Photothermal behavior for two-dimensional nanoparticle ensembles: optical coupling and thermal accumulation effects

Minggang Luo,¹,² Junming Zhao,¹,³, * Linhua Liu,⁴ and Mauro Antezza²,⁵, †

¹ School of Energy Science and Engineering, Harbin Institute of Technology, 92 West Street, Harbin 150001, China
² Laboratoire Charles Coulomb (L2C) UMR 5221 CNRS-Université de Montpellier, F- 34095 Montpellier, France
³ Key Laboratory of Aerospace Thermophysics, Ministry of Industry and Information Technology, Harbin 150001, China
⁴ School of Energy and Power Engineering, Shandong University, Qingdao 266237, China
⁵ Institut Universitaire de France, 1 rue Descartes, F-75231 Paris Cedex 05, France

(Dated: January 6, 2022)

Light-assisted micro-nanoscale temperature control in complex nanoparticle network attracts lots of research interests. Many efforts have been put on the optical properties of the nanoparticle networks and only a few investigations on its light-induced thermal behavior was reported. We consider two-dimensional (2D) square-lattice nanoparticle ensemble made of typical metal Ag with a radius of 5 nm. The effect of complex optical coupling and thermal accumulation on the light-induced thermal behavior in plasmonic resonance frequency (around 383 nm) is analyzed by means of the Green’s function approach. Regime borders of both optical coupling and thermal accumulation effects on the photothermal behavior of 2D square-lattice nanoparticle ensemble are figured out clearly and quantitatively. A dimensionless parameter ϕ is defined as the ratio of full temperature increase to that without considering the optical coupling or thermal accumulation to quantify the optical coupling and thermal accumulation effects on photothermal behavior. The more compact the nanoparticle ensemble is, the stronger the optical coupling on thermal behavior is. When the lattice spacing increases to tens of nanoparticle radius, the optical coupling becomes insignificant. When ϕ = 1 (lattice spacing increases to hundreds of nanoparticle radius), the thermal accumulation effects are weak and can be neglected safely. The polarization-dependent distribution of temperature increase of nanoparticles is observed only in the compact nanoparticle ensemble, while for dilute ensemble, such polarization-dependent temperature increase distribution can not be observed anymore. This work may help for the understanding of the light-induced thermal transport in the 2D particle ensemble.

I. INTRODUCTION

Nanoscale temperature control in complex plasmonic nanoparticle ensembles attracts a lot of interest in the fields of physics [1–4], chemistry [5–8], and biology [9–11], to name a few, where the light illumination is an efficient and common ingredient. Such light-assisted temperature control approach has many applications, ranging from hyperthermia therapy to additive manufacturing. Nanoparticles inside the ensemble often obtain energy from the incident light and then work as heat sources heating each other. Hence, the photothermal behavior for nanoparticle ensemble is a coupling optical-thermal process and should be analysed from both optical (light absorption and scattering) and thermal (heat dissipation) sides.

To well understand the light-induced thermal behavior of nanoparticle ensembles, it’s better for us to investigate the optical properties of the nanoparticle ensembles at first. Multiple scattering in nanoparticle ensembles will inevitably affect light absorption and finally affect the light-induced thermal behavior, which is named optical coupling effect. The coupled dipole method (CDM) is a well-known tool used for investigation on the properties of small particles [12, 13]. In CDM, the effect of mutual optical coupling in the nanoparticle ensemble on the light-absorption is considered by using the external electric field experienced by each nanoparticle rather than the direct incident field (i.e., illumination field). When nanoparticles are far enough away from each other, hence, they can be treated as optically independent. That is the external electric field experienced by nanoparticles can directly be approximated to the incident field. Some other methods (e.g., finite element method [14] and the finite difference time domain method [15], generalized multiparticle Mie method [16, 17] and boundary element method [18–20], to name a few) can also be applied to investigate optical properties of nanoparticle ensembles.

Then, with aforementioned methods in hand, the optical properties of nanoparticle ensembles have been investigated widely and have been reviewed in the literature[21–23]. Collective lattice resonance for disordered and quasi-random ensembles [24], and ordered arrays [25–29] of nanoparticles was analyzed. Effect of array structure on plasmonic resonance wavelength was analyzed theoretically for the silver nanoparticle ensembles [30], where extinction spectra shift was reported. Evlyukhin et al. [31] and Zundel et al. [32] systematically analyzed the finite-size effect on the optical response for periodic arrays of nanostructures (e.g., nanoparticles and graphene nanodisks). Besides above theoretical predictions, electromagnetic interactions in plasmonic nanoparticle ensembles were investigated and the spectra shift was demonstrated experimentally [33]. Aforementioned investigations on the optical response of nanoparticle ensembles provide the fundamental knowledge from the optical side to understand the photothermal behavior of nanoparticle ensembles.

* jmzhao@hit.edu.cn (Junming Zhao)
† mauro.antezza@umontpellier.fr (Mauro Antezza)
From the thermal side, light absorption by each nanoparticle in the ensemble will heat the whole nanoparticle ensemble cumulatively, namely accumulative (collective) heating effect, which was predicted theoretically [34–38] and demonstrated experimentally [39–42]. For small particle (point dipole approximation valid), a Green’s function approach combined the coupled dipole method (also namely discrete dipole approximation method) with the thermal Green’s function together for the light-induced thermal behavior modeling of nanoparticle ensembles was proposed by Baffou et al. (2010) [36], where the steady-state temperature distribution throughout arbitrary complex plasmonic systems can be calculated easily.

