A normal-diffusion theory for near-field radiative heat transfer in dense nanoparticle systems

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ABSTRACT

We derived a normal-diffusion type governing equation for near-field radiative heat transfer (NFRHT) in dense nanoparticle systems at continuum-scale directly from fluctuational electrodynamics formulation at particle-scale. The near-field radiative thermal conductivity (NFRTC) tensor is discovered naturally as the coefficient of diffusion term, which results in a new formula for calculating NFRTC. A first-order term is shown to appear in the governing equation, which reveals a convection-like transfer behavior of NFRHT in highly asymmetrical systems of particles. For systems of uniformly distributed particles, the governing equation reduces to the classical heat diffusion equation. This work rigorously builds a bridge between particle-scale and continuum-scale NFRHT in particulate system, which paves a way for understanding and analysis of heat diffusion in large-scale particulate system with near-field interaction.

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Radiative heat exchange at subwavelength (thermal wavelength $\lambda_r = h c / k_B T$) separation distance can exceed the limit of two blackbodies due to the tunneling of evanescent waves. This super-Planckian transfer behavior contains rich physics and promising application potential, has attracted considerable research interests recently [1-6]. The fluctuational electrodynamics theory was widely accepted as a rigorous theoretical framework for NFRHT analysis [7-15], and has been proved by several recent experimental investigations [16-18]. In dense nanoparticle system where nanoparticles lie in the near field of each other, multiple scattering of thermally excited evanescent wave will dominate NFRHT. The complex many-body interaction significantly influences NFRHT characteristics of dense particulate system [19, 20]. Several theoretical approaches capable of analyzing NFRHT in system composed of many particles have been developed, e.g., the many-body radiative heat transfer theory [21-23], scattering matrix method [24-28], the trace formulas [28-30] and the quasi-analytic solution [31, 32]. Even though, due to enormous computational effort, it's usually technically impractical to directly apply the existed theoretical frameworks suitable for particle-scale to large system composed of a great number of particles.

In practice, continuum approach at macro-scale is an essential way to deal with NFRHT in large-scale particulate system. Ben-Abdallah et al. [3] developed a continuum theory for NFRHT in particulate system for the first time, in form of fractional-order diffusion equation (FODE), based on fluctuational electrodynamics. An anomalous diffusion behavior (heat super-diffusion) was observed in plasmonic nanostructure networks driven by collective near-field interaction. It is noted that numerical solution of fractional-order partial differential equation is cumbersome since traditional numerical method cannot be applied directly. Kinetic theory based on Boltzmann transport equation (BTE) for thermal photons is another continuum approach to deal with NFRHT in dense particulate system [33-37]. The limitations of the kinetic theory were analyzed.
systematically recently by comparing to fluctuational electrodynamics results \cite{36, 37}. It was shown that the mismatch between the resonant mode and the thermal accessible mode significantly limits the application of the kinetic theory. Assuming validity of the Fourier’s law for NFRHT in particulate system, the classical heat diffusion equation (HDE) can be considered as another way of continuum approach, and the key is to calculate the effective radiative thermal conductivity from particle-scale analysis. Tervo \textit{et al.} \cite{38} proposed a method to calculate the NFRTC for arbitrary three-dimensional particulate systems. However, the applicability of the HDE for NFRHT in particulate system has not been examined thoroughly, especially for system with non-uniform distribution of particles.

Though a number of works have applied the BTE and HDE to study NFRHT in dense particulate system \cite{33-38}, both of the equations have not been rigorously derived from the first principle, i.e., fluctuational electrodynamics, and their rationality are not known clearly. It is still an open problem whether it is possible to directly derive a normal-diffusion type (or HDE type) governing equation, being alternative to the FODE \cite{3}, for NFRHT in the dense particulate system from the exact fluctuational electrodynamics. Since the normal-diffusion type equation can be solved as easily as the HDE, it will largely facilitate practical applications.

