:

$$\epsilon_r(z, \omega) = \Xi(z) \cdot L(\omega) \quad (3.21)$$

where $L(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\gamma\omega}$, with $\omega_p = 7 \left(\frac{2\pi c}{a} \right)$ and $\gamma = 0.03 \left(\frac{2\pi c}{a} \right)$; c represents the speed of light in vacuum and $a = 1\mu\text{m}$. $\Xi(z) = 2 - \delta(z - |z|)$ is the spatial dependent factor in the form of the step function, resulting in the piece-wisely homogeneous permittivity in the $z > 0$ and the $z < 0$ regions, respectively.

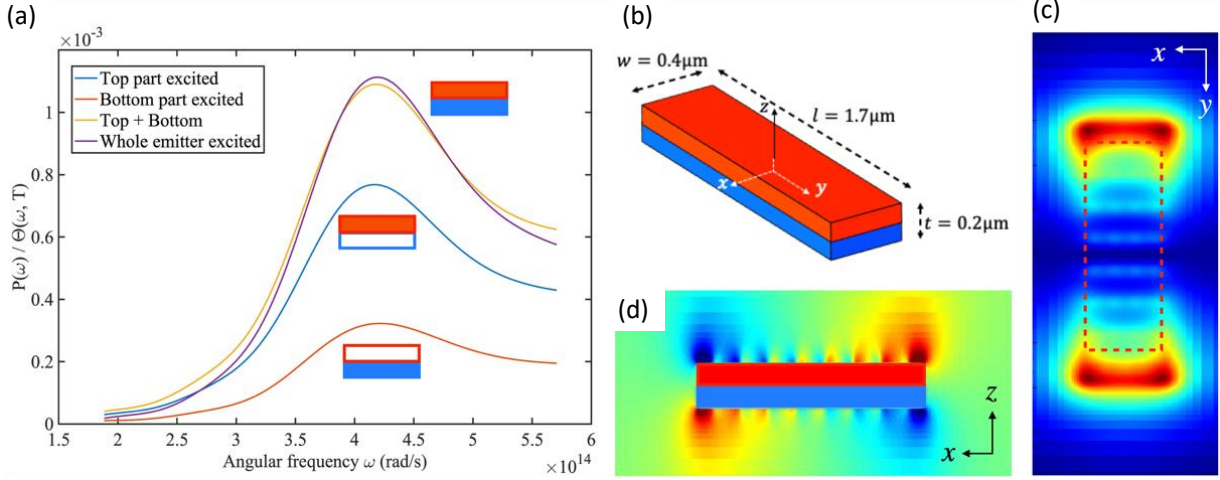


Figure 3.3 (a) Simulated radiative spectra of the slab emitter with permittivity inhomogeneity when the entire emitter is excited (pulper line) and only a homogeneous part is excited (blue and orange lines). Excited parts are marked by solid rectangles. (b) Perspective view of the slab emitter. (c) $|E|^2$ profile in the $x - y$ plane. Red dashed lines represent the border of the emitter. (d) E_z profile in the $x - z$ plane. The red and blue rectangles represent the cross section of the emitter.

The simulated radiative spectra were shown in Fig. 3.3(a). A resonance peak at $\omega_0 = 4.3 \times 10^{14} \text{ rad/s}$ was observed. The blue line and the orange line showed the contributions when only one homogeneous part was excited in the WCE method. The summation of the two, shown in the yellow line, matches the direct simulated spectra (pulper line) when the WCE method is applied to the entire emitter in an inhomogeneous manner, which validates the WCE method in inhomogeneous cases.

Figures 3.3(c) and (d) showed the field profiles at the resonance frequency in the $x - y$ plane and $x - z$ plane, respectively. These two field profiles indicate that the thermal radiation from the emitter at the resonance frequency follows the mode of the dipole radiation since the emitted field is mainly concentrated at the two ends of the emitter and shows an anti-symmetric pattern across the cross section of the emitter.

In summary, the WCE method provides a direct approach to simulate thermal radiation spectra of plasmonic thermal emitters in sub-wavelength sizes by expanding the Poynting vector of the thermal radiation onto Poynting vectors from known current modes, which can be simulated in most Maxwell equations' solvers. Despite its advantages, the WCE method has never been adapted to inhomogeneous cases. In our formulation, we explicitly formulated the method in an inhomogeneous manner. The new formulation allows us to apply this method to any inhomogeneous system where the V_E^2 term in equation (3.6) can be expressed as a function of positions, which enables the simulation of more complicated emitter systems. As proof of concept,

we validated the new formulation of the WCE method through a case with a piece-wisely homogeneous permittivity.

4. Design and analysis of electrically modulated thermal metasurfaces [*Frontiers in Energy*, 17, 134–140, 2022]

We studied the size effect of electrically modulated thermal metasurfaces consisting of an array of nanoplasmonic emitters, by designing gold nanorod array (GNA) based metasurfaces with a nearly perfect narrowband emission, as shown in Fig. 4.1(a). The resonance intensity from the finite GNA can be strongly converged to the value comparable to the infinite case. The resulting small footprint allows the metasurface thermal emitters to respond at a nanosecond timescale. More importantly, this strong resonance is to some extent insensitive to the aspect ratio of the nanorod array, which serves as a new degree of freedom to achieve the temperature uniformity of less than 1 K. The comprehensive analysis presented in this work thus provides a guideline for designing high-performance thermal infrared metasurfaces.

The optical response of the metasurface is modelled in Ansys Lumerical finite-difference time-domain solutions. Then, electrothermal simulations are conducted in COMSOL Multiphysics. The GNA is positioned on the multilayer consisting of a Si substrate, a 300 nm thick layer of SiO₂ insulation layer, a 200 nm thick Al reflector, and an 100 nm thick dielectric spacer made of Al₂O₃, as shown in Fig. 4.1(b). The Al reflector also functions as a heater, where a current is passed through to heat up the GNA.

In the optical simulations, the GNA and beneath substrate are illuminated by a plane wave source polarized in the x-direction, as indicated by the blue arrow in Fig. 4.1(b). A reflective power monitor is placed above the source to capture the reflective energy and thus the resonance feature of the GNA. The thermal simulation combines an electric current module, governed by the electric current equations, with a 3D heat conduction module, governed by heat diffusion equations. In the electric current module, a fixed current of $I_0 = 250$ mA passes through the Al reflector, and the calculated volumetric Joule heat generation is set to be the heat source in the heat conduction module. The bottom surface of the silicon substrate is fixed to have a constant temperature of $T_0 = 293.15$ K. The transient simulation starts with the overall initial temperature of T_0 , and the temperature profiles are captured within the time from 0 to 2000 ns.

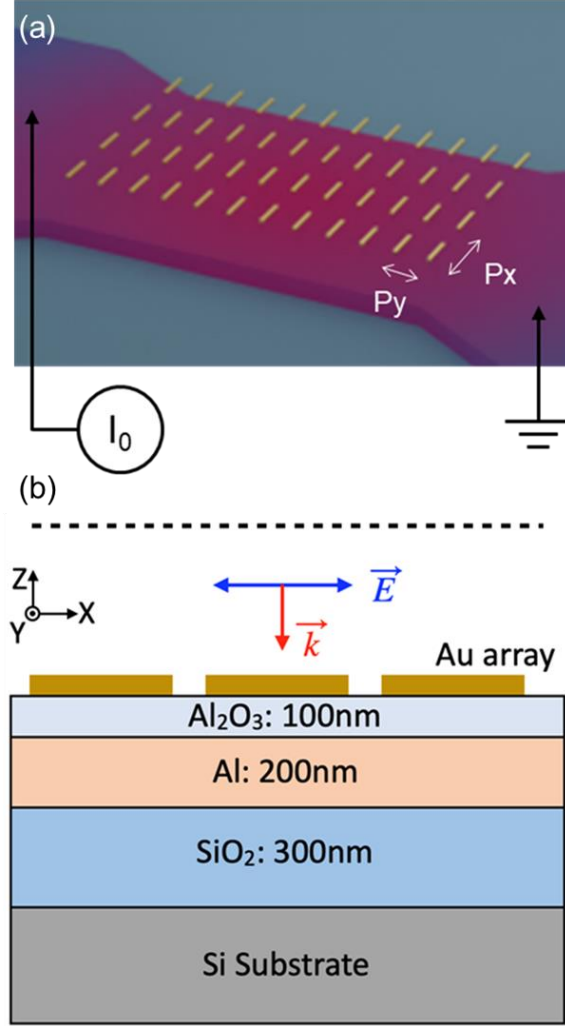


Figure 4.1 (a) Schematic of a metasurface based on gold nanorod arrays. The Al_2O_3 spacer is hidden to reveal the beneath Al reflector, which also functions as the heater (Joule heating is applied via a fixed current I_0). P_x and P_y represent the periodicities in the x-direction and the y-direction, respectively. (b) x-z view of the simulated device. The dimensions of each gold nanorod are 700 nm by 50 nm by 50 nm in the x, y, and z directions. The size and the location of the reflective power monitor (dashed line) is fixed among simulations.

To analyze the finite size effect on the metasurface, we first simulate the reflective spectrum of an infinite GNA as a reference. Typically for an infinite GNA, the resonance frequency in the reflective spectra is determined by the localized surface plasmon resonance (LSPR) of the single gold nanorod resonator, and the resonance strength is governed by both the LSPR and the packing density (periodicity). Here, we design the GNA with the single nanorod sized as 700 nm by 50 nm by 50 nm in the x, y, and z directions and the periodicities set to be $P_x = P_y = 800$ nm. The simulated spectrum is plotted as the black line in Fig. 4.2(a) and features a first-order resonant peak centered around 1046 nm in the wavelength. Figure 4.2(b) illustrates the $|\vec{E}|^2$ profile at the resonance frequency, which indicates a longitude LSPR mode. High-order mode at around 870 nm in Fig. 4.2(a) corresponds to the weak periodic brightness near the center of the nanorod in Fig. 4.2(b).

The effect of the finite array size on the GNA metasurface is then investigated with a different finite number of $N \times N$ arrays from $N = 1$ to $N = 9$, as shown in Fig. 4.2(a). Although for simulations involving finite size arrays, it is inherently impossible to eliminate the absorption effect of the simulation boundaries (perfectly matched layer), which will inevitably result in an underestimation of the reflectance. We thus, in all finite array size simulations, fix the monitor size (fully covered the 9 by 9 arrays) and the distance between the monitor and the metasurface. By doing so, we can mimic the real experimental condition where those arrays are measured by the same spectrometer. Hence those reflectance spectra are comparable, and the spectral emissivity of those finite size metasurfaces can be inferred based on Kirchhoff's law.

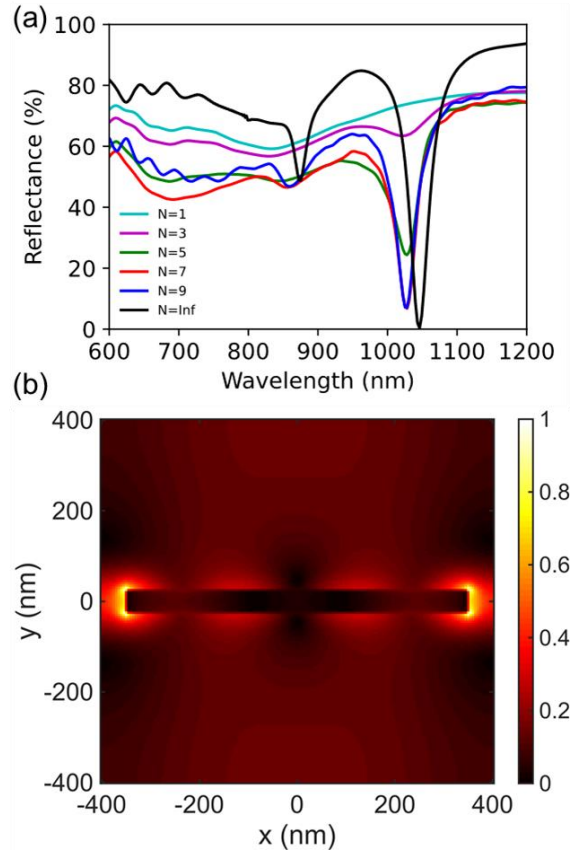


Figure 4.2 (a) Simulated reflectance spectra for different array sizes. (b) Simulated $|E|^2$ profile of a single unit cell in the infinite array at the resonance wavelength of 1046 nm.