By means of the Green’s function approach, the effect of plasmonic (optical) coupling on the photothermal behavior of an ensemble with three-dimensional (3D) random distribution of nanoparticles was analyzed by Siahpoush et al. [43]. They found that the optical coupling can reduce the temperature increase of nanoparticles as compared to the case without optical coupling at the plasmonic resonance wavelength of single nanoparticle. Due to the thermal accumulation effect, when calculating the temperature increase of an arbitrary nanoparticle from a nanoparticle ensemble, we often can’t simply treat the nanoparticle solely, just like the rest nanoparticles don’t exist [40, 44]. Though the optical coupling and thermal accumulation on the photothermal behavior of nanoparticle ensembles already have been investigated, no clear and quantitative regime borders of these two effects have been reported yet. The method to figure out regime of these two effects will greatly facilitate investigation of the photothermal behavior of nanoparticle ensembles, which is the motivation of this work.

It should be noted that, due to the multiple scattering, the light may attenuate along its propagation direction in the 3D random nanoparticle ensemble, which can also reduce the temperature increase of nanoparticles. It is hard to tell the effect of light attenuation on temperature increase from the optical coupling on inhibition of the temperature. In this work, to have a clear understanding optical coupling effect on photothermal behavior without any interference of the light attenuation, we consider the two-dimensional nanoparticle ensembles, of which the extension direction is perpendicular to the light propagation direction. In addition, light absorption of nanoparticle ensemble is polarization-dependent [14, 22, 45]. The effect of light polarization on the photothermal behavior of ensemble composed of only a few or tens of nanoparticles has been analyzed already [14, 20, 46, 47]. As the number of nanoparticles in the ensemble increases, the mutual interaction between nanoparticles in the ensemble becomes more and more complex, of which it’s still remained unclear if the light polarization effect on the photothermal behavior still exists.

We address the aforementioned missing points in this paper, where photothermal behavior of two-dimensional nanoparticle ensembles is investigated by means of the Green’s function approach [36]. This work is organized as follows. In Sec. II, the discrete dipole approximation method for light scattering and the thermal Green’s function method for steady temperature spatial distribution are presented in brief, with considering the mutual optical coupling interaction and thermal accumulation effect. In addition, the physical model of the 2D nanoparticle ensemble considered in this work is also given. In Sec. III, a method to clearly and quantitatively figure out the regime borders of both the optical coupling and the thermal accumulation will be proposed as the focus of this work. Effects of both the optical coupling and the thermal accumulation on photothermal behavior, as well as the relation between the two effects, will be discussed.

**II. THEORETICAL MODELS**

In this section, we describe the physical system and the theoretical models: (1) the discrete dipole approximation method describing the light scattering and absorption of nanoparticle ensembles and (2) thermal Green’s function method describing the steady temperature spatial distribution for the nanoparticle ensembles. The SI unit system is used.

**A. Physical systems: 2D nanoparticle ensembles**

We investigate the thermal behavior of the 2D nanoparticle ensemble \(N \times N\) square-lattice ensemble illuminated by an incident light, as shown in Fig. 1. The size of the nanoparticle is supposed to be small compared to the wavelength of the incident light, which results in the validity of the point dipole approximation of the nanoparticle. The incident light wave vector is against the positive \(z\) axis direction and the polarization direction is parallel to the \(x\) axis. The ensemble is parallel to the \(xoy\) plane. The lattice spacing is \(L\). Nanoparticle radius is \(a\). When analyzing the relation between the optical coupling and collective effects on the light-induced thermal behavior of the ensemble, a chain of nanoparticles of interest near the \(x\) axis is extracted out of the 2D square-lattice nanoparticle ensemble in the green frame and is defined by \(y = L/2\) and \(z = 0\). When analyzing the thermal accumulation effect, the nanoparticle in the first sector closest to the origin is assigned as the central nanoparticle of the ensemble. The central nanoparticle position is defined as \(x = y = L/2\).

**B. Discrete dipole approximation method**

When the dipolar nanoparticle ensemble is illuminated by an incident light characterized by the electric field amplitude \(E^{\text{inc}}(r)\), the optical coupling between each nanoparticle results in an external field \(E^{\text{ext}}(r)\) experienced by each nanoparticle, which is quite different from the incident one and yields [13, 32]
where $k = \sqrt{\varepsilon_m \omega / c}$ is the wave vector in the host medium, $\varepsilon_m$ is the host medium relative permittivity, $\varepsilon_0$ is vacuum permittivity, $N_i = N \times N$ is the total number of nanoparticles in the 2D square-lattice nanoparticle ensemble, $G(r_i, r_j) = \frac{e^{ikr}}{4\pi r} \left[ 1 + \frac{ikr-1}{k^2r^2} \right] I_3 + \frac{3-3ikr-k^2r^2}{k^2r^2} \hat{r} \otimes \hat{r}$ is the electric Green’s function connecting two nanoparticles at $r_i$ and $r_j$, $r$ is the magnitude of the separation vector $r = r_i - r_j$, $\hat{r}$ is the unit vector $r/r$, $I_3$ is the $3 \times 3$ identity matrix, $p_j$ is the dipole moment located at $r_j$, which yields

$$p_j = \varepsilon_0 \varepsilon_m a_j |E^{ext}(r_j)|,$$  
where $a_j$ is the polarizability of the $j$th particle defined as follows.