In this letter, a normal-diffusion type governing equation for the NFRHT in arbitrary dense particulate system is derived rigorously starting from the exact fluctuational electrodynamics formulation at particle-scale. Both uniform and non-uniform distribution of particles are considered. A convection-like first-order term is discovered for the first time to appears in the governing equation for system with asymmetric distribution of particles, which reveals a convection-like transfer behavior of NFRHT in highly asymmetrical systems of particles. The NFRTC tensor is obtained naturally as the coefficient of the diffusion term during the derivation.
FIG. 1 Schematic of NFRHT in particulate system analyzed at particle-scale (left) and continuum-scale (right). The continuum-scale governing equations for NFRHT are derived directly from the discrete form of energy balance equation at particle-scale based on the fluctuational electrodynamics formulation.

To start the analysis, a particulate system with near-field radiative heat transfer is considered, which is depicted in Fig. 1, and our goal is to derive a governing equation for NFRHT at continuum-scale, as shown in Fig. 1. The energy balance of the central particle $P$ can be calculated at the particle-scale as [3]

$$\sum_{i=1}^{N_p} Q_{i\rightarrow P} + S_P = \frac{dE_P}{dt},$$

(1)

where the subscript $i$ denotes the $i$-th neighboring particle at $r_i$ of the central particle $P$ at $r_P$, $N_p$ is the number of neighboring particles, $Q_{i\rightarrow P}$ [W] is the net exchanged radiative power between $i$ and $P$, $S_p$ [W] is the energy received from the thermal bath and external source, and $E_p = C_p T$ [J] is the internal energy of the particle, where $C_p$ [J/K] is the heat capacity. To facilitate the calculation of NFRHT between particles, thermal conductance ($U$) between particle $i$ and $P$ is defined as follows.
where \( l_i = |\mathbf{r}_i - \mathbf{r}_p| \) is the separation distance between the neighboring particle \( i \) and the central particle \( P \), \( T_i \) and \( T_P \) are the temperatures of particle \( i \) and particle \( P \), respectively, and \( \phi(l_i, T) = 3\int_0^{+\infty} \frac{d\omega}{2\pi} T_{iP}(\omega) \Theta(\omega, T) \) is the transferred radiative power by particle \( i \) at \( T \) to particle \( P \), where \( \Theta(\omega, T) = h\omega \left[ \exp\left( \frac{h\omega}{k_B T} \right) - 1 \right] \) is the mean energy of Planck oscillator, and \( T_{iP}(\omega) \) is the near-field transmission coefficient between particle \( i \) and particle \( P \). Considering both the contributions of electric and magnetic polarizations of particles, \( T_{iP}(\omega) \) can be calculated from \([19, 22]\)

\[
T_{iP}(\omega) = \frac{4}{3} k^4 \left[ \text{Im} \left( \chi'_E \right) \text{Im} \left( \chi'_M \right) \text{Tr} \left( G_{iP}^{EE} G_{iP}^{EE} \right) + \text{Im} \left( \chi'_E \right) \text{Im} \left( \chi'_M \right) \text{Tr} \left( G_{iP}^{EM} G_{iP}^{EM} \right) + \text{Im} \left( \chi'_H \right) \text{Im} \left( \chi'_M \right) \text{Tr} \left( G_{iP}^{ME} G_{iP}^{ME} \right) + \text{Im} \left( \chi'_E \right) \text{Im} \left( \chi'_H \right) \text{Tr} \left( G_{iP}^{MM} G_{iP}^{MM} \right) \right],
\]

where \( k \) is wave vector, \( \chi'_E = \alpha_E - \frac{ik^3}{6\pi} |\alpha_E|^2 \) and \( \chi'_H = \alpha_H - \frac{ik^3}{6\pi} |\alpha_H|^2 \), \( \alpha_E \) and \( \alpha_H \) are the electric and magnetic polarizability and are obtained from the extinction cross section with the help of the first order Lorenz-Mie coefficients \([39, 40]\), \( G_{iP}^{EE} \), \( G_{iP}^{ME} \), \( G_{iP}^{EM} \) and \( G_{iP}^{MM} \) are the Green’s functions in many particles system.