From Fig. 4.2(a), we can find that the finite array preserves the narrowband feature of the infinite one and the first-order resonance starts to saturate when $N = 7$, which corresponds to a metasurface with an area of only about $2.2 \mu\text{m}^2$. This enables the device to have a thermal response down to nanoseconds as discussed later in electrothermal simulations. Compared to the case of infinite array size, a red shift of resonance can be clearly observed. It depends on the packing density or periodicity of the nanorod array, as described by the tight-binding model. With an increasing number of nanorods, the dipole moment can be effectively elongated, causing a longer resonance wavelength.

The influence of the GNA arrangement to the reflectance spectra is also studied. To achieve this, we keep the total number of unit cells around 49, which corresponds to the 7 by 7 array marking the resonance saturation, but vary the aspect ratios N_x/N_y of the array (i.e., number of unit cells along the axial direction (x-direction) divided by the number along the other (y-direction)). For arrays with different aspect ratios, the contrast ratio C_r of the reflectance spectrum, defined as:

$$C_r = [1 - \min(R)] / [1 - \text{mean}(R)] \quad (4.1)$$

is evaluated. In Eq. (4.1), $\min(R)$ represents the minimum value in the reflectance spectrum and $\text{mean}(R)$ represents the spectral average of the reflectance spectrum. Hence, the contrast ratio physically represents to which degree the resonance feature stands out of the baseline of the spectrum. Therefore, a large C_r value is targeted.

From Fig. 4.3(a), for $N_x/N_y > 1$, the resonance can maintain its intensity even with only a small number of arrays in the perpendicular direction (y-axis) due to the highly polarized nature of the GNA resonance. On the other hand, the arrays with an aspect ratio $N_x/N_y < 1$ show a significantly decreased contrast ratio. A detailed comparison can be found in Fig. 4.3(b), where $N_x/N_y = 16/3$ almost keeps the same resonance intensity as that of the unbiased case $N_x/N_y = 7/7$ while the peak height of $N_x/N_y = 3/16$ is reduced by almost a half and broadened. The relaxation of the requirement for the number of arrays in the direction perpendicular to polarization can be found later to significantly improve the temperature uniformity of the GNA when considering the electrothermal active emission. It is thus a valuable factor in design, especially at the nanoscale where the controlling parameters are limited.

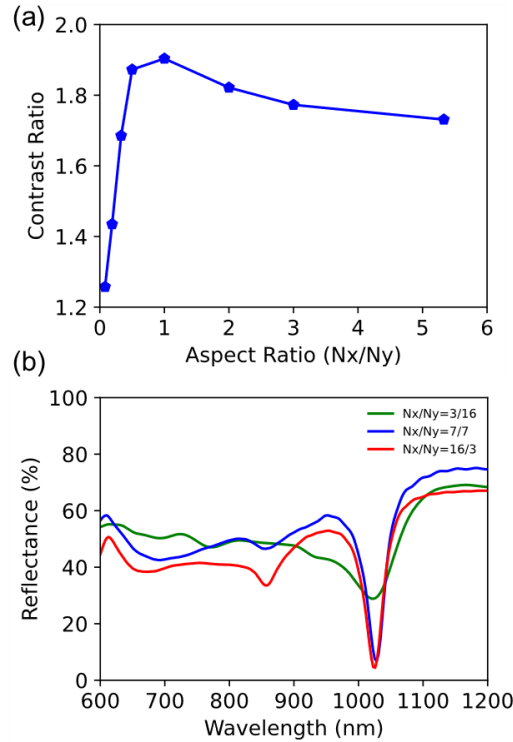


Figure 4.3 (a) Contrast ratios C_r of finite GNAs with different aspect ratios. (b) Simulated reflectance spectra for aspect ratios of $N_x/N_y = 16/3$, $7/7$ and $3/16$, respectively.

The GNA is then electrically heated, and its emission is analyzed in both transient and steady states. We first simulate the response time (defined as 90% of the rise time responding to the on-switch current injection) for the device under different array sizes and aspect ratios. With the increased array size, the response time increases dramatically, as shown in Fig. 4.4, since a larger size induces a larger thermal mass which significantly reduces the cutoff modulation frequency. This shows the importance of finding the minimal size of the finite metasurface showing enough resonance contrast. For our device, with a saturation size of $N = 7$, we can achieve a response time of about 600 ns, which outperforms the majority of the current tunable metasurface designs.

Under the similar array size around $N = 7$, containing 49 unit cells, we further study the influence of the aspect ratio. Even under an extremely large aspect ratio change ($N_x/N_y = 4/12 \sim 12/4$), we can keep the device response time down to nanoseconds, with a fluctuation of only around 100 ns (red-dashed box). Combined with our previous discussion that a large N_x/N_y has less influence on the resonance contrast of the x-polarized metasurface, the aspect ratio can thus hold the metasurface a stable performance both optically with a narrowband, nearly perfect emission and electrothermally with an ultrafast response.

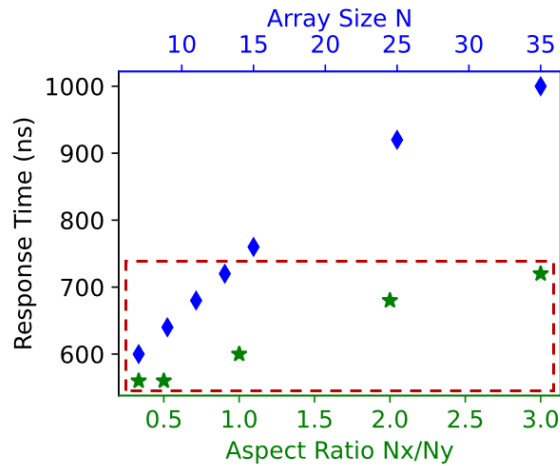


Figure 4.4 Response time for arrays in different sizes N (blue diamonds) and aspect ratios N_x/N_y (green stars).

Therefore, under a certain window, the aspect ratio can be set freely to benefit other aspects of our device performance, among which the temperature uniformity of the metasurface is fundamentally important. A reliable thermal emission typically favors a stable and uniform temperature, which also simplifies the design processes. Even when pursuing some advanced nonequilibrium emission, controlling temperature uniformly at a small local area is a prerequisite for most cases.

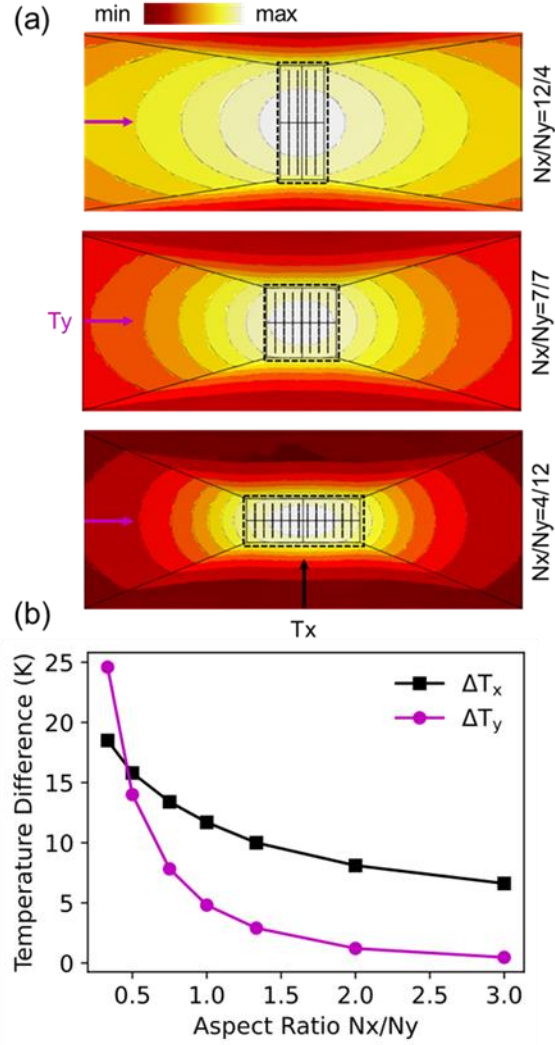


Figure 4.5 (a) From top to bottom: normalized isothermal contours for arrays in aspect ratio $N_x/N_y = 12/4$, $7/7$, and $4/12$, respectively. (b) Temperature difference along x-axis ΔT_x and along y-axis ΔT_y for arrays in different aspect ratios.

The temperature uniformity of the GNA metasurface is then illustrated by the 2D normalized isothermal contours, as shown in Fig. 4.5(a), together with the geometry of the arrays and electrodes. The GNA with a larger N_x/N_y is found to have much sparser isothermal contours, indicating a smaller temperature gradient. To specify the details, we also define the temperature difference ($T_{\max} - T_{\min}$) inside the array area enclosed by black-dashed boxes in Fig. 4.5(a). The temperature difference for both axes, as shown in Fig. 4.5(b), decreases with increasing N_x/N_y because of the thermal conduction from the electrode, which substantially modifies the temperature profile of the metasurface. Another indication of the impact from the electrode is that the decrease of temperature difference along the y-axis is faster than that of the x-axis due to the more effective heat conduction through the electrode metals. With N_x/N_y increasing up to about 3, the temperature difference can even reach sub-Kelvin along the y-axis while keeping a high resonance contrast ratio of 1.8 and a fast response time of 720 ns. It should be noted that here we only consider a common trapezoidal electrode, whose material and shape can be further optimized together with the GNA aspect ratio to enhance the temperature uniformity. Finally, the degree of

freedom in aspect ratio tuning can also improve the power injection into the metasurface by the impedance matching based on the maximum power transfer theorem.

In summary, we design a thermal metasurface based on a finite-size gold nanorod array, which, with only several unit cells, can achieve a nearly perfect narrowband emission close to that of the infinite case. Because of its small volume, the metasurface can achieve a fast response down to a nanosecond level. Moreover, the aspect ratio of this finite array can be tuned to greatly enhance the temperature uniformity to a sub-Kelvin temperature difference while maintaining the high resonance contrast ratio and the nanosecond response time. The analysis methodology can be extended to polarization-independent metasurfaces, such as the crossbar array that shows better emission efficiency in sensing applications. Furthermore, our analysis can also be extended to handle inhomogeneous temperature cases under the framework of a local Kirchhoff's law. With these extensions, the analyses of the size and aspect ratio of the finite-size metasurfaces thus pave the way to optimize the electrothermal infrared devices.

5. Electrically driven thermal infrared metasurface: Fast Response, uniform temperature, and narrowband direct emission [Applied Physics Letters, 121, 131703, 2022]

We experimentally demonstrated an electrically driven thermal infrared metasurface based on a gold nanorod array, which also verifies the modelling results above. The overall device, as shown in Fig. 5.1(a), is a metal-insulator-metal metasurface with the center-contacted electrode line design allowing a fast Joule heating to electrically excite the emission without deteriorating the optical resonance. By tuning the gold nanorod dimensions, we can obtain the emission at different resonant wavelengths. In Figs. 5.1(b) and (c), the scanning electron microscope (SEM) images show a nice fabrication of two nanorod arrays with resonances at $5.24\ \mu\text{m}$ and $3.61\ \mu\text{m}$, respectively.