$$a_j = \left( \frac{1}{a_j^0} - \frac{i k^3}{6 \pi} \right)^{-1},$$  
where $a_j^0$ is the Clausius-Mossotti polarizability defined as [23, 48]

$$a_j^0 = 4\pi a^3 \frac{\varepsilon(\omega) - \varepsilon_m}{\varepsilon(\omega) + 2\varepsilon_m},$$

where $a$ is the nanoparticle radius. $\varepsilon(\omega)$ is the relative permittivity of the nanoparticle. Then, the external field experienced by all nanoparticles in the ensemble can be rearranged in a compact and explicit way as follow:

$$E^{ext}(r_j) = E^{inc}(r_j) + \frac{k^2}{\varepsilon_0 \varepsilon_m} \sum_{j \neq i} G(r_i, r_j) \cdot p_j,$$  

where $3N \times 3N$ matrix is defined as follows.

$$A_k = \sum_{i=1}^{3N} G_i(r_i, r_j) Q_j,$$

where $G_{ij} = G(r_i, r_j)$, $\delta_{3N}$ is the $3 \times 3$ identity matrix.

### C. Thermal Green’s function method

In the following, it is supposed that the thermal conductivity of the nanoparticle is much higher than the one of the host medium. Under such approximation, the temperature can be considered as uniform inside nanoparticle [36]. For the considered metal Ag nanoparticle ensembles embedded in water, the thermal conductivity of metal particle (Ag, $\kappa_{\text{particle}} = 429 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$) is much higher than that of host medium (water, $\kappa = 0.6 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$). The temperature increase $\Delta T_i$ (relative to the ambient temperature) inside the $i$th nanoparticle of the ensemble induced by the incident light yields [36]

$$\Delta T_i = \sum_{j=1}^{N_i} G_i(r_i, r_j) Q_j,$$

where $Q_j = \frac{1}{2} \sigma_{abs} n c w |E^{inc}(r)|^2$, $\sigma_{abs} = k \Im(\alpha_j) - \frac{k^4}{6\pi} |\alpha_j|^2$ is the absorption cross section of the $j$th nanoparticle and $n = \sqrt{\varepsilon_m}$ is the refractive index of the host medium. The thermal Green’s function $G_i(r_i, r_j)$ is the scalar Green’s function $G(r_i, r_j)$ at $r_j$, which is associated to the following Poisson equation with a Dirac source distribution $\delta(r-r_j)$ in an infinite isotropic medium.

$$\nabla \cdot (-\kappa \nabla T(r)) = \sum_{j=1}^{N_i} Q_j \delta(r-r_j).$$

Such scalar Green’s function $G(r_i, r_j)$ yields

$$G(r_i, r_j) = \frac{1}{4\pi \kappa |r_i - r_j|},$$

where $\kappa$ is the thermal conductivity of the host medium. In particular, $G_i(r_i, r_j) = 1/(4\pi \kappa |r_i - r_j|)$ and $G_i(r_i, r_j) = 1/(4\pi \kappa a)$. It is worthwhile to mention that the thermal emission by the particles may also affect the temperature increase $\Delta T_i$. 

---

**FIG. 1.** Structure diagram of the two-dimensional $N \times N$ square-lattice nanoparticle ensemble. Light-induced thermal behavior of the ensemble is investigated. The lattice spacing is $L$. Nanoparticle radius is $a$. A chain of nanoparticles near the $x$ axis is extracted out of the 2D square-lattice nanoparticle ensemble in the green frame and is defined by $y = L/2$ and $z = 0$. 

\[ E^{ext}(r_j) = E^{inc}(r_j) + \frac{k^2}{\varepsilon_0 \varepsilon_m} \sum_{j \neq i} G(r_i, r_j) \cdot p_j, \]

where $k = \sqrt{\varepsilon_m \omega / c}$ is the wave vector in the host medium, $\varepsilon_m$ is the host medium relative permittivity, $\varepsilon_0$ is vacuum permittivity, $N_i = N \times N$ is the total number of nanoparticles in the 2D square-lattice nanoparticle ensemble, $G(r_i, r_j) = e^{ikr} \left[ 1 + \frac{ikr-1}{k^2r^2} \right] I_3 + \frac{3-3ikr-k^2r^2}{k^2r^2} \hat{r} \otimes \hat{r}$ is the electric Green’s function connecting two nanoparticles at $r_i$ and $r_j$, $r$ is the magnitude of the separation vector $r = r_i - r_j$, $\hat{r}$ is the unit vector $r/r$, $I_3$ is the $3 \times 3$ identity matrix, $p_j$ is the dipole moment located at $r_j$, which yields

$$p_j = \varepsilon_0 \varepsilon_m a_j E^{ext}(r_j), \quad a_j = \left( \frac{1}{a_j^0} - \frac{i k^3}{6 \pi} \right)^{-1}, \quad a_j^0 = 4\pi a^3 \frac{\varepsilon(\omega) - \varepsilon_m}{\varepsilon(\omega) + 2\varepsilon_m},$$

where $a$ is the nanoparticle radius. $\varepsilon(\omega)$ is the relative permittivity of the nanoparticle. Then, the external field experienced by all nanoparticles in the ensemble can be rearranged in a compact and explicit way as follow:
The net emitted power from the particle $j$ to the thermal bath can be defined as $\mathcal{P}_{j \to B} = \sigma_{\text{abs}}\sigma_B(T_j^4 - T_{\text{env}}^4)$ [49, 50], where $\sigma_B$ is Stefan-Boltzmann constant, $T_{\text{env}}$ is the environment temperature, $T_j$ is the temperature of the particle $j$. For the considered geometry where the power absorbed from the incident light is much more significant than the power exchanged with the environment, $\mathcal{P}_{j \to B}$ can be neglected with respect to $Q_i$ in Eq. (7).