Assuming the temperature field is locally continuous in the particulate system, the temperature of the \( i \)-th neighboring particle \( (T_i) \) can be approximated by Taylor expansion as

\[
T_i = T_p + l_i \mathbf{e}_i \cdot \nabla T + \frac{1}{2} (l_i \mathbf{e}_i \cdot \nabla)^2 T + \frac{1}{6} (l_i \mathbf{e}_i \cdot \nabla)^3 T + O(l_i^4),
\]

where \( \mathbf{e}_i \) is the unit vector along \( l_i \).

(2)

(3)

(4)
where $\mathbf{e}_i = (\mathbf{r}_i - \mathbf{r}_p) / l_i$ is a unit vector pointing from particle $P$ to particle $i$. According to Eqs. (2) and (4), the net exchanged power between particles $i$ and $P$ can be calculated from

$$Q_{i\rightarrow P} = U_{i\rightarrow P} \left[ l_i \mathbf{e}_i \cdot \nabla T + \frac{1}{2} (l_i \mathbf{e}_i \cdot \nabla)^2 T + \frac{1}{6} (l_i \mathbf{e}_i \cdot \nabla)^3 T + O(l_i^3) \right].$$

(5)

Omit the high order terms and substitute Eq. (5) into Eq. (1), the energy balance equation can be rewritten as

$$\sum_{i=1}^{N_p} U_{i\rightarrow P} \left[ l_i \mathbf{e}_i \cdot \nabla T + \frac{1}{2} (l_i \mathbf{e}_i \cdot \nabla)^2 T \right] + S_P = \frac{dE_p}{dt}.$$  

(6)

Equation (6) is a second-order partial differential equation, much similar to a diffusion-type equation, but the connection between particle-scale parameters and continuum-scale parameters is still unclear and some manipulation are needed for further simplification. Consider a virtual control volume $V_{\text{cell}}$ occupied by the particle $P$ shown in Fig. 1, which is location dependent for system of non-uniform distribution of particles, and Eq. (6) divided by $V_{\text{cell}}$ yields

$$\mathbf{h}_{\text{non-loc}} \cdot \nabla T + \mathbf{K}_{\text{non-loc}} : \nabla \nabla T + S = \frac{dE}{dt},$$  

(7)

where $\mathbf{h}_{\text{non-loc}} = \sum_{i=1}^{N_p} \frac{U_{i\rightarrow P}}{V_{\text{cell}}} l_i \mathbf{e}_i [\text{W}/(\text{m}^2.\text{K})]$], $\mathbf{K}_{\text{non-loc}} = \sum_{i=1}^{N_p} \frac{U_{i\rightarrow P}}{2V_{\text{cell}}} l_i^2 \mathbf{e}_i [\text{W}/(\text{m}^2.\text{K})]$, $S = S_P / V_{\text{cell}} [\text{W}/\text{m}^3]$, $E = E_p / V_{\text{cell}} [\text{J}/\text{m}^3]$ are the continuum-scale parameters. $\mathbf{h}$ is a heat transfer coefficient vector that accounts for asymmetric NFRHT in system with non-uniform distribution of particles (hence called asymmetric radiative heat transfer coefficient in the following), $\mathbf{K}$ is an effective radiative heat conductivity tensor for NFRHT. The subscript ‘non-loc’ (short for ‘non-local’) indicates the parameters are dependent not only on the temperature of the local central particle $T_P$, but also that of all neighboring particles $T_i$. It is noted that $U_{i\rightarrow P}$ is a function of $T_i$, $T_P$ and $l_i$. The non-local parameters are not easy to be calculated.
In the following, the governing equation with parameters (\( h \) and \( K \)) dependent only on \( T_p \), namely, local parameters, is derived. By using second order Taylor expansion of \( \varphi(l, T) \) at \( T_p \), the \( U_{i=\alpha P} \) can be rewritten as

\[
U_{i=\alpha P}(T, T_p, l) = U_0(T, l) + \frac{1}{2} \frac{\partial U_0(T, l)}{\partial T_p} (T - T_p),
\]