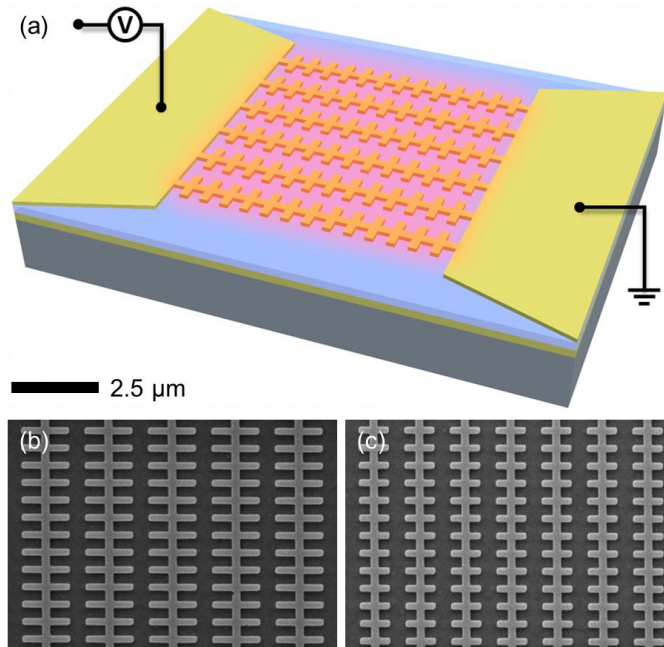


Figure 5.1 (a) Overview of the electrically driven thermal infrared metasurface. (b) SEM image of gold nanorod array with resonance at $5.24\ \mu\text{m}$ and its centered-contact electrode line. (c) SEM image of metasurface with the same design but resonating at $3.61\ \mu\text{m}$.

The center-contacted electrode line design is verified by the finite-difference time-domain simulations. As shown in Fig. 5.2(a), there is little difference in the reflectance between the nanorod array with and without the electrode lines for both $5.24\ \mu\text{m}$ and $3.61\ \mu\text{m}$ resonances. The mechanism can be clearly shown in the mode profiles of $5.24\ \mu\text{m}$ resonance in Fig. 5.2(b). The electrode line only contacts the area of nanorod with modest field intensity making it almost identical to the field distribution of the one without electrode line. The electrode line can be changed freely to adjust the impedance for a better power injection, although the resonance can be undermined if the size is beyond the weak-field area. This approach also keeps the metasurface simple with the nanorod plasmonic resonators that we can tune the narrowband emission straightforwardly compared to some void meta-design.

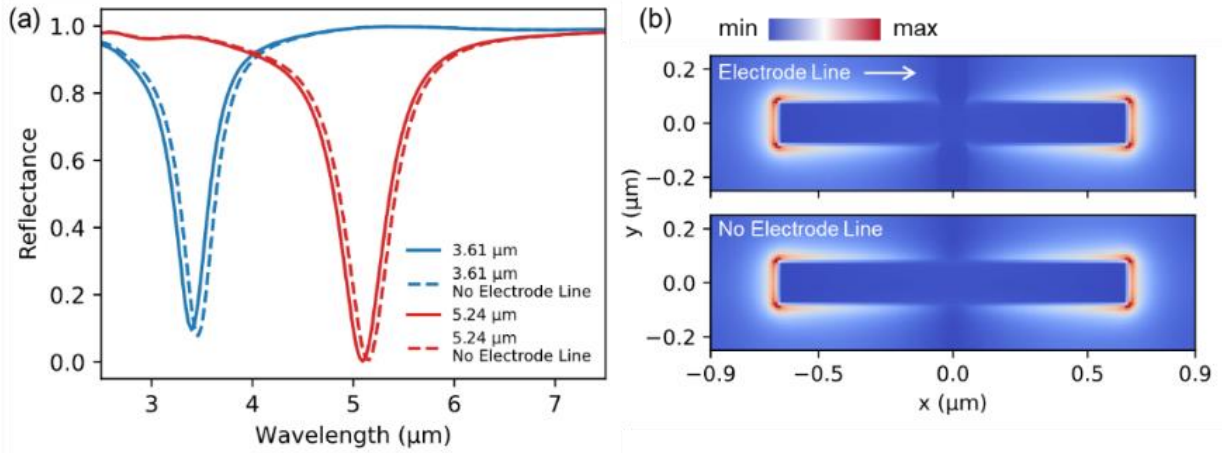


Figure 5.2 (a) The reflectance of metasurfaces with and without electrode line for both $5.24\ \mu\text{m}$ and $3.61\ \mu\text{m}$ resonances. (b) The electric field profile of the nanorod at $5.24\ \mu\text{m}$ resonance with and without electrode line.

We then characterize the performance of our metasurface in spatial, temporal, and spectral domains. In Fig. 5.3(a), the temperatures of the metasurfaces are measured by the thermal mapping system. The inset shows an emission image of the array with a quite uniform hot area, whose average temperature and temperature standard deviation are also shown. With increasing voltages, the average temperatures increase parabolically due to the Joule heat generation of V^2/R where R is the electrical resistance. The temperature standard deviation, representing the temperature non-uniformity, also increases with the temperatures. The higher temperatures tend to make the parabolic shape of temperature from Joule heating more prominent. However, with the metasurfaces of size $50\ \mu\text{m} \times 50\ \mu\text{m}$ in this temperature range, the temperature profiles keep good uniformity. As shown in Fig. 5.3(b), the contours of the metasurfaces show that the temperature maintains flat along the length of $50\ \mu\text{m}$ for both X and Y axes (defined in the inset of Fig. 5.3(a)). This uniformity is good enough for us to approximate Kirchhoff's law, which is also verified by our direct emission measurement later.

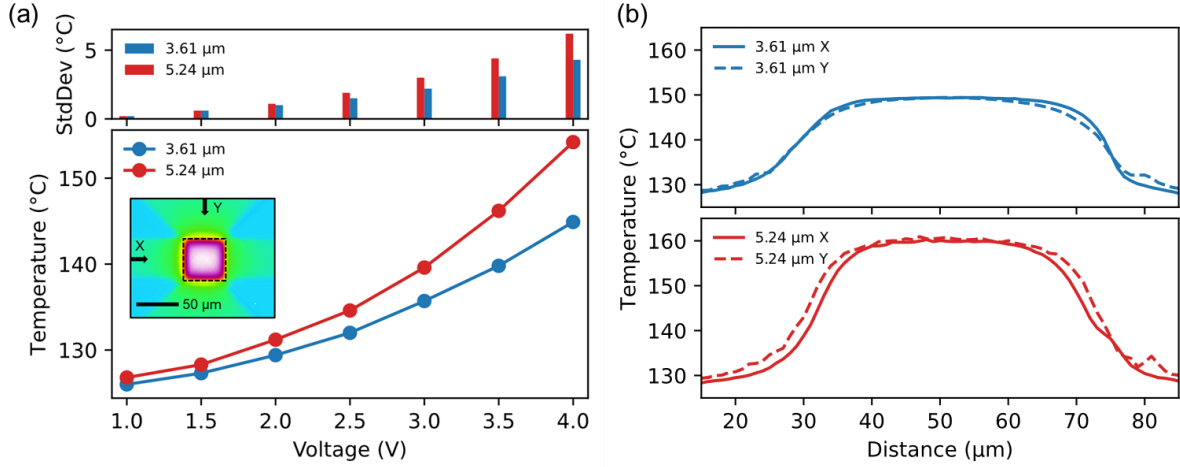


Figure 5.3 (a) The thermal mapping of temperatures of metasurfaces at different voltages. (b) The temperature contours of metasurfaces along X and Y axes.

The metasurfaces also show a fast response with rise time of 16.13 μs and 17.55 μs for 5.24 μm and 3.61 μm resonances, respectively, as shown in Fig. 5.4. The response speed is mainly dependent on the heat conduction to the substrate thus the array size and substrate materials play a major role, which is analyzed from our modeling work above.

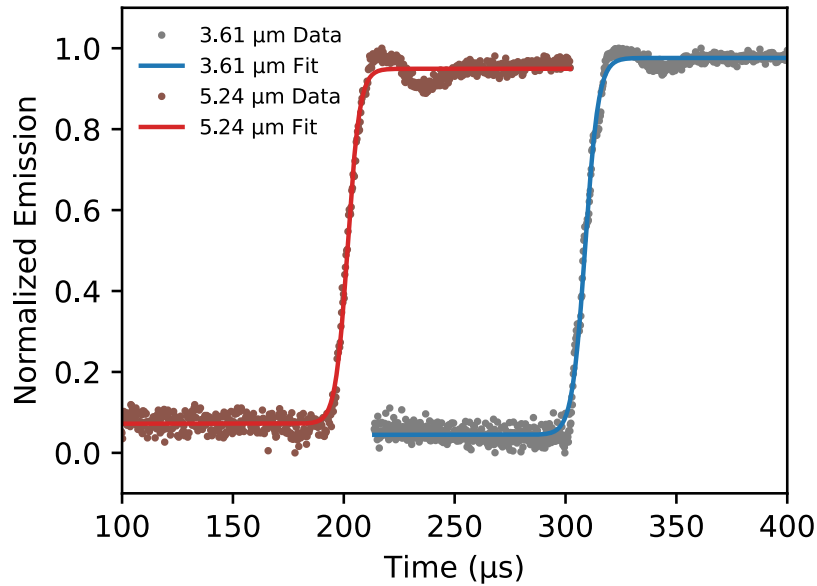


Figure 5.4 The rise time of metasurfaces.

Finally, the narrowband direct emission is measured by our FTIR system. The direct emission is the product of the blackbody emission and the emissivity, which equals absorptance in the approximation of Kirchhoff's law. As shown in Fig. 5.5(a), even at a low temperature, the metasurface with the 5.24 μm resonance has a higher emission peak than the one with 3.61 μm due to the better overlapping of the blackbody emission and emissivity. To verify our direct

emission, a careful calibration can be done based on different global heating. The details can be found in our report about the FTIR in the next section. Then we can recover the emissivities well consistent with the ones measured from reflectance, for both $5.24\ \mu\text{m}$ and $3.61\ \mu\text{m}$ resonances, as shown in Figs. 5.5(b) and (c).

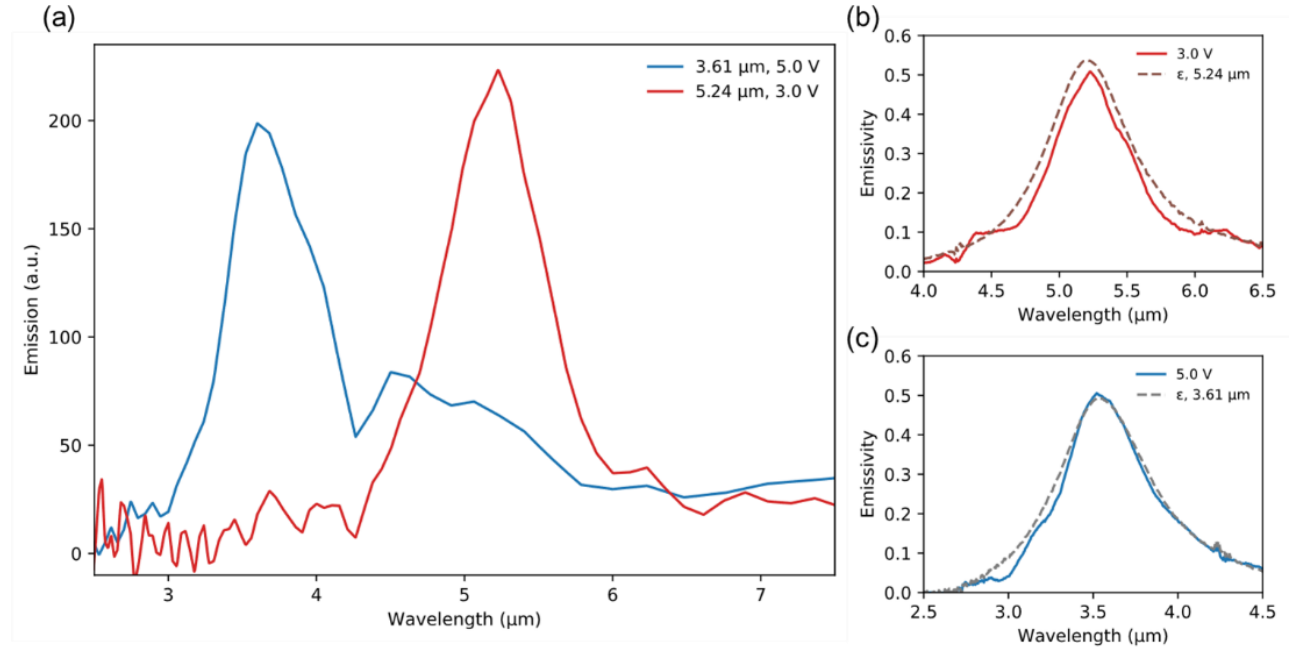


Figure 5.5 (a) The direct emission spectra of metasurfaces for both $5.24\ \mu\text{m}$ and $3.61\ \mu\text{m}$ resonances. (b-c) The recovered emissivities from direct emission measurement and are consistent with the ones from reflectance measurements.