Another important factor that may significantly influence the temperature increase of nanoparticles is the thermal boundary resistance around the particles, which is well acknowledged to play a key role in nanoparticle-based experiments. The molecular coating acts as a thermal boundary resistance, thus molecular coating on the nanoparticle surface can influence the heat transfer between the inner plasmonic nanoparticle and the outer surrounding host medium [51]. It has been demonstrated that this thermal boundary resistance effect indeed can affect the temperature inside nanoparticle, however such thermal boundary resistance would not change the temperature distribution in the host medium [36].

III. RESULTS AND DISCUSSION

The photothermal behavior of two-dimensional nanoparticle ensembles are analyzed with particular focus on the optical coupling and thermal accumulation effects. Nanoparticle radius $(a)$ is 5 nm. Nanoparticles are composed of metal Ag, of which the dielectric function is from Ref. [52]. Incident light wavelength is fixed at 383 nm, which is corresponding to the plasmonic resonance of Ag nanoparticle embedded in water with the relative permittivity of $\epsilon_R = 1.77$ and thermal conductivity of $\kappa = 0.6 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$. Though we only focus on the optical coupling effect at resonance frequency in this work, it is noted that effect of optical coupling on the photothermal behavior is wavelength-dependent. We discussed the optical coupling effect on photothermal behavior by considering only water as a host medium in this work. As indicated by our previous work [53], the relative permittivity of the host medium significantly affects the nanoparticle polarizability, which is strongly relevant to the temperature increase of nanoparticles determined by Eq. (7) combined with $\sigma_{\text{abs}} = k \text{Im}(a_j) - \frac{k^2}{4\pi} |a_j|^2$ and $Q_i = \frac{1}{2} \sigma_{\text{abs}} n c e_0 |\mathbf{E}^{\text{ext}}(\mathbf{r})|^2$. Thus, by tailoring host medium relative permittivity, one may control the photothermal behavior of nanoparticles consequently. The incident light intensity $I_0$ is 0.5 mW·µm$^{-2}$. The lattice spacing $L$ is at least three times larger than the particle radius, which makes the dipole approximation valid [54–56]. The dimensionless parameters $X$ and $Y$ are defined as $x/L(N-1)$ and $y/L(N-1)$, respectively.

A. Relation between the optical coupling and thermal accumulation effects

The light-induced thermal behavior of the nanoparticle ensemble is affected by two main factors, i.e., (a) the optical coupling effect [43] and (b) thermal accumulation effect [2, 36]. We clarify the two factors in brief at first and then discuss the relation between them.

Optical coupling effect The optical coupling effect concerns the light scattering and absorption of the nanoparticles. As mentioned in Sec. II B, the external field $\mathbf{E}^{\text{ext}}(\mathbf{r}_j)$ experienced by the $i$th nanoparticle at $\mathbf{r}_i$ can be obtained by Eq. (1), where the second term on the right hand is corresponding to the optical coupling. When the optical coupling is negligible, the external field $\mathbf{E}^{\text{ext}}(\mathbf{r}_j)$ is directly equal to the illuminating incident field $\mathbf{E}^{\text{inc}}(\mathbf{r}_i)$ by neglecting the second term at the right hand in Eq. (1).

Thermal accumulation effect The thermal accumulation effect concerns the thermal diffusion process in the nanoparticle ensemble. As mentioned in Sec. II C, the temperature increase $\Delta T$ experienced by the $i$th nanoparticle calculated by Eq. (7) has two contributions: (a) its own heat generation $\Delta T_S$ and (b) the heat generated by the rest other $N_i - 1$ nanoparticles $\Delta T^{\text{ext}}$ under illumination in the ensemble, which yields

$$\Delta T = \Delta T_S + \Delta T^{\text{ext}},$$

where $\Delta T_S = G_i(\mathbf{r}_i, \mathbf{r}_j)Q_i$ is the temperature increase of the $i$th nanoparticle due to its own heat generation, $\Delta T^{\text{ext}} = \sum_{j \neq i} G_i(\mathbf{r}_i, \mathbf{r}_j)Q_j$ is the temperature increase of the $i$th nanoparticle due to the heat generated by the rest other $N_i - 1$ nanoparticles.

The thermal accumulation effect is corresponding to the second term on the right hand of Eq. (10). When the thermal accumulation effect is negligible, the second term at the right hand in Eq. (10) is negligible. Each nanoparticle in the nanoparticle ensemble can be treated as an isolated hotspot.

To quantitatively analyze the optical coupling and the thermal accumulation effect, a parameter $\varphi$ is define as follow:

$$\varphi = \frac{\Delta T}{\Delta T_{\varphi = 0} \text{ or } S},$$

where $\Delta T_{\varphi = 0} \text{ or } S$ are corresponding to the temperature increase without the optical coupling effect and the temperature increase due to only its own heat generation, respectively. We show the dependence of $\varphi$ for both optical coupling effect and thermal accumulation effect on two parameters 1) the dimensionless lattice spacing $L/a$ and 2) the dimensionless $X$ in Fig. 2. We considered $14 \times 14$ square-lattice nanoparticle ensembles. The blue-dash line corresponding to the dimensionless parameter defined in Ref.[2] $\xi = L/(3a \sqrt{N}) = 1$ is added for reference. $\zeta >> 1$ is for the negligible thermal accumulation effect. $\zeta << 1$ is for the strong thermal accumulation effect.
The optical coupling effect can compete with the thermal accumulation effect for the dense nanoparticle ensembles and can also cooperate with the thermal accumulation effect to enhance $\Delta T$. For the loose enough ensembles, both the optical coupling effect and the thermal accumulation effect are negligible and the plasmonic nanoparticle can be treated as the isolated hotspot safely.