where \( U_0(T, l) = \lim_{T \to T_p} U_{i=\alpha P}(T, T_p, l) = \frac{\partial \varphi(T_p, l)}{\partial T_p} \). Substitute Eq. (8) and \( T_i - T_p \approx l_i e_i \cdot \nabla T \) to Eq. (7), it yields

\[
\mathbf{h}'_{loc} \cdot \nabla T + \nabla \cdot \left( \frac{\partial \mathbf{K}_{loc}}{\partial T} \right) \nabla T + \mathbf{K}_{loc} : \nabla \nabla T + S = \frac{dE}{dt},
\]

where the asymmetric radiative heat transfer coefficient vector \( \mathbf{h}_{loc} \) and the effective NFRTC tensor \( \mathbf{K}_{loc} \) are defined as follows.

\[
\mathbf{h}'_{loc}(T, \mathbf{r}) = \frac{1}{V_{cell}(\mathbf{r})} \sum_{i=1}^{N_p} U_0(T, l_i(\mathbf{r})) l_i(\mathbf{r}) e_i,
\]

\[
\mathbf{K}_{loc}(T, \mathbf{r}) = \frac{1}{2} \frac{1}{V_{cell}(\mathbf{r})} \sum_{i=1}^{N_p} U_0(T, l_i(\mathbf{r})) l_i^2(\mathbf{r}) e_i e_i.
\]

Equation (9) is in non-conservative form, which can be further rewritten into a conservative form of normal-diffusion equation as follows (derivation details refer to the Supp. Material),

\[
\mathbf{h}_{loc} \cdot \nabla T + \nabla \cdot \left( \mathbf{K}_{loc} \cdot \nabla T \right) + S = \frac{dE}{dt},
\]

where the asymmetric radiative heat transfer coefficient vector in the conservative form governing equation, \( \mathbf{h}_{loc} \), has a much complex form given as follows.
\[ h_{\text{loc}} = \frac{1}{V_{\text{cell}}} \sum_{i=1}^{N_e} U_0(T, l_i(\mathbf{r})) l_i \mathbf{e}_i - \frac{1}{2} \frac{1}{V_{\text{cell}}} \sum_{i=1}^{N_e} l_i^2 \frac{\partial U_0(T, l_i(\mathbf{r}))}{\partial l_i} \nabla l_i \mathbf{e}_i \]

\[ -\frac{1}{V_{\text{cell}}} \sum_{i=1}^{N_e} U_0(T, l_i(\mathbf{r})) l_i \nabla l_i \mathbf{e}_i - \frac{1}{2} \nabla \left( \frac{1}{V_{\text{cell}}} \sum_{i=1}^{N_e} U_0(T, l_i(\mathbf{r})) l_i^2 \mathbf{e}_i \right) \]

The finally obtained normal-diffusion type governing equation, namely, Eq. (12) and Eq. (9), are the core findings of the present study. Note that the derivation is general and can be applied to one-, two- and three-dimensional (1D, 2D and 3D) particulate system.

By looking at Eq. (12), it is in a form similar to the HDE. An obvious difference still exists, namely, there is an additional convection-like term (the first-order term), which accounts for the asymmetric NFRHT in system with non-uniform distribution of particles, characterized by \( h_{\text{loc}} \).

Hence the derivation indicates that NFRHT in particulate system contains not only a diffusion process of heat transfer, but also a convection-type heat transfer process, and the latter shows the NFRHT has a preferential direction caused by the asymmetric distribution of particles in the system. During the derivation, the NFRTC tensor \( K_{\text{loc}} \) is revealed naturally as the coefficient of the diffusion term, indicating the validity of Fourier’s law for the diffusion-type NFRHT process.

As for uniform (isotropic) distributed particulate systems, there will be \( h_{\text{loc}} = 0 \) according to Eq. (13) due to symmetry, and Eq. (12) will become the same as the HDE with \( K_{\text{loc}} \) as the heat conductivity tensor.