6. Direct thermal infrared emission measurement by microscopic FTIR [Physical Review Applied, 19 (3), 034040, 2023.]

With the advent of novel thermal infrared microdevices, conventional characterizations, such as the simple reflectance that indirectly indicates the emissivity of a device by Kirchhoff's law, are far from enough. Directly catching the thermal infrared signal can provide a lot more information concerning the device performance and becomes an indispensable process for future device applications in lighting, sensing, and energy harvesting. Compared to typical fluorescence or laser spectroscopy, the thermal infrared light from microdevices is rather weak and buried in a strong environmental thermal background and different kinds of electronic noises during the signal collection. Here, we set up a microscopic lock-in Fourier-Transform Infrared Spectrometer (FTIR) system that can significantly reduce both the optical and electrical noises allowing us to measure the direct emission resonances from the electrothermal metasurfaces. In this report, we first elaborate the signal detection process focusing on the combination of global and Joule heating in boosting the signal-to-noise ratio (SNR). This microscopic lock-in FTIR can then serve as an ultrasensitive detector that is fast enough to record the spectral-resolved response of the device in the frequency domain. Finally, a blackbody-free calibration is proposed to give the true emission power from the device.

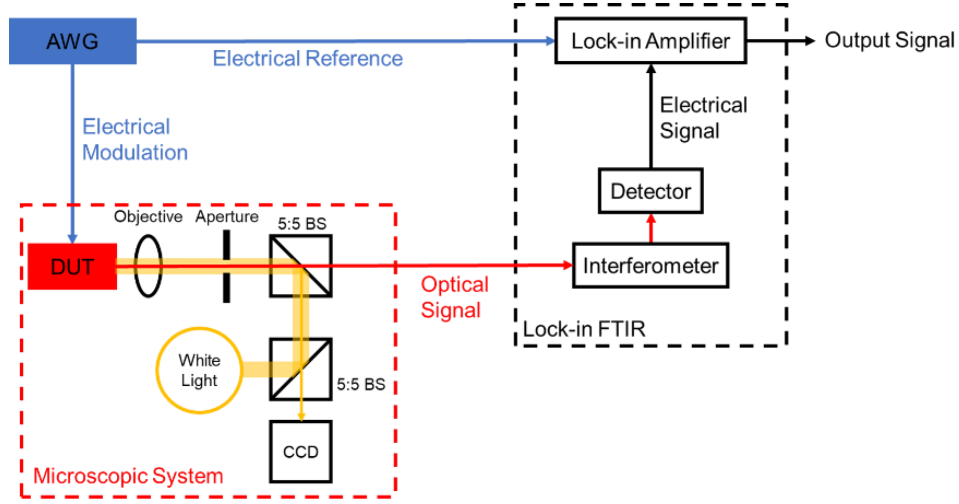


Figure 6.1 Microscopic lock-in FTIR setup.

As shown in Fig. 6.1 for the microscopic lock-in FTIR setup, the modulated optical signal of the device under test (DUT) is extracted by a microscope with an aperture. The microscope allows us to find and focus on the DUT active area when it is illustrated by a white light source and helps to extract the DUT signal when it directs the thermal infrared light into the FTIR. The aperture can block the noises from structures surrounding the DUT and provide some spatial-resolved capability when the DUT emission is inhomogeneous. The signal-to-noise ratio (SNR) improvement is mainly finished by the lock-in FTIR subsystem, whose lock-in amplifier (LIA) works as a dynamic noise filter centered at the modulation frequency.

To describe the overall signal process, we start with the voltage modulation from an arbitrary wave generator (AWG) as $V(t)$, and then the Joule heating generated in the DUT is $Q(t) = V^2(t)/R$ where R is the DUT electrical resistance. The DUT temperature response ΔT can be given by

$$T(t) - T_0 = \Delta T(t) = Q(t)R_t = \frac{V^2(t)R_t}{R}$$

where R_t is the effective thermal resistance of the substrate underneath the metasurface and T_0 is the temperature of the substrate and is set by the global heating. In this lumped thermal circuit model, we ignore the natural convection and radiation from the top of metasurface for its temperature dependence. With the known temperature $T(t)$, we can then theoretically compute the spectral thermal emission based on Planck's law in a time-pointwise manner

$$I[\lambda, T(t)] = \varepsilon(\lambda)I_{BB}[\lambda, T(t)].$$

Which is functional of t and is difficult to handle in practice. To simplify the process, we need to separate the time dependence of temperature $T(t)$ as

$$T(t) = T_0 + \Delta T(t) = T_0 + T_{DC} + T_m(t) = T_{avg} + T_m(t),$$

with T_{DC} is the time-independent part of temperature generated by the root-mean-square (RMS) Joule heating power and $T_m(t)$ is the time-dependent part of temperature for modulated thermal emission. We can further group T_0 and T_{DC} as the average temperature T_{avg} measured from the QFI thermal mapping measurement. Since $T_m(t) \ll T_{avg}$, the functional $I_{BB}(\lambda, T)$ can be Taylor expanded with respect to T with the linearization as

$$I(\lambda, T) = \varepsilon(\lambda)I_{BB}(\lambda, T_{avg}) + \varepsilon(\lambda) \frac{\partial I_{BB}}{\partial T}(\lambda, T_{avg})T_m(t),$$

which is then simplified as a function of t . This thermal emission (optical signal) together with the optical noises $I_{ON}(\lambda, T_{ON})$ is then transformed into the electrical voltages by the MCT detector as

$$V_{in}(\lambda, t) = R(\lambda)[I(\lambda, T) + I_{ON}] = R(\lambda) \left[\varepsilon(\lambda)I_{BB}(\lambda, T_{avg}) + \varepsilon(\lambda) \frac{\partial I_{BB}}{\partial T}(\lambda, T_{avg})T_m(t) + I_{ON}(\lambda, T_{ON}) \right] + I_{EN},$$

where $R(\lambda)$ is the response function whose wavelength dependence comes from both MCT responsivity and all the optical path components. $I_{ON}(\lambda, T_{ON})$ is the thermal infrared (optical) background noise including the room-temperature environmental thermal background and radiation from structures surrounding the DUT. I_{EN} is the unavoidable electrical noise generated during the data collection process, which becomes critical when we are extracting weak signals from micro-devices.

The LIA noise reduction is quite mature and straightforward, but how to excite the electrothermal metasurface with the combination of global heating and Joule heating in the modulation frequency remains unexplored. They play an important role in SNR improvement and emission power calibration. Starting with the noise spectral density of the input $V_{in}(\lambda, t)$, as shown in Fig. 6.2(a), we can clearly observe the drastically increasing noises when the modulation frequency approaches to 0 Hz, where it is impossible to measure the thermal emission signal among these color noises. However, with a modulation frequency of 2600 Hz, the signals are moved to the spectral density with only small white noise, which can be further reduced by a low-pass filter. With increasing global temperatures, the signal intensities also increase because the global temperature T_0 makes $I_{BB}(\lambda, T_{avg})$, i.e., $T_{BB}(\lambda, T_0 + T_{DC})$ overlap better with $\varepsilon(\lambda)$, although global heating itself is not modulated at 2600 Hz. This is also shown in the equation of $V_{in}(\lambda, t)$ with the term $\partial I_{BB}(\lambda, T_{avg})/\partial T$ for the modulated part of signal. The drawbacks of the global heating can also be obviously found to significantly increase the non-modulated part of signal, which has no difference from the non-modulated noises I_{ON} or I_{EN} to the LIA. Fortunately, the dynamic reserve, i.e., the ratio of the largest tolerable noise to the full-scale signal, of modern LIAs typically can tolerate this amount of non-modulated global heating.

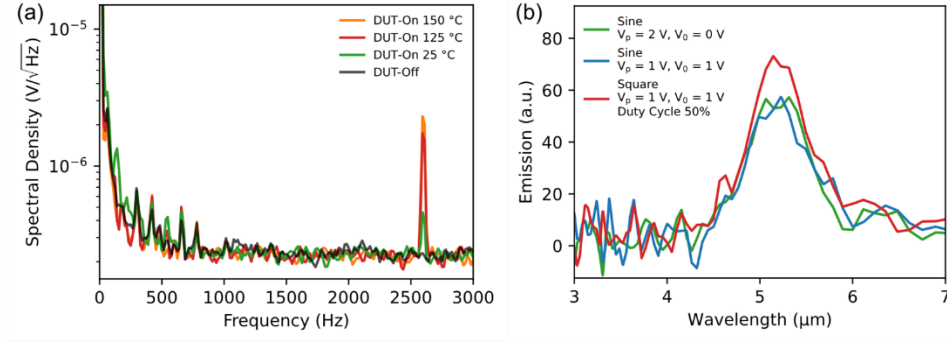


Figure 6.2 (a) Input spectral density for LIA with different global heating temperatures. (b) Measured signals with different Joule heating modes.

We then specify the Joule heating mode $Q(t)$, which plays a fundamental role in our LIA signal detection. More specifically, $Q(t)$ depends on the DC offsets and AC waveshapes. To figure out the influence of the DC bias on the final detected signal, we can choose the simplest sine waveshape with a DC offset as

$$V(t) = V_p \sin(\omega t) + V_0 ,$$

and then the modulated part of LIA input is

$$V_{in,m}(\lambda, t) = R(\lambda)\varepsilon(\lambda) \frac{\partial I_{BB}}{\partial T}(\lambda, T_{avg}) \left[\left(\frac{2V_0 V_p R_t}{R} \right) \sin(\omega t) + \left(-\frac{V_p^2 R_t}{2R} \right) \cos(2\omega t) \right] ,$$

which is caught by the LIA and mixed with a reference $V_r = \sin(\omega_r t + \Delta\phi)$ where $\Delta\phi$ is the fixed phase difference between the input signal V and reference V_r , and can be finally cancelled out with a dual-phase demodulation.

$$\begin{aligned} V_{mix} &= V_{in,m}(\lambda, t) \cdot V_r(t) \\ &= R(\lambda)\varepsilon(\lambda) \frac{\partial I_{BB}}{\partial T}(\lambda, T_{avg}) \left\{ \left(\frac{V_0 V_p R_t}{R} \right) \cos[(\omega_r - \omega)t] \right. \\ &\quad - \left(\frac{V_0 V_p R_t}{R} \right) \cos[(\omega_r + \omega)t] - \left(\frac{V_p^2 R_t}{4R} \right) \sin[(\omega_r + 2\omega)t] \\ &\quad \left. + \left(\frac{V_p^2 R_t}{4R} \right) \sin[(\omega_r - 2\omega)t] \right\} \end{aligned}$$

If we pick the ω term with $\omega_r = \omega$, the DC term output after the low-pass filter is

$$V_{out,\omega} = R(\lambda)\varepsilon(\lambda) \frac{\partial I_{BB}}{\partial T}(\lambda, T_{avg}) \frac{V_0 V_p R_t}{\sqrt{2}R} ,$$

and if we pick up the 2ω term with $\omega_r = 2\omega$,

$$V_{out,2\omega} = R(\lambda)\varepsilon(\lambda) \frac{\partial I_{BB}}{\partial T}(\lambda, T_{avg}) \frac{V_p^2 R_t}{4\sqrt{2}R} ,$$

where the $\sqrt{2}$ is for the RMS power output.