B. Optical coupling effect on photothermal behavior

In this section, the optical plasmonic coupling effects on the photothermal behavior of the 2D finite-size square-lattice nanoparticle ensemble are analyzed. It’s worthwhile to mention that the optical plasmonic coupling inhibits the temperature increase of the 3D randomly distributed nanoparticle ensemble [43].

The temperature increase distribution for 2D ensemble with several different lattice spacings is shown in Fig. 3 (L = 3a, 10a and 110a, respectively): (a)-(c) the temperature increase distribution with optical coupling effect, (d)-(f) the temperature increase distribution without optical coupling effect. Nanoparticle number for each lateral edge $N = 40$.

For dense ensembles ($L = 3a$ and 10a), the temperature increase with optical coupling as shown in Fig. 3(a) and (b) is much less than that without the optical coupling as shown in Fig. 3(d) and (e), respectively. For the dilute ensemble ($L = 110a$), the temperature increase distribution with optical coupling as shown in Fig. 3(c) is nearly the same as...
that without the optical coupling as shown in Fig. 3(f). The strong optical coupling significantly inhibits the temperature increase of the nanoparticles in the 2D square-lattice ensemble. As the lattice spacing increasing, the decreasing optical coupling accounts for the negligible inhibition on the temperature increase.

We extract a chain of nanoparticles of interest near the x axis out of the 2D square-lattice nanoparticle ensemble, as shown in Fig. 1. The chain is defined by $y = L/2$ and $z = 0$. The temperature increase along the nanoparticle chains of interest with two different lattice spacings ($L = 3a$ and 110$a$) is shown in Fig. 4. The dimensionless and the real separation distance between the neighboring peaks shown in Fig. 4 are $\Delta X \approx 0.02596$ and $\Delta x \approx L$ for the considered two ensembles, which is corresponding to the $\Delta x \approx L$.

As shown in Fig. 3 (a), (b) and (c), it is also noted that the temperature increase for the dense ensemble ($L = 3a$) is angle-dependent (seen from the ensemble center), which is quite different from the angle-independent temperature increase for dilute ensembles ($L = 10a$ and 110$a$). For the dilute nanoparticle ensembles, the optical coupling becomes less important and thus each particle works as a separate heat source for temperature increase, which results in the angle-independent temperature distribution. The optical coupling in the dense particle ensemble is expected to be strong and polarization-dependent, which may account for the angle-dependent temperature distribution. Hence, to some extend, we can tailor the temperature increase distribution by light polarization. However, it’s worthwhile to mention that the light-induced heat transfer behavior in the nanostructures (e.g., dense particle ensembles) is not always polarization-dependent. Recently, a polarization-independent isosbestic temperature increase behavior of nanostructures was proposed [45, 47].

C. Thermal accumulation effect

The dependence of $\Delta T$ of the central particle from the 2D square-lattice ensemble on the nanoparticle number $N$ in each lateral edge is shown in Fig. 5: (a) with optical coupling effect and (b) without optical coupling effect. Four different lattice spacings are considered, $L = 3a$, 10$a$, 30$a$ and 3000$a$, respectively. The temperature increase $\Delta T$ from the center to the boundary of the ensemble along the x axis.

As shown in Fig. 3 (a), (b) and (c), it is also noted that the temperature increase for the dense ensemble ($L = 3a$) is angle-dependent (seen from the ensemble center), which is quite different from the angle-independent temperature increase for dilute ensembles ($L = 10a$ and 110$a$). For the dilute nanoparticle ensembles, the optical coupling becomes less important and thus each particle works as a separate heat source for temperature increase, which results in the angle-independent temperature distribution. The optical coupling in the dense particle ensemble is expected to be strong and polarization-dependent, which may account for the angle-dependent temperature distribution. Hence, to some extend, we can tailor the temperature increase distribution by light polarization. However, it’s worthwhile to mention that the light-induced heat transfer behavior in the nanostructures (e.g., dense particle ensembles) is not always polarization-dependent. Recently, a polarization-independent isosbestic temperature increase behavior of nanostructures was proposed [45, 47].
(a) with optical coupling

(b) without optical coupling

FIG. 5. Dependence of $\Delta T$ of the central particle on the nanoparticle number $N$ in each lateral edge: (a) with optical coupling effect and (b) without optical coupling effect. Lattice spacing $L = 3a$, $10a$, $30a$ and $3000a$. Metal Ag is used. The temperature increase $\Delta T = 9.8$ K for the isolated single nanoparticle is also added for reference.

creases nonlinearly with $N$. While for the loose ensemble ($L = 30a$), a linear dependence of $\Delta T$ on $N$ can be observed, which is consistent with the reported results in Ref.[36]. For the compact ensemble (e.g., $L = 3a$), the temperature increase $\Delta T$ for the central nanoparticle is even less than that of the isolated single nanoparticle. For all the considered ensembles with different lattice spacings, the temperature increase $\Delta T$ without optical coupling effect increases linearly with $N$, as shown in Fig. 5(b). The strong optical coupling for the compact ensembles accounts for the non-linear dependence of the $\Delta T$ on $N$, observed in Fig. 5(a).

For the extremely loose ensemble ($L = 3000a$), the optical coupling and thermal accumulation effects are negligible, which accounts for the same temperature increase $\Delta T$ for the central nanoparticle as that of the isolated single nanoparticle.