In the following, we apply the developed normal-diffusion theory to analyze the steady-state NFRHT in 1D linear nanoparticle chains, 2D and 3D nanoparticle networks in vacuum. The calculated temperature field distribution based on the normal-diffusion theory are compared with the results solved directly from the exact fluctuational electrodynamics [21, 22] for verification. The calculated NFRTC from the new theory [\( K_{\text{loc}} \), Eq. (11)] are also compared with that predicted
using kinetic theory. During the calculation, the separation distance between any two particles in the systems is limited no shorter than $3a$ to ensure the validity of the dipole approximation [41].

Firstly, NFRHT in one-dimensional linear chains of uniform distribution of particles are considered. The left and right end of the chains are connected to hot (at 400 K) and cold reservoir (at 300 K), respectively, as shown in Fig. 2(a). The chains are composed of 60 nanoparticles, and three kinds of materials, namely, SiC, gold, and hBN, are considered. Structure information of the particle chains and optical properties of the materials considered in this work are consistent with that of the Refs. [36, 37]. Nanoparticle radius $a$ are 5 nm and 25 nm, and the lattice spacing $s$ are $4a$ and $7a$ for gold and hBN chain, respectively. While for SiC chain, $a$ is 25 nm and $s$ is $3a$. A heating power is added to each particle in the chains, the heating power at particle-scale ($S_p$) are $3\times10^{-13}$ W, 0 W and $2\times10^{-15}$ W for SiC, gold, and hBN nanoparticles, respectively. The equivalent volumetric heat source at continuum-scale ($S$) can be calculated by $S = S_p/V_{cell} = S_p/\pi sa^2$. The temperature distributions in the nanoparticle chains solved at continuum-scale based on the developed normal-diffusion theory [Eqs. (12) and (11)], and the HDE with NFRTC calculated using the kinetic theory, are shown in Fig. 2(a). The temperature distributions solved directly at particle-scale based on the fluctuational electrodynamics are considered as exact results for comparison. The temperature distribution obtained from the proposed normal-diffusion theory agrees very well with the exact result, which gives the maximum relative errors of 0.8%, 0.4% and 0.5% for SiC, Au and hBN chains, respectively. Dependence of the NFRTC on temperature for the three considered chains are shown in Fig. 2(b). The NFRTC for SiC and hBN chains are much larger than that of the Au chain, ascribed to the strong thermally accessible coupling in the former, as compared to the latter. The kinetic theory is completely inappropriate for calculating NFRTC of the Au chain due to the mismatch between the resonance frequency and the Planck’s window.
Generally, HDE combined with kinetic theory for NFRTC overestimates the temperature, as shown in Fig. 2 (a), which is attributed to the inappropriate NFRTC, as shown in Fig. 2 (b). Details on the solution of the derived governing equation for the NFRHT in linear chains are provided in the Supplemental Material.

**FIG. 2** (a) Temperature distributions in the nanoparticle chains solved based on the continuum-scale normal-diffusion theory developed in this work, and the classical heat diffusion equation with the NFRTC calculated using the kinetic theory, which are compared with the exact results calculated from the fluctuational electrodynamics at particle-scale. The left and right end of the chains are connected to hot (at 400 K) and cold reservoir (at 300 K). The chains contain 60 nanoparticles, which are composed of SiC \((a=25\,\text{nm}, s=3a)\) [37], Au \((a=5\,\text{nm}, s=4a)\) and hBN \((a=25\,\text{nm}, s=7a)\) [36], and the nanoparticles are heated with power \(S_p = 3 \times 10^{11} \, \text{W}, \, 0 \, \text{W} \text{ and } 2 \times 10^{15} \, \text{W}\), respectively. (b) The near-field radiative thermal conductivity as a function of temperature \(T\) for different nanoparticles chains, calculated from the normal-diffusion theory and the kinetic theory.