Therefore, it is interesting to find that we can use the first harmonic for the Joule heating signal pickup, unlike the previous studies which always focus on the full-wave rectified sine wave with 2ω for reference frequency. More importantly, the filtered output signal $V_{out,\omega}$ is proportional to the bias V_0 and it means we can go beyond the power limit of an arbitrary wave generator (AWG) by in series connecting a DC source to it when more power is needed to improve SNR. Although counterintuitive, the DC Joule heating is fundamentally different from the global heating since it contributes to the modulated signal based on its electrical interference with the AC joule heating. It is also noted that $V_{out,\omega} = V_{out,2\omega}$ with $V_0 = V_p/4$, and with this single input, the LIA should pick up the output of ω the same as 2ω (several more spectra may be needed here). Or the above analysis can be verified by choosing two inputs: a sine pulse train with $V_p = 1V$ and $V_0 = 1V$ and set the LIA to pick up the signal with ω , and this should be the same as a sine wave with $V_p = 2V$ and $V_0 = 0V$ when the LIA picks up the signal with 2ω , as shown in Fig. 6.2(b).

The output signal can also benefit from the waveshape of the modulation. If we change the sine pulse train to a square pulse train with the same $V_p = 1V$ and $V_0 = 1V$, the first harmonic picked up by the LIA should be $(4/\pi)V_{out,\omega}$. This increment in output signal can be clearly observed in Fig. 6.2(b) with the peak ratio at $5.24 \mu m$ is about 1.22. More importantly, the square waveshape introduces the duty cycle, which is an additional control factor that can increase the contrast between T_{DC} and $T_m(t)$. Since the LIA only picks up the $T_m(t)$ emission signal, it is possible for us to use a pulse train with a high peak but a small duty cycle making the $T_m(t)$ emission power high but the average power low, if the DUT can response to this narrow pulse (it is possible for modern thermal infrared microdevices). It can be beneficial not only to protect the DUT from high power injection but also to achieve a highly spatial-localized emission. For general different waveshapes, only the first harmonic will be picked up and the previous conclusions of DC bias V_0 in Joule heating keep valid.

With the increasing modulation frequency, the emission decreases since the DUT cannot respond to the fast-changing electrical excitation, as shown in Fig. 6.3(a). This changing of emission peak height with the modulation frequencies can be fitted and gives the DUT cutoff frequency as $65553.88 Hz$, which corresponds to the response time of $15.25 \mu s$, as shown in Fig. 6.3(b). The response time can then be verified by the fall time from our transient measurement as $14.72 \mu s$, as shown in the inset.

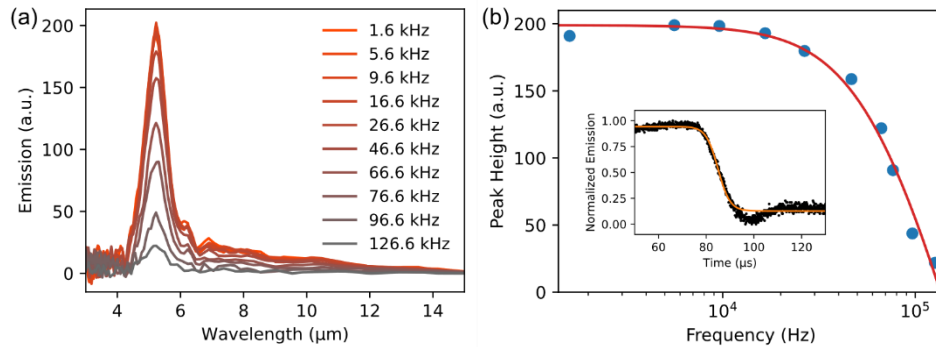


Fig. 6.3 (a) Emission spectra for different modulation frequencies up to no response from DUT. (b) The emission peak height decreases with increasing modulation frequencies showing the DUT cutoff frequency as 65553.88 Hz, which is consistent the insert transient measurement showing the DUT response time as 14.72 μ s.

The importance here is that our dynamic response characterization is spectral resolved, i.e., for each modulation frequency, we can obtain the full spectrum of the emission. This can be particularly useful when the modulation depth is no longer the emission power. For example, some tunability may induce the emission peak shift, and our spectral-resolved response measurement can directly monitor the peak changing, which may not change simultaneously with the emission power.

In addition, with the help of LIA, our lock-in FTIR system is ultrasensitive, giving a clear contrast of the emission signal from on-state to off-state even it is buried in very strong noises. The measurable speed range is limited by the MCT detector ($> 20\text{MHz}$) and the LIA bandwidth (50MHz). It is also noted that the time constant of the low-pass filter, which is typically long for noise reduction, has nothing to do with this frequency-domain measurement since the signal at this point is at DC after mixing.

It is also worth noting that our temperature modulation $T(t)$ follows strictly with the Joule heating $Q(t)$ based on a simple thermal circuit with the effective thermal resistance R_t of the substrate. A more detailed analysis can be given to solve the heat diffusion equation, including the spatial effect, to give a more accurate time-dependent $T(t)$, and thus the time-dependent emission $I(\lambda, t)$, which may have some different frequency dependences in different modulation frequency regimes. Ideally, our system can monitor these phenomena as well when denser frequency points are given.

To specify this system response function for our emission power calibration, we choose the thermal excitation as the square pulse train

$$V = V_0 + \sum_{n=1}^{\infty} \frac{4V_p}{n\pi} \sin(nD\pi) \cos(n\omega t)$$

with $V_p = V_0 = V/2$, i.e., the low-level voltage is always set to 0 V and the high-level voltage is set to V. The duty cycle D is set to 50% and the modulation frequency is 2600 Hz. Only the first harmonic is picked by the LIA and its RMS output voltage is

$$V_{out,\omega} = R(\lambda)\varepsilon(\lambda) \frac{\partial I_{BB}}{\partial T}(\lambda, T_{avg}) \frac{V^2 R_t}{\sqrt{2}\pi R}$$

with $(V^2 R_t)/(\sqrt{2}\pi R)$ having the dimension as temperature. To avoid complicated modelling and measurement, we can group

$$m(\lambda, V) = R(\lambda) \frac{V^2 R_t}{\sqrt{2}\pi R}$$

as the effective response function with the unit $[a.u. \cdot K/nW]$ ($V_{out,\omega}$ has an arbitrary unit), and $(\partial I_{BB})/\partial T$ has the unit $[nW/K]$. The $I_{BB}(\lambda, T)$ is the Planck's law in nW computed as

$$I_{BB}(\lambda, T) = \frac{c_1 \times A \times \Omega \times 10^9}{\lambda^5 \left(e^{\frac{c_2}{\lambda T}} - 1 \right)}$$

with $c_1 = 1.191042 \times 10^8 W/m^2 \cdot sr \cdot \mu m^{-4}$ and $c_2 = 1.438775 \times 10^4 K/\mu m$, after including the size $A = 50 \mu m \times 50 \mu m$ of metasurface and the solid angle $\Omega = 0.53$ of the microscope (Thorlabs reflective objective LMM40X-P01).

Therefore, the effective response function $m(\lambda, V)$ is a double-variable function of λ and V , but is independent of the temperature T_{avg} and thus can be given by

$$m(\lambda, V) = \frac{V_{out,rms}(\lambda, V, T_{avg,1}) - V_{out,rms}(\lambda, V, T_{avg,2})}{\varepsilon(\lambda) \left[\frac{\partial I_{BB}}{\partial T}(\lambda, T_{avg,1}) - \frac{\partial I_{BB}}{\partial T}(\lambda, T_{avg,2}) \right]},$$

where $T_{avg,1}$ and $T_{avg,2}$ are changed by the global heating with the same modulation voltage. We then specify the expression by controlling the input variables as either λ' or V' . As shown in Fig. 6.4(a), with different modulation voltages $V = V'$, we have the system responses $m(\lambda, V')$ as the functions of λ , which are parallel to each other at least in the range near the resonance where the SNR is high. That means the $R(\lambda)$ inside $m(\lambda, V)$ is independent of the modulation voltage. Indeed, the $R(\lambda)$ is pure from the MCT and optical path signal collection. As the wavelengths approach $4.5 \mu m$ or $6.0 \mu m$, the CO_2 and H_2O absorption disturb the response function a lot. We also include another electrothermal metasurface resonating at $3.61 \mu m$ and similar parallel response functions $m(\lambda, V')$ are shown in Fig. 6.4(b). Again, when the wavelengths are far away from the resonance, the response function tends to be deformed due to the CO_2 absorption near $4.5 \mu m$ and KBr beam splitter limit at around $3 \mu m$. Furthermore, we choose the resonance wavelength at each metasurface (gray dashed lines) and plot the response function $m(\lambda', V)$ for a specific wavelength as a function of V . A clear parabolic relation $m \propto V^2$ can be observed in Fig. 6.4(c) and follows well the analytical expression of the effective response function.

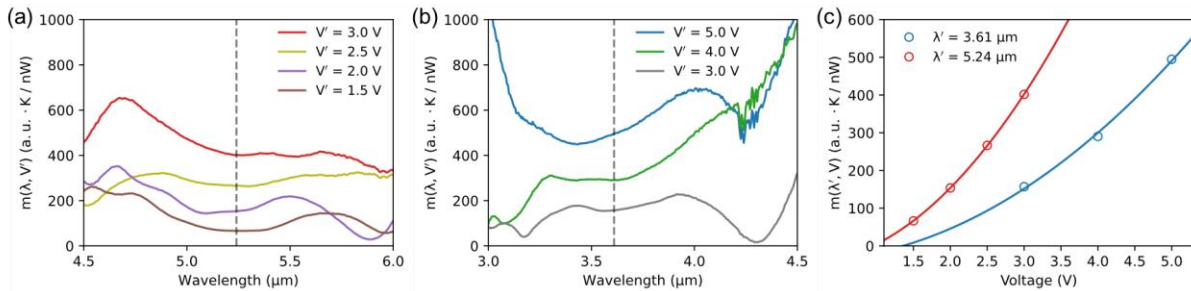


Figure 6.4 (a) The effective response function $m(\lambda, V')$ as a function of wavelength λ for different voltages $V = V'$. (b) The $m(\lambda, V')$ for another electrothermal metasurface with resonance at $3.61 \mu m$. (c) The $m(\lambda', V)$ as a function of voltage V for resonance peaks $\lambda' = 5.24 \mu m$ and $3.61 \mu m$.

Finally, with the measured effective response function $m(\lambda, V)$, we can recover the emissivity with

$$\varepsilon(\lambda, T_{avg}, V) = \frac{V_{out,\omega}}{\frac{\partial I_{BB}}{\partial T}(\lambda, T_{avg}) R(\lambda) \frac{V^2 R_t}{\sqrt{2\pi R}}} = \frac{V_{out,\omega}}{\frac{\partial I_{BB}}{\partial T}(\lambda, T_{avg}) m(\lambda, V)}.$$

As shown in Fig. 6.5, with different global heating T_0 for T_{avg} and modulation voltage V , we can have the same emissivity as the one measured from the FTIR reflectance. This verifies our whole calibration analysis.