From Fig. 5(a) and (b), in general, for an ensemble composed of a certain number of nanoparticles, $\Delta T$ without the optical coupling effect increases monotonically with the lattice spacing, while $\Delta T$ with the optical coupling effect increases at first and then decreases with the lattice spacing. The dependence of $\Delta T$ for the central particle on the lattice spacing $L/a$ is shown in Fig. 6. Ensembles of three different sizes are considered, $N = 2$, 10 and 20, respectively. Both the $\Delta T$ with and without optical coupling effects are considered. From $L = L_{OC}$, $\Delta T$ with and without optical coupling effect starts to be identical, where the optical coupling is negligible. From $L = L_C$, $\Delta T$ for ensembles composed of different number of nanoparticles starts to be identical, where the thermal accumulation effect is negligible. The length $L_C$, where the thermal accumulation effect is negligible is much larger than the length $L_{OC}$ where optical coupling starts to be less important. When the thermal accumulation effect is negligible, the optical coupling also cannot be observed. However, when the optical coupling effect is negligible, the thermal accumulation effect on photothermal behavior can exist if the lattice spacing $L$ is not large enough.

FIG. 6. Dependence of $\Delta T$ for the central particle on the lattice spacing $L/a$. Different ensemble size are considered, $N = 2$, 10 and 20. Both the $\Delta T$ with and without optical coupling effects are considered. From $L = L_{OC}$, $\Delta T$ with and without optical coupling effect starts to be identical. From $L = L_C$, $\Delta T$ for ensembles composed of different number of nanoparticles starts to be identical.

When considering no optical coupling effect, the temperature increase $\Delta T$ for the central nanoparticle decreases monotonically with $L/a$. The pure thermal accumulation effect accounts for the monotonic dependence of $\Delta T$ on
When considering the optical coupling effect, $\Delta T$ increases at first and then decreases with $L/a$, as shown with the symbol lines in Fig. 6. The thermal accumulation effect together with the optical coupling effect account for the non-monotonic dependence of $\Delta T$ on $L/a$.

We also can notice that small oscillations of $\Delta T$ occur at around few tens of $L/a$ in Fig. 6. Note that $a = 5\text{nm}$, so the lattice spacings $L$ corresponding to small oscillations are comparable to or even larger than the considered wavelength (383 nm). According to Zou and Schatz (2004) [57], for a fixed wavelength, extinction efficiency of two-dimensional hexagonal arrays is not monotonously dependent on the lattice spacing, as shown in FIG. 3 of Ref. [57]. Such non-monotonously-dependent optical properties may account for the small oscillations of $\Delta T$ when increasing $L/a$ for two-dimensional nanoparticle arrays.

IV. CONCLUSION

Light-induced heat transfer of the two-dimensional nanoparticle ensembles embedded in water is investigated by means of the Green's function approach with focus on proposing a clear regime map of both the optical coupling and the thermal accumulation. The dimensionless parameter $\varphi$ is defined to quantify the optical coupling and collective effects on photothermal behavior. For 2D ordered nanoparticle ensemble, similar to the 3D random nanoparticle ensemble, optical coupling can also inhibit temperature increase $\Delta T$ of nanoparticles. The more compact the nanoparticle ensemble is, the stronger the optical coupling on thermal behavior is. When the lattice spacing increases to tens of nanoparticle radius, the optical coupling becomes insignificant. In addition, when $\varphi \approx 1$ and lattice spacing increases to hundreds of nanoparticle radius, the thermal accumulation effects are weak and can be neglected safely. The distribution of temperature increase of nanoparticle is polarization-dependent, especially for the compact nanoparticle ensemble. For dilute ensemble, such polarization-dependent temperature increase distribution can not be observed. The temperature increase $\Delta T$ of the center of ensemble increases linearly with increasing the ensemble size without considering optical coupling. However, when considering optical coupling, for the compact ensemble, $\Delta T$ increases non-linearly with increasing the ensemble size. For a fixed ensemble size, $\Delta T$ increases non-linearly with increasing the lattice spacing. This work may help for the understanding of the light-induced thermal transport in the 2D particle ensemble.

ACKNOWLEDGMENTS

The support of this work by the National Natural Science Foundation of China (No. 51976045) is gratefully acknowledged. M. G. Luo also thanks for support from the China Postdoctoral Science Foundation (2021M700991). In addition, we acknowledge greatly the kind and helpful suggestions from the two anonymous reviewers.