Then, NFRHT in linear chains of non-uniform distribution of hBN nanoparticles are considered, where the particles are distributed highly asymmetric, to demonstrate the convection-type NFRHT behavior. The particles are packed closely on the left side (at 400 K) and sparsely towards the right side (at 300 K). Two chains with different non-uniformity marked as Case 1 and Case 2, and a uniform-distributed chain marked as Case 3, are considered. The temperature
distributions in the three nanoparticle chains calculated at continuum-scale based on the normal-diffusion theory [Eq. (9)], and the HDE with $\mathbf{K}_{\text{loc}}$, are shown in Fig. 3 (a). Note that the HDE can be considered the omission of the convection term in the normal-diffusion equation [Eq. (12)], hence it cannot well account for the convection-type heat transfer in the asymmetric particle chains. For Case 3, the particles are uniformly distributed, the HDE predict the temperature distribution with good accuracy. While for Case 1 and Case 2, the particles are distributed unevenly, the HDE significantly underestimates the temperature distributions. In all the cases, the normal-diffusion theory predicts the temperature distributions with good accuracy compared to the exact results.

Strong non-linearity of temperature distribution is observed for the chain with high asymmetry of particle distribution. At particle-scale, the temperature of a nanoparticle in both Case 1 and Case 2 is more significantly affected by its left neighbor (where the particles are denser) than the right neighbor nanoparticles, since the near-field interaction from its left neighbor particle is much stronger than that from the right. Hence a preferred direction heat transfer exists along the chain from left to right, which is a convection-like process with convection direction to the right. The HDE has not taken this convection-like process into consideration, which is thus not sufficient to describe the NFRHT in highly asymmetrical systems. An asymmetric radiative heat transfer coefficient vector $\mathbf{h}_{\text{loc}}$ appears to characterize the convection-like process in the normal-diffusion theory for NFRHT in dense particulate system. The NFRTC and the asymmetric radiative heat transfer coefficient vary significantly from place to place, as shown in Fig. 3 (b) and (c), which are obtained from the original coefficients data at the position of the particles using the linear interpolation (details refer to Supplemental Material). Negative value of asymmetric radiative heat transfer coefficient shows a direction of convection-like heat transfer process from left to right. Both $\mathbf{K}_{\text{loc}}$ and $\mathbf{h}_{\text{loc}}$ decrease dramatically (several orders of magnitudes) from left side to the right.
side of the chain, consistent with the decreasing of near-field interaction with particle separation distance. From the above observation, special attention should be paid on NFRHT in non-uniform distributed particulate systems, where the HDE may not work well.

**FIG. 3** (a) Temperature distributions in the three hBN nanoparticle chains, the leftmost particle is fixed at 400 K, the rightmost particle is fixed at 300 K, which are calculated at continuum-scale based on the normal-diffusion theory developed in this work [Eq. (9)], and the classical heat diffusion equation (HDE). The results from exact fluctuational electrodynamics are shown as reference. The three different particle distributions are shown as inset. The radius $a$ of hBN nanoparticle is 25 nm. The number of particles in the chain is 20. The particles disperse more and more unevenly from the Case 3 to the Case 1. Particle coordinates, as well as some other necessary information for solving the continuum governing equation refer to the Supplemental Material. The spatial dependent near-field radiative thermal conductivity $K_{\text{loc}}$ and the asymmetric radiative heat transfer coefficient $h'_{\text{loc}}$ at 300 K are shown in (b) and (c), respectively.

Finally, the normal-diffusion theory is applied to 2D and 3D particulate networks to demonstrate that the derived Eq. (11) is a powerful tool for exploring the NFRTC of complex
multi-dimensional particulate networks. For 2D particulate networks, four different structures, i.e., random distribution, and square, hexagonal and honeycomb lattice distributions are considered. For 3D particulate networks, only random distribution is considered. Based on the normal-diffusion theory, the NFRTC is the key parameter to characterize the radiative heat transfer in statistically uniform particulate systems, since the asymmetric radiative heat transfer coefficient $h_{loc} = 0$. For the uniform particulate systems, average separation distance between neighboring nanoparticles, $c$, significantly affects the near-field interaction. A similar parameter is the volumetric filling fraction $f_v$. The dependence of the NFRTCs of the considered different 2D and 3D particulate networks of SiC nanoparticles with radius of 100 nm (at 300 K) on the filling fraction $f_v$ and $c$ are shown in Fig. 4. In addition, the NFRTC of 1D linear chains at different $f_v$ is also shown for reference. From computational resources consideration, the calculations are performed for systems of the size of 1000 nanoparticles.
FIG. 4 Dependence of near-field radiative thermal conductivity on the filling fraction for 1D, 2D and 3D uniform-distributed SiC particulate networks at 300 K. The calculations are performed for systems of the size of 1000 nanoparticles. Diagrams of the structures of the considered particulate systems are given in the insets. For 2D particulate networks, four different structures, i.e., random distribution, and square, hexagonal and honeycomb lattice distributions are considered. For 3D particulate networks, only random distribution is considered. The separation distance \( c \) of particles with \( f_v \) for the 1D case is added as the top axis. Analysis on the spectral radiative thermal conductivity is provided in the Supplemental Material for reference.