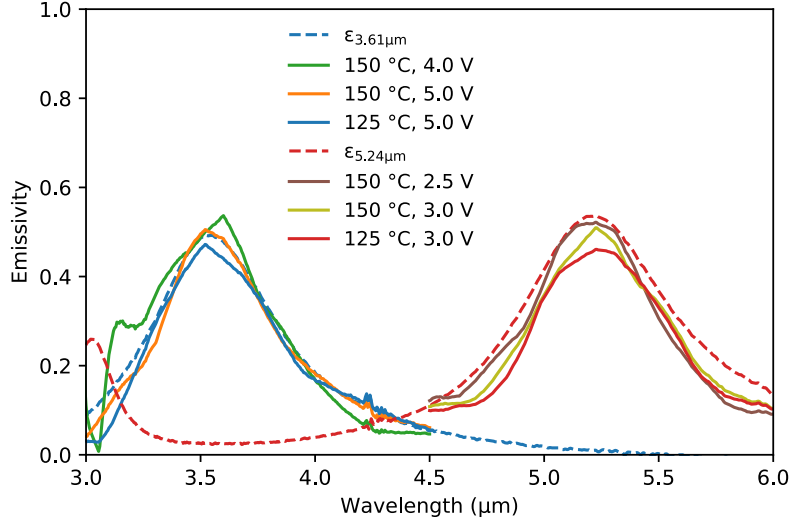


Figure 6.5 Recovered emissivity from the effective response function with different global heating and modulation voltages.

With more careful electrical and thermal measurements, it is possible for us to obtain the specific values of R and R_t and then for the true emission power from the metasurfaces. This blackbody-free calibration can be beneficial since little error comes from the measurement system, and with the DUT themselves, only measurements in different global heating or modulation voltages are needed. In addition, a blackbody is typically hard to achieve for Joule heating devices since conductivity always makes reflectivity. On the other hand, some infrared LED sources are typically expensive, cooling-required, and even uncalibrated.

7. Far-field thermal radiation from degenerate resonant thermal emitters [1 manuscript in submission to *Science*]

We designed and demonstrated a pair of thermal-driven micro-sized pixel twins inspired by the leafhopper-produced brochosome structures (Fig. 7.1(a) and inset), which are hollow spherical structures with distributed open pores bridging the inner and outer surfaces. These brochosome-like pixel (BLP) twins are featured with either open pores (op-BLPs, Fig. 7.1(b)) or closed pores (cp-BLPs, Fig. 7.1(c)). The diameter of pores d and the diameter of BLPs D are the key design parameters. We started the design by adopting the ratio $d/D = 0.28$, which is the average value acquired from natural brochosomes generated by various leafhoppers, and then optimized such ratio for the desired functions. As illustrated in Fig. 7.1(d), they show distinct emissivities when thermally excited, which serve as infrared counterparts of the binary states 0 and 1 used in the

traditional electronic approaches. Therefore, information can be stored in the ‘bitmap’ arranged with the pixel twins and extracted under infrared imaging systems. At the same time, BLPs of the two types share similar appearances under visible imaging systems, so that the stored information is encrypted against visible-range reading. To achieve the proposed encryption functions, we simulated the optical responses of both BLPs based on their 3D models in both the visible and infrared wavelength ranges to demonstrate the wavelength-dependent distinguishability between the twins, which sets the foundation of the physical mechanism of the encryption. We then fabricated BLP arrays of both kinds to experimentally measure their infrared emissivities and a 2.5:1 contrast was achieved, which agrees well with our simulations. Finally, bitmaps arranged with the BLP twins were designed and fabricated to store both simple patterns and QR codes as proofs of concept. Both the visible and infrared images of the fabricated bitmaps were shown to demonstrate that the stored information was encrypted for infrared extraction only. Such a wavelength-dependent visibility makes the BLP twins potentially applicable for anticounterfeiting or data encryption.

The total irradiance reaching the image plane of an imaging system from a BLP determines its appearance. Furthermore, the total irradiance can be decomposed into the thermal emission from the BLP, which dominates in the infrared spectrum since BLPs will be thermally excited for data extraction, and the scattered light from the BLP with respect to the environmental illumination in the visible spectrum. To enable thermal encryption, distinctive infrared emission but similar visible scattering of op-BLPs and cp-BLPs are desired. To demonstrate this, we performed finite-difference time-domain (FDTD) simulations for both types of BLPs in both the infrared and the visible ranges. The diameter of BLPs D is designed to be 20 μm , and the pore size d is 3.7 μm , which fulfills the requirement of our infrared measurement, as described below. The material of BLPs is chosen to be nickel (Ni) or other reflective metals, which provides a high permittivity contrast between the BLPs and the background.

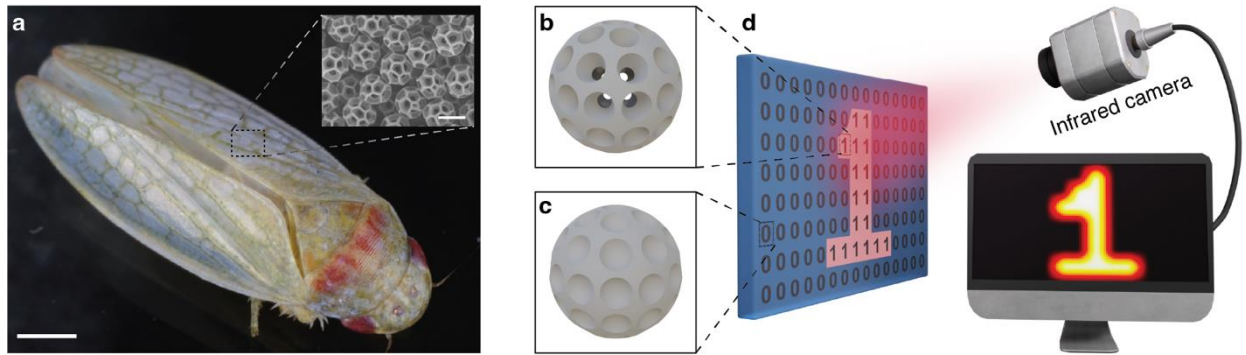


Figure 7.1 Encryption by brochosome-like pixel twins. (a) An optical image of a leafhopper *Gyponana Serpenta*. Scale bar: 1 mm. Inset: a scanning electron microscopy (SEM) image of leafhopper generated brochosomes. Scale bar: 500 nm. (b) and (c), 3D models of brochosome-like pixel (BLP) twins with open pores (op-BLPs) and closed pores (cp-BLPs), respectively. (d) Schematic of the encryption and extraction of information by BLPs. Information is encrypted in the binary array formed by BLPs, which cannot be distinguished by human eyes but can be extracted under infrared imaging systems.

In the IR range, we simulated the spectral absorption cross sections (σ) for both types of BLPs. The magnitude of σ also indicates the emission capability of the two structures, based on Kirchhoff's law. Figure 7.2(a) showed the results for a single op-BLP and a single cp-BLP. For comparison, we included the result of a Ni sphere with the same diameter (20 μm). Within the wavelength range from 2.5 μm to 8 μm , the op-BLP showed an enhanced absorption cross section compared to those of the cp-BLP and the sphere. The contrast of σ between the op-BLP and cp-BLP structures can be further illustrated in Fig. 7.2(b), where we plotted their excess absorption cross section ($\Delta\sigma = \sigma_{BLP} - \sigma_{sphere}$) with respect to that of the sphere. The enhanced σ of the op-BLP within this wavelength range is attributed its open pores and thus the exposed cavity. As shown in Fig. 7.2 (c) and (d), the electromagnetic (EM) field penetrates the op-BLP (Fig. 7.2(c)) and is trapped within the exposed cavity, and eventually absorbed due to the electron damping effect of Ni. Such a cavity effect is not observed in the cp-BLP due to the lack of open pores and exposed cavity (Fig. 7.2(d)), which results in the contrast of σ between these two structures. Note that such a high contrast gradually decreases after it reaches its maximum value at the wavelength around 5 μm , because the long-wavelength EM waves cannot penetrate BLPs even with the existence of the open pores. In our previous work, we showed that the cutoff wavelength of the penetration is around 1.5 times of the characteristic size of the pores. We, therefore, choose the size of BLPs accordingly to ensure a strong infrared contrast over the entire working range of our infrared imaging system, but it can be optimized for other wavelengths as well. Also, the multi-pore structures of BLPs are critical to achieve such the high contrast because simple spherical structures with fewer or only one open pore will reduce the amount of light reaching the cavity and thus will result in reduced absorption cross sections.

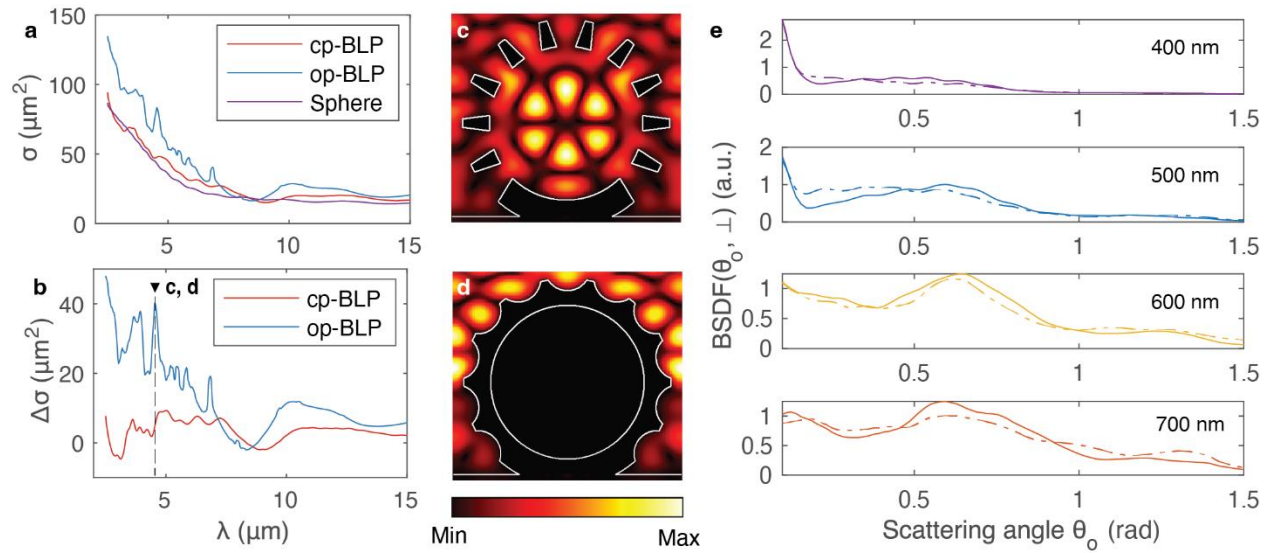


Figure 7.2 Optical characteristics of single brochosome-like pixels (BLPs). (a) Simulated absorption cross-sectional areas σ of a BLP with open pores (op-BLP), a BLP with closed-pores (cp-BLP), and a sphere. The diameter of the simulated BLPs and sphere is 20 μm , and the material is nickel (Ni). (b) Excess absorption cross-sectional areas $\Delta\sigma$ of BLPs compared to the sphere. Dashed lines marked the wavelength, 4.5 μm , at which field profiles are shown. (c) and (d) Simulated spatial distribution of the magnitude of the electrical field (wavelength is 4.5 μm) near an op-BLP and a cp-BLP, respectively. e, Simulated scattering distribution functions in the visible wavelength range for both a

cp-BLP (solid lines) and an op-BLP (dashed lines) with respect to normal illumination. Similar visible scattering behaviors of both the BLPs ensure that they cannot be easily distinguished in the visible range.