[1] G. Baffou, F. Cichos, and R. Quindt, Applications and challenges of thermoplasmonics, Nature Materials 19, 946 (2020).
[2] G. Baffou, I. Bordacchini, A. Baldi, and R. Quindt, Simple experimental procedures to distinguish photothermal from hot-carrier processes in plasmonics, Light Sci. Appl. 9, 108 (2020).
[3] O. Blum and N. T. Shaked, Prediction of photothermal phase signatures from arbitrary plasmonic nanoparticles and experimental verification, Light Sci. Appl. 4, e322 (2015).
[4] S. A. Maier, M. L. Brongersma, P. G. Kik, S. Meltzer, A. A. G. Requicha, and H. A. Atwater, Plasmonics—a route to nanoscale optical devices, Adv. Mater. 13, 1501 (2001).
[5] X. L. Li, X. Zhang, H. O. Everitt, and J. Liu, Light-induced thermal gradients in ruthenium catalysts significantly enhance ammonia production, Nano Lett. 19, 1706 (2019).
[6] L. Zhou, D. F. Swearer, C. Zhang, H. Robatjazi, H. Zhao, L. Henderson, L. Dong, P. Christopher, E. A. Carter, P. Nordlander, and N. J. Halas, Quantifying hot carrier and thermal contributions in plasmonic photocatalysis, Science 362, 69 (2018).
[7] K. Saha, S. S. Agasti, C. Kim, X. Li, and V. M. Rotello, Gold nanoparticles in chemical and biological sensing, Chem. Rev. 112, 2739 (2012).
[8] P. V. Kamat, Photophysical, photochemical and photocatalytic aspects of metal nanoparticles, J. Phys. Chem. B 106, 7729 (2002).
[9] M. B. Cortie, D. L. Cortie, and V. Timchenko, Heat transfer from nanoparticles for targeted destruction of infectious organisms, Int. J. Hyperthermia 34, 157 (2018).
[10] H. Šípová, L. Shao, N. Odebo Länk, D. Andrén, and M. Käll, Photothermal DNA release from laser-tweezed individual gold nanomotors driven by photon angular momentum, ACS Photonics 5, 2168 (2018).
[11] S. Jones, D. Andrén, P. Karpinski, and M. Käll, Photothermal heating of plasmonic nanoantennas: Influence on trapped particle dynamics and colloid distribution, ACS Photonics 5, 2878 (2018).
[12] O. Merchiers, F. Moreno, F. González, and J. M. Saiz, Light scattering by an ensemble of interacting dipolar particles with both electric and magnetic polarizabilities, Phys. Rev. A 76, 043834 (2007).
[13] G. W. Mulholland, C. F. Bohren, and K. A. Fuller, Light scattering by agglomerates: Coupled electric and magnetic dipole method, Langmuir 10, 2533 (1994).
[14] R. Borah and S. W. Verbruggen, Coupled plasmon modes in 2D gold nanoparticle clusters and their effect on local temperature control, J. Phys. Chem. C 123, 30594 (2019).
[15] A. E. Ershov, V. S. Gerasimov, R. G. Bikbaev, S. P. Polyutov, and S. V. Karpov, Mode coupling in arrays of Al nanoparticles, J. Quant. Spectrosc. Radiat. Transf. 248, 106961 (2020).
[16] Y. L. Xu and N. G. Khlebtsov, Orientation-averaged radiative properties of an arbitrary configuration of scatterers, J. Quant. Spectrosc. Radiat. Transf. 79-80, 1121 (2003).
[17] B. Khlebtsov, V. Zharov, A. Melnikov, V. Tuchin, and N. Khlebtsov, Optical amplification of photothermal therapy
with gold nanoparticles and nanoclusters, Nanotechnology 17, 5167 (2006).

[18] F. J. García De Abajo and A. Howie, Relativistic electron energy loss and electron-induced photon emission in inhomogeneous dielectrics, Phys. Rev. Lett. 80, 5180 (1998).

[19] F. J. García de Abajo and A. Howie, Retarded field calculation of electron energy loss in inhomogeneous dielectrics, Phys. Rev. B 65, 115418 (2002).

[20] G. Baffou, R. Quidant, and F. J. García de Abajo, Nanoscale control of optical heating in complex plasmonic systems, ACS Nano 4, 709 (2010).

[21] F. J. García De Abajo, Colloquium: Light scattering by particle and hole arrays, Rev. Mod. Phys. 79, 1267 (2007).

[22] M. B. Ross, C. A. Mirkin, and G. C. Schatz, Optical properties of one-, two-, and three-dimensional arrays of plasmonic nanostructures, J. Phys. Chem. C 120, 816 (2016).

[23] V. G. Kravets, A. V. Kabashin, W. L. Barnes, and A. N. Grigorenko, Plasmonic surface lattice resonances: A review of properties and applications, Chem. Rev. 118, 5912 (2018).

[24] V. I. Zakomirnyi, S. V. Karpov, H. Ågren, and I. L. Rasskazov, Collective lattice resonances in disordered and quasi-random all-dielectric metasurfaces, J. Opt. Soc. Am. B 36, E21 (2019).

[25] A. D. Utyushev, V. I. Zakomirnyi, A. E. Ershov, V. S. Gerasimov, S. V. Karpov, and I. L. Rasskazov, Collective lattice resonances in all-dielectric nanostructures under oblique incidence, Photonics 7, 24 (2020).

[26] A. Manjavacas, L. Zundel, and S. Sanders, Analysis of the limits of the near-field produced by nanoparticle arrays, ACS Nano 13, 10682 (2019).

[27] V. I. Zakomirnyi, A. E. Ershov, V. S. Gerasimov, S. V. Karpov, H. Ågren, and I. L. Rasskazov, Collective lattice resonances in arrays of dielectric nanoparticles: a matter of size, Opt. Lett. 44, 5743 (2019).

[28] M. Ramezani, G. Lozano, M. A. Verschueren, and J. Gómez-Rivas, Modified emission of extended light emitting layers by selective coupling to collective lattice resonances, Phys. Rev. B 94, 125406 (2016).

[29] S. Rodríguez, M. Schaaftsma, A. Berrier, and J. G. Rivas, Collective resonances in plasmonic crystals: Size matters, Physica B 407, 4081 (2012).

[30] L. L. Zhao, K. L. Kelly, and G. C. Schatz, The extinction spectra of silver nanoparticle arrays: Influence of array structure on plasmon resonance wavelength and width, J. Phys. Chem. B 107, 7343 (2003).