As shown in Fig. 4, under the same \( f_v \), the NFRTC for 3D particulate networks is significantly greater than that of the 2D particulate networks and the NFRTC for 2D particulate networks is significantly greater than that of the 1D linear chains. Since more neighboring particles exist in high-dimensional particulate networks, which will provides more heat transfer channels as compared to the relatively low-dimensional ones, which is consistent with the reported observation in the systems of only a few nanoparticles [21] or hundreds of nanoparticles [20]. For 1D linear chains, the dependence of the NFRTC on the filling fraction \( f_v \) varies radically, from \( \sim f_v \) to \( \sim f_v^{4.7} \).
with increasing $f_v$, which is ascribed to collective near-field interaction. The $f_v$ dependence of NFRTC for the 2D and 3D particulate networks is found to be different from that of the 1D particulate networks. No obvious change of the $f_v$ dependence of NFRTC for the 2D and 3D particulate networks are observed, which may be due to limited parameter range. When considering sufficiently sparse 2D and 3D particulate networks where near-field interaction is weak, the $f_v$ dependence of the NFRTC is expected to have a radical change as compared to that of the dense systems. For 2D particulate networks, the NFRTC and its dependence on $f_v$ for the different structures show slightly difference. For high $f_v$ (~0.3), the NFRTC of square lattice shows the highest value, whereas that of the random distribution shows lowest value. For low $f_v$ (~0.045), the NFRTC of honeycomb lattice shows the highest value, while that of square lattice shows lowest value. The number of proximity particles and the separation distance work together to determine the near-field interaction in the particulate networks. For higher filling fraction, the two factors are comparable, which account for the highest NFRTC of the square lattice network at high $f_v$ (~0.3). For small filling fraction, the minimum separation distance dominates near-field interaction, which accounts for the highest NFRTC of the honeycomb network at low $f_v$ (~0.045).

In summary, this work rigorously builds a bridge between particle-scale and continuum-scale NFRHT in particulate system. A normal-diffusion type governing equation is derived for NFRHT in dense particulate system at continuum-scale from the exact fluctuational electrodynamics formulation. A first-order term is shown to appear naturally due to the asymmetry of the particulate system, which reveals a convection-like transport behavior of the NFRHT in the highly asymmetrical systems. The NFRTC tensor is discovered as the coefficient of diffusion term and a new formula (Eq. (11)) for calculating NFRTC is given. For uniform particulate system, the derived normal-diffusion equation reduces to the classical heat diffusion equation. The accuracy
of the proposed theory is verified by comparison with exact results from fluctuational electrodynamics solution at particle-scale. The temperature field predicted by the proposed theory agrees very well the exact solutions both for systems of uniform and nonuniform distribution of particles. Whereas, the HDE cannot well capture the asymmetric heat transfer behavior for NFRHT in highly asymmetrical systems of particles. The new NFRTC formula (Eq. (11)) is demonstrated to be versatile and a powerful tool to calculate the NFRTC of multi-dimensional particulate systems. The NFRTC is observed to increase with particle filling fraction for 1D, 2D and 3D particulate system, and its dependence on filling fraction has a radical change for 1D particulate system due to the collective near-field effect. This work paves the way for understanding and analysis of heat diffusion in large-scale dense particulate system with near-field interaction. In prospect, the characteristics of heat diffusion process in particulate system of variety of materials, structures, and dimensions, especially the nonuniform distributed system, the effect of many-body interaction on the NFRTC, and coupled-mode heat transfer analysis in large-scale particulate system with considering NFRHT, to name a few, can be explored based on the proposed theory.