In the visible range, the bi-directional scattering distribution function (BSDF) characterizes the scattering behavior of BLPs, and thus their appearances under visible imaging systems. Conventionally, acquiring $\text{BSDF}(\theta_o, \theta_i)$ requires the simulations/measurements of the energy distribution at any given scattering angle θ_o with respect to illuminations from various directions θ_i . Given that microscopies need to be used to view microscale BLPs, where illuminations will mostly come from the vertical direction, we limited the visible range simulation of BLPs to the scattering distribution functions with respect to the normal illumination only, i.e., $\text{BSDF}(\theta_o, \perp)$. In Fig. 7.2(e), we compared the simulated results for both an op-BLP (solid lines) and a cp-BLP (dashed lines) at various visible wavelengths. Within the entire visible spectrum, their scattering distribution functions turn out to be similar, which can be attributed to the exact same arrangement of the surface features (either open pores or closed pores) in both structures. Such similar scattering distribution functions ensure that the pixel twins cannot be easily distinguished under visible imaging systems, and thus provide the encryption for the information coded in the BLP bitmaps.

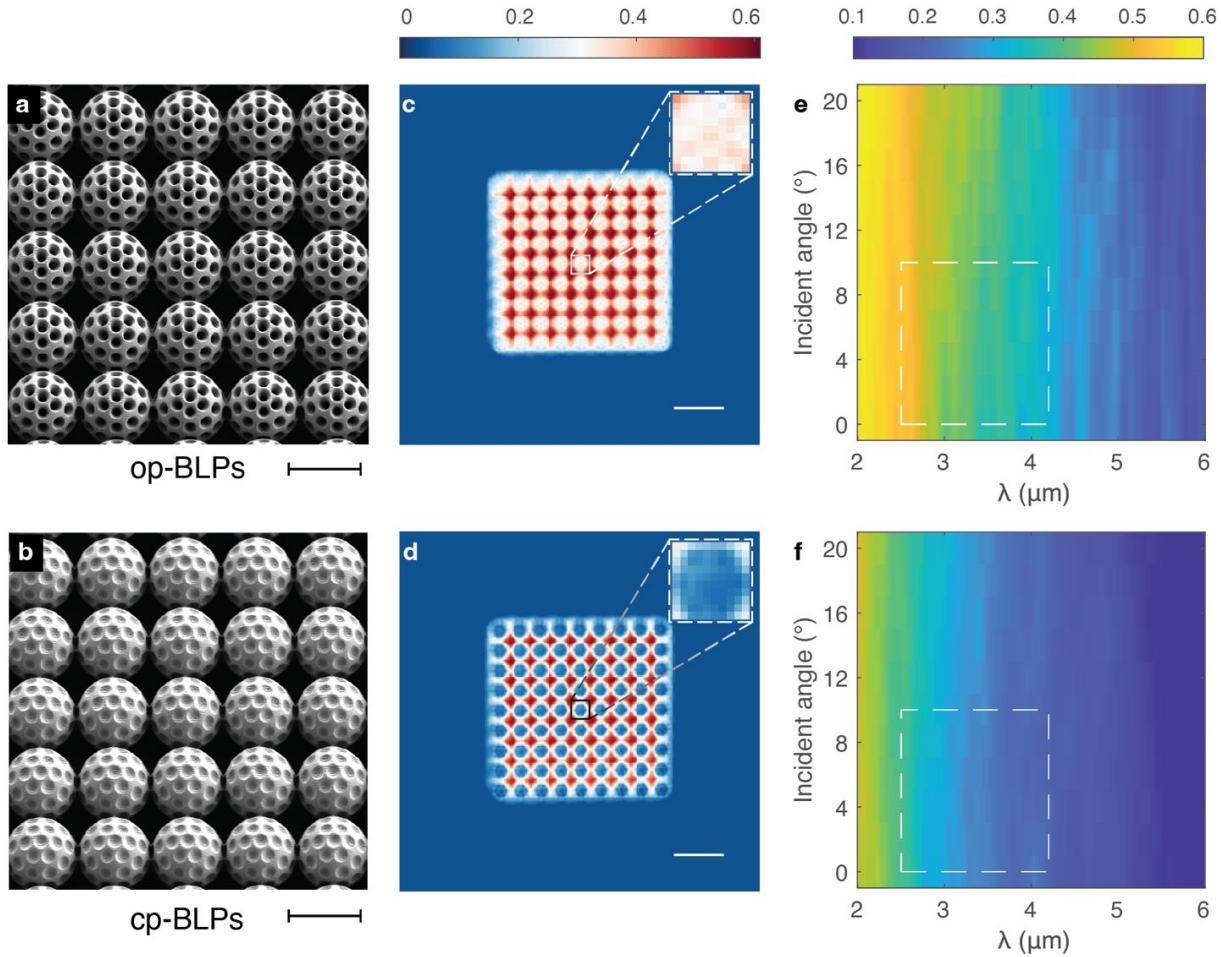


Figure 7.3 Emissivities of fabricated brochosome-like particle arrays. (a) and (b) SEM images of fabricated op-BLP array and cp-BLP array, respectively. Scale bar: 20 μm . (c) and (d) Measured spatial distribution of emissivities of op-BLP and cp-BLP arrays, respectively. Scale bar: 50 μm . Insets: Typical local enlarged images of single BLPs from which their emissivities are evaluated. (e) and (f) Simulated spectral-directional absorptivity of op-BLP and cp-BLP

arrays, respectively. The averaged emissivities are integrated within domains are marked by the white boxes, which match well with the spatially averaged experimental results.

According to Kirchhoff's Law, the difference between absorption cross sections of op-BLPs and cp-BLPs results in an emissivity contrast, which can be captured and measured by infrared imaging systems. To quantify such contrast, we fabricated BLP arrays of both kinds respectively (see Fig. 7.3(a) and (b)) and measured their spatial distributions of emissivity with an infrared thermal mapping system. Both the measured arrays consist of 9-by-9 BLPs whose diameters are 20 μm and were fabricated by the two-photon nanoscale 3D printing method and then coated with ~ 100 nm Ni. Figures 7.3(b) and (c), plot the spatial distributions of the measured emissivities for the op-BLP and cp-BLP arrays, respectively, from which the spatially averaged emissivities were calculated. Specifically, we calculated the emissivities for both arrays by averaging among the central 7 by 7 BLPs (the BLPs located at the edges of the two arrays are excluded to eliminate the edge effect), from which the averaged emissivity is evaluated to be 0.419 for the op-BLP array, and 0.3073 for the cp-BLP array. We also performed the Fourier transform infrared reflection measurement for both the arrays, from which an equivalent difference in their infrared emissivities can be found. Here we noticed the voids between neighboring BLPs in both arrays showed high emissivities (~ 0.5) because these voids also act as cavities and thus enhance the local emission in both arrays, which causes the underestimation of the emissivity contrast between different BLPs since they will act as single pixels. Therefore, we also calculated the averaged emissivities of single BLPs of both kinds by averaging local emissivities within randomly chosen parts containing only one BLPs (typical ones are shown in insets of Fig. 7.3(b) and (c)). The averaged emissivities of single op-BLPs and cp-BLPs are then evaluated to be 0.3329 and 0.1329, respectively, which leads to an infrared contrast ratio around 2.5: 1. In comparison, the optical images of the two arrays were analyzed via the quantitative color pattern analysis method, and they shared same pattern energy distribution, indicating the indistinguishability of the two arrays by human eyes.

To verify the infrared measurement, we performed FDTD simulations of the spectral-directional absorptivity $\alpha_{\lambda,\theta}$ for both the measured arrays, where θ represents the illumination angle with respect to the normal direction, and the corresponding spectral-directional emissivity $\varepsilon_{\lambda,\theta} = \alpha_{\lambda,\theta}$ can be acquired based on Kirchhoff's Law. Therefore, the emissivity can be calculated as:

$$\varepsilon = \frac{1}{AB} \int_{\lambda_{min}}^{\lambda_{max}} \int_0^{\theta_{max}} \varepsilon_{\lambda,\theta} \sin\theta \cos\theta E_{\lambda,b}(\lambda, T) d\theta, \quad (7.1)$$

where

$$A = \int_0^{\theta_{max}} \sin\theta \cos\theta d\theta, \quad (7.2)$$

and

$$B = \int_{\lambda_{min}}^{\lambda_{max}} E_{\lambda,b}(\lambda, T) d\lambda, \quad (7.3)$$

in which $E_{\lambda,b}(\lambda, T)$ represents the blackbody radiation. θ_{max} , λ_{min} , λ_{max} , and T are set to 10° , 2.5 μm , 4.2 μm , and 323.15 K, respectively, to match with the specifications of the thermal mapping system used in measurement. Figure 7.3(e) and (f) showed the simulated spectral-directional emissivity (absorptivity) of the two arrays, in which the white lines highlight the

integration region in Eq. 7.1, according to which the emissivities of the op-BLP array and cp-BLP array are estimated to be 0.449 and 0.301, respectively, and both are consistent with the measured results.

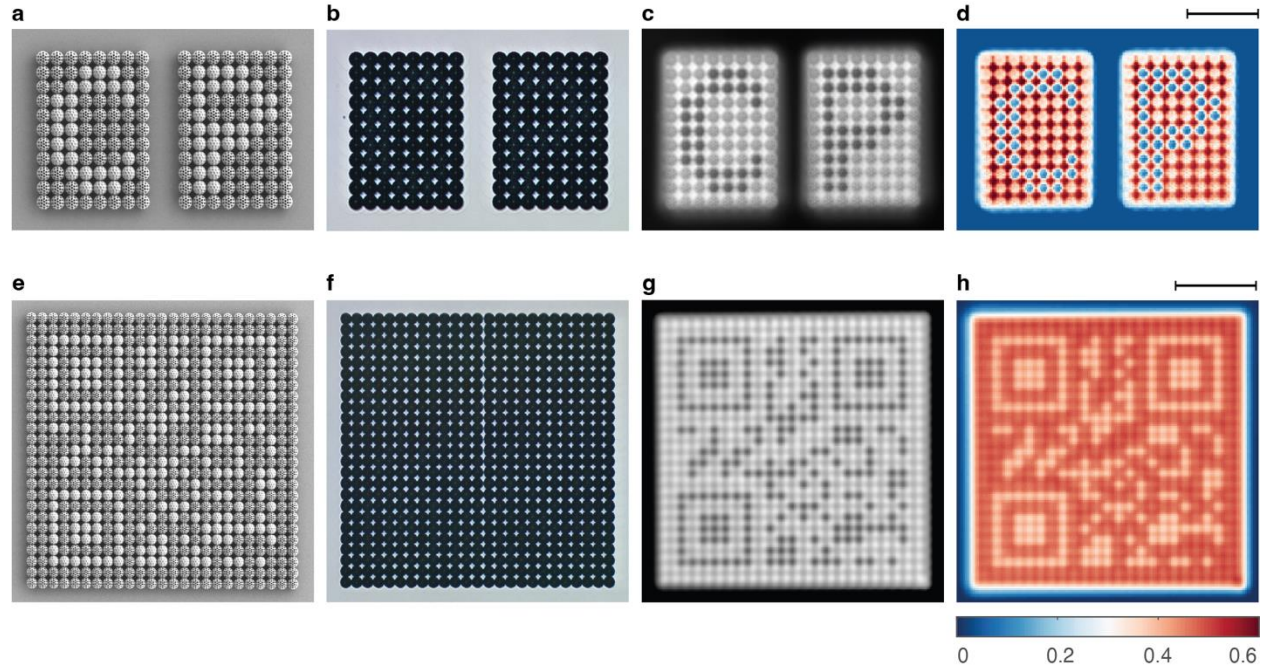


Figure 7.4 Infrared encryption by brochosome-like pixel arrays showing letters ‘C’ and ‘P’, and a quick-response code (QR code). (a) and (e) SEM images of BLP arrays patterned to encrypt letters ‘C’ and ‘P’ and a QR code (storing the address of the homepage of Carnegie Mellon University), respectively. (b) and (f) Optical images of the BLP arrays. The encrypted information is invisible. c and g, Infrared images of the BLP arrays, revealing the encrypted information. (d) and (h) Calculated spatial distribution of emissivity based on the infrared images, showing the contrast between the encrypted patterns and the background. The infrared image and emissivity distribution of the QR code sample are defocused for an easier scan. Scalebars: top row 100 μm , bottom row 150 μm .