[31] A. B. Evlyukhin, C. Reinhardt, A. Seidel, B. S. Luk’yanchuk, and B. N. Chichkov, Optical response features of Si-nanoparticle arrays, Phys. Rev. B 82, 045404 (2010).

[32] L. Zundel and A. Manjavacas, Finite-size effects on periodic arrays of nanostructures, J. Phys. Photonics 1, 015004 (2018).

[33] A. Bouhelier, R. Bachelot, J. S. Im, G. P. Wiederrecht, G. Lerondel, S. Kostcheev, and P. Royer, Electromagnetic interactions in plasmonic nanoparticle arrays, J. Phys. Chem. B 109, 3195 (2005).

[34] G. Baffou and R. Quidant, Thermo-plasmonics: using metallic nanostructures as nano-sources of heat, Laser & Photon. Rev. 7, 171 (2013).

[35] L.-W. Un and Y. Sivan, Size-dependence of the photothermal response of a single metal nanopshere, J. Appl. Phys. 126, 173103 (2019).

[36] G. Baffou, R. Quidant, and C. Girard, Thermoplasmonics modeling: A Green’s function approach, Phys. Rev. B 82, 165424 (2010).

[37] R. Gillibert, E. Colas, M. L. de La Chapelle, and P. G. Guicciardi, Heat dissipation of metal nanoparticles in the dipole approximation, Plasmonics 15, 1001–1005 (2020).

[38] A. Heber, M. Selmke, and F. Cichos, Metal nanoparticle based all-optical photothermal light modulator, ACS Nano 8, 1893 (2014).

[39] H. H. Richardson, M. T. Carlson, P. J. Tandler, P. Hernandez, and A. O. Govorov, Experimental and theoretical studies of light-to-heat conversion and collective heating effects in metal nanoparticle solutions, Nano Lett. 9, 1139 (2009).

[40] G. Baffou, P. Berto, E. Bermúdez Ureña, R. Quidant, S. Monneret, J. Polleux, and H. Rigneault, Photoinduced heating of nanoparticle arrays, ACS Nano 7, 6478 (2013).

[41] G. Baffou, E. Ureña, P. Berto, S. Monneret, R. Quidant, and H. Rigneault, Deterministic temperature shaping using plasmonic nanoparticle assemblies, Nanoscale 6, 8984 (2014).

[42] C. Moularas, Y. Georgiou, K. Adamska, and Y. Deligianakis, Thermoplasmonic heat generation efficiency by non-monodisperse core-shell AgO@SiO2 nanoparticle ensemble, J. Phys. Chem. C 123, 22499 (2019).

[43] V. Siahpoush, S. Ahmadi-kandjani, and A. Nikniazi, Effect of plasmonic coupling on photothermal behavior of random nanoparticles, Opt. Commun. 420, 52 (2018).

[44] A. O. Govorov, W. Zhang, T. Skeini, H. Richardson, J. Lee, and N. A. Kotov, Gold nanoparticle ensembles as heaters and actuators: melting and collective plasmon resonances, Nanoscale Res. Lett. 1, 84 (2006).

[45] L. X. Ma and C. C. Wang, Isosbestic light absorption by metallic dimers: effect of interparticle electromagnetic coupling, Appl. Opt. 59, 1028 (2020).

[46] Y. Ren, Q. Chen, H. Qi, L. Ruan, and J. Dai, Phase transition induced by localized surface plasmon resonance of nanoparticle assemblies, Int. J. Heat Mass Transf. 127, 244 (2018).

[47] K. Metwally, S. Mensah, and G. Baffou, Isosbestic thermoplasmonic nanostructures, ACS Photonics 4, 1544 (2017).

[48] P. O. Chapuis, M. Larroche, S. Volz, and J.-J. Greffet, Radiative heat transfer between metallic nanoparticles, Appl. Phys. Lett. 92, 3303 (2008).

[49] P. Ben-Abdallah, R. Messina, S.-A. Biets, M. Tschikin, K. Joullain, and C. Henkel, Heat superdiffusion in plasmonic nanostructure networks, Phys. Rev. Lett. 111, 174301 (2013).

[50] V. Yannopapas and N. V. Vitanov, Spatiotemporal control of temperature in nanostructures heated by coherent laser fields, Phys. Rev. Lett. 110, 044302 (2013).

[51] J. Alper and K. Hamad-Schifferli, Effect of ligands on thermal dissipation from gold nanorods, Nano Lett. 9, 1893 (2009).

[52] E. Palik, Handbook of Optical Constants of Solids (Academic, New York, 1998).

[53] M. G. Luo, J. M. Zhao, L. H. Liu, B. Guizal, and M. Antezza, Many-body effective thermal conductivity in phase-change nanoparticle chains due to near-field radiative heat transfer, Int. J. Heat Mass Transf. 166, 120793 (2021).

[54] P Ben-Abdallah, S.-A. Biets, and K. Joullain, Many-body radiative heat transfer theory, Phys. Rev. Lett. 107, 114301 (2011).

[55] J. Dong, J. M. Zhao, and L. H. Liu, Radiative heat transfer in many-body systems: Coupled electric and magnetic dipole approach, Phys. Rev. B 95, 125411 (2017).

[56] P Ben-Abdallah, Multitip near-field scanning thermal microscopy, Phys. Rev. Lett. 123, 264301 (2019).

[57] S. L. Zou and G. C. Schatz, Narrow plasmonic/photonic extinction and scattering line shapes for one and two dimensional silver nanoparticle arrays, J. Chem. Phys. 121, 12606 (2004).