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References

[1] P. S. Venkataram, S. Molesky, W. L. Jin, and A. W. Rodriguez, Phys. Rev. Lett. 124, 013904 (2020).
[2] G. T. Papadakis, S. Buddhiraju, Z. X. Zhao, B. Zhao, and S. H. Fan, Nano Lett. 20, 1654 (2020).
[3] P. Ben-Abdallah, R. Messina, S.-A. Biehs, M. Tschikin, K. Joulain, and C. Henkel, Phys. Rev. Lett. 111, 174301 (2013).
[4] J. DeSutter, L. Tang, and M. Francoeur, Nat. Nanotechnol. 14, 751 (2019).
[5] M. Ghashami, H. Geng, T. Kim, N. Iacopino, S. K. Cho, and K. Park, Phys. Rev. Lett. 120, 175901 (2018).
[6] K. Kim, B. Song, V. Fernández-Hurtado, W. Lee, W. Jeong, L. J. Cui, D. Thompson, J. Feist, M. T. H. Reid, and F. J. García-Vidal, Nature (London) 528, 387 (2015).
[7] S. M. Rytov, Y. A. Kravtsov, and V. I. Tatarskii, Principiles of statistical radiophysics. 3. Elements of Random Fields (Springer-Verlag, Berlin, 1989).
[8] D. Polder and M. Van Hove, Phys. Rev. B 4, 3303 (1971).
[9] R. Carminati and J.-J. Greffet, Phys. Rev. Lett. 82, 1660 (1999).
[10] A. Narayanaswamy and G. Chen, Appl. Phys. Lett. 82, 3544 (2003).
[11] G. Domingues, S. Volz, K. Joulain, and J.-J. Greffet, Phys. Rev. Lett. 94, 085901 (2005).
[12] J. J. Loomis and H. J. Maris, Phys. Rev. B 50, 18517 (1994).
[13] A. V. Shchegrov, K. Joulain, R. Carminati, and J.-J. Greffet, Phys. Rev. Lett. 85, 1548 (2000).
[14] A. I. Volokitin and B. N. J. Persson, Phys. Rev. B 63, 205404 (2001).
[15] A. I. Volokitin and B. N. J. Persson, Phys. Rev. B 69, 045417 (2004).
[16] E. Rousseau, A. Siria, G. Jourdan, S. Volz, F. Comin, J. Chevrier, and J.-J. Greffet, Nat. Photonics 3, 514 (2009).
[17] S. Shen, A. Narayanaswamy, and G. Chen, Nano Lett. 9, 2909 (2009).
[18] R. S. Ottens, V. Quetschke, S. Wise, A. A. Alemi, R. Lundock, G. Mueller, D. H. Reitze, D. B. Tanner, and B. F. Whiting, Phys. Rev. Lett. 107, 014301 (2011).
[19] M. G. Luo, J. Dong, J. M. Zhao, L. H. Liu, and M. Antezza, Phys. Rev. B 99, 134207 (2019).
[20] M. G. Luo, J. M. Zhao, L. H. Liu, and M. Antezza, Phys. Rev. B 102, 024203 (2020).
[21] P. Ben-Abdallah, S.-A. Biehs, and K. Joulain, Phys. Rev. Lett. 107, 114301 (2011).
[22] J. Dong, J. M. Zhao, and L. H. Liu, Phys. Rev. B 95, 125411 (2017).
[23] M. Nikbakht, Phys. Rev. B 96, 125436 (2017).
[24] R. Messina and M. Antezza, Phys. Rev. A 89, 052104 (2014).
[25] R. Messina and M. Antezza, Phys. Rev. A 84, 042102 (2011).
[26] R. Messina and M. Antezza, Europhys. Lett. 95, 61002 (2011).
[27] L. X. Zhu and S. H. Fan, Phys