The BLP twins represent a pair of binary states under infrared imaging systems due to their emissivity contrast, meanwhile their similar appearances under visible imaging systems make them indistinguishable to human eyes. Such a wavelength-dependent distinguishability sets the foundation of the encrypted data storage based on BLPs. After experimental optimizations of BLPs in terms of their arrangement, the shape, and the size of the pores, we demonstrated the proposed encryption method by two examples as shown in Fig. 7.4. As the first example, Figure 7.4(a) shows a SEM image of two BLP arrays, where we patterned the letters ‘C’ and ‘P’ using the cp-BLPs in the background formed by op-BLPs. Here the size of BLP pixels is 20 μm , resulting in an effective pixel density of 1270 pixel per inch, which is more than 5 times denser than typical micro-LED displays used in smart devices. Figure 7.4(b) shows the visible optical images of the two arrays using an optical microcopy with a 20X magnification objective, from which the two kinds of BLPs are indistinguishable and no patterns can be identified. However, the emissivity difference between the BLPs of different kinds provides ample contrast to visualize the encrypted patterns in the infrared range, as shown in Fig. 7.4(c), which is taken with the same magnification but by the

infrared camera in our thermal mapping system. Figure 7.4(d) plots the calculated spatial distribution of emissivity based on Fig. 7.4(c), for which one can tell the typical emissivity of each individual cp-BLPs (pixels forming the letters) is around 0.15, compared to ~ 0.35 for individual op-BLPs, further verifying the theoretical contrast ratio around 2.5: 1. Furthermore, thanks the highly symmetrical arrangement of pores on BLP surfaces, the two kinds of pixels are able to maintain their infrared emission contrast even when viewed from different angles. The contrast between the BLP twins, and consequently the ‘C’ and ‘P’ patterns can still be clearly viewed under IR microscopes even when they are tilted up to 6° . Note here the maximum viewing angle is not limited by the BLPs, but the focal depth of the IR microscope. Such a property provides tolerance to angular misalignments in the imaging system and thus makes the data extraction process more robust.

By combining the BLP arrays with other data encoding protocols, the encryption of multi-media information can be achieved. Here, in the second example, we fabricated a BLP array patterned into a quick-response code (QR code, version 1, ECC level L) containing the address of the homepage of Carnegie Mellon University, with the cp-BLPs forming the QR code and op-BLPs for the background, as shown in Fig. 7.4(e). The size of the QR code is 21 pixel by 21 pixel with a 2-pixel margin on each sides, resulting in a physical size of 0.5 mm by 0.5 mm. Compared with other nano/microstructure-based QR codes, our pixel size, as well as total QR code size, is much smaller than those based photonic crystals, and on par with the one based on imprinted polyvinyl alcohol nanostructures, even though our QR code is designed to work in a much longer wavelength. Following the similar idea as the first demo, the QR code is encrypted against the visible scanning, as shown in Fig. 7.4(f) for the visible optical image. In comparison, the infrared images of the BLP pixel array (Fig. 4(g)) and calculated emissivity (Fig. 7.4(h)) clearly reveal the encrypted QR code, thus allowing fast scanning by most QR code scanning applications on smart devices. Though counterintuitive, we found that blurring the IR images of the BLP QR code, which can be done either by slightly defocusing the IR camera while taking images, or numerically, by applying Gaussian filters or generalized Gaussian filters to well-focused IR images, will eliminate the local emissivity variations within single BLPs so that they can better represent the binary digits as in traditional QR codes, which will make the BLP QR code easier to scan. The application of the BLP encryption to QR codes provides protection against normal scanning tools yet retain the convenience of revealing the coded information with appropriate imaging devices working in the infrared range, which is also potentially applicable for anticounterfeiting.

In summary, inspired by leafhopper-generated brochosomes, we have designed a pair of brochosome-like pixel (BLP) twins with either open pores or closed pores to represent a pair of binary states in the infrared range. Due to their similar visible range scattering behavior, the BLP twins share a similar appearance under visible range imaging systems. Meanwhile, the open pores and thus the exposed cavity of op-BLPs lead to the enhanced absorption cross sections compared those with closed pores, which translates into a 2.5: 1 emissivity contrast between the two pixels and can be captured by infrared imaging systems. Such a wavelength-dependent distinguishability of the two kinds of pixels sets the foundation for the encrypted data storage. Theoretically, by optimizing the size and arrangement of the pores, BLPs with various emissivities could be designed to represent data/images in a continuous, instead of binary, manner. Ultimately, the designed BLPs allow encoding of data in a pixel-by-pixel manner, and only require simple thermal excitation for data extraction. This provides a new method of encrypted data storage or other technologies requiring selective visibility.

What opportunities for training and professional development has the project provided?

If the research is not intended to provide training and professional development opportunities or there is nothing significant to report during this reporting period, state "Nothing to Report." Describe opportunities for training and professional development provided to anyone who worked on the project or anyone who was involved in the activities supported by the project. "Training" activities are those in which individuals with advanced professional skills and experience assist others in attaining greater proficiency. Training activities may include, for example, courses or one-on-one work with a mentor. "Professional development" activities result in increased knowledge or skill in one's area of expertise and may include workshops, conferences, seminars, study groups, and individual study. Include participation in conferences, workshops, and seminars not listed under major activities.

This project has provided opportunities for training and professional development of a number of students at Carnegie Mellon University.

Graduate Students:

In this project, 5 graduate students (Zhuo Li; Xiu Liu; Hyeonggyun Kim; Lin Jing; Jiayu Li) have been involved in this project. This project provided them extensive scientific training and professional development on photonics, heat transfer, and materials science. Zhuo Li, Xiu Liu, and Hyeonggyun Kim joined Carnegie Mellon University in Fall 2019, and selected the research topics in this project for their PhD theses. Hyeonggyun Kim is also co-advised by Prof. Gianluca Piazza, who is the Co-PI of this project. This project also partially supported Jiayu Li's and Lin Jing's thesis work. Jiayu Li and Lin Jing have defended their PhD theses in September 2020 and February 2023, respectively.

Undergraduate Students:

In this project, 4 undergraduate students, Yutian Cai, Andrew Fu and Muzaffer Tasoglu, Hannah Zhai, participated in the research work.

Course Development:

To enrich the education of nanophotonics and radiative heat transfer at Carnegie Mellon University, the latest research results gained from this project have been incorporated in three courses: "24-627 Nanophotonics: Fundamentals and Applications", "24-322 Thermal Fluids Experimentation", and "24-321 Heat Transfer" taught by the PI.

How have the results been disseminated to communities of interest?

If there is nothing significant to report during this reporting period, state “Nothing to Report.”

Describe how the results have been disseminated to communities of interest. Include any outreach activities that have been undertaken to reach members of communities who are not usually aware of these research activities, for the purpose of enhancing public understanding and increasing interest in learning and careers in science, technology, and the humanities.

(1) PhD Dissertations

- Jiayu Li, “Thermal Radiation from Nanophotonic Structures: Theory, Numerical Simulation and Applications,” *PhD thesis*. His PhD work was partially supported by this project, including nanoplasmonic emitter design and simulation, etc.
- Lin Jing, “Advanced Nanostructures for Thermal Management and Heat Regulation,” *PhD thesis*. Her PhD work was partially supported by this project.

(2) Journal Publications

- H. Salihoglu, J. Shi, Z. Li, S. Shen, “Nonlocal near-field radiative heat transfer by transdimensional plasmonics,” *under review in Physical Review Letters*.
- H. Yun, X. Liu, H. Salihoglu, Z. Li, S. Shen, “Electromechanically reconfigurable plasmonic nanoslits as active infrared perfect absorbers,” *submitted to APL Photonics*.
- L. Jing, X. Liu, H. Salihoglu, X. Luo, H. Yun, Z. Wang, S. Shen, “Electrically driven nanoantenna metasurface for coherent thermal emission,” *submitted to APL Photonics*.
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- X. Liu, H. Salihoglu, X. Luo, H. S. Yun, L. Jing, B. Yu, S. Shen, “Direct measurement of electrically modulated far-field thermal infrared emission and its dynamics,” **Physical Review Applied**, 19 (3), 034040, 2023.
- X. Liu, L. Jing, X. Luo, B. Yu, S. Du, Z. Wang, H. Kim, Y. Zhong, S. Shen, “Electrically driven thermal infrared metasurface with narrowband emission,” **Applied Physics Letters**, 121, 131703, 2022.
- H. Salihoglu, X. Liu, S. Shen, “Theory of thermal radiation from a nanoparticle array,” **Applied Physics Letters**, 121 (24), 241701, 2022.
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- J. Li, Z. Li, X. Liu, S. Maslovski, S. Shen, “Active control of thermal emission by graphene-nanowire coupled plasmonic metasurfaces,” **Physical Review B**, 106, 115416, 2022.
- X. Liu, Z. Li, Z. Wang, H. S. Yun, S. Shen, “Design and analysis of electrothermal metasurfaces,” **Frontiers in Energy**, 17, 134–140, 2022.
- Z. Li, J. Li, X. Liu, H. Salihoglu, S. Shen, “Wiener chaos expansion method for thermal

radiation from inhomogeneous structures,” *Physical Review B*, 104 (19), 195426, 2021.

- J. Li, B. Yu, S. Shen, “Scale Law of Far-Field Thermal Radiation from Plasmonic Metasurfaces,” *Physical Review Letters*, 124, 137401, 2020.

(3) Conference Posters/Presentations

- J. Li, S. Shen, “Scale law of far-field thermal radiation from plasmonic metasurfaces.” SPIE Photonic West, San Francisco, CA, February 2020.
- Z. Li, J. Li, S. Shen, “Thermal Radiation Spectra of Nanoplasmonic Structures: Simulation, Theory and Application.” Twenty-First Symposium on Thermophysical Properties, Boulder, CO, June 2021.
- Xiu Liu, Sheng Shen, “Design and analysis of electrothermal active metasurfaces.” *ASME IMECE 2022 Conference*, Columbus, OH, Oct. 2022.
- Zhuo Li, Sheng Shen, “Active Control of Thermal Emission by Graphene-Nanowire Coupled Plasmonic Metasurfaces.” *ASME IMECE 2022 Conference*, Columbus, OH, Oct. 2022.
- Hakan Salihoglu, Sheng Shen, “Far-field thermal emission from nanoparticle array.” *ASME IMECE 2022 Conference*, Columbus, OH, Oct. 2022.

(4) Invited Talks and Seminars

- Sheng Shen, “Nanoplasmonic Thermal Emission in Near- and Far-Fields.” *KITP Program and Workshop-Emerging Regimes and Implications of Quantum and Thermal Fluctuational Electrodynamics*, Santa Barbara, CA, July 2022.
- Sheng Shen, Sibley School of Mechanical and Aerospace Engineering, Cornell University, September 2021.
- Sheng Shen, Department of Mechanical Engineering, Stevens Institute of Technology, November 2021.
- Sheng Shen, Department of Mechanical and Aerospace Engineering, University of Notre Dame, April 2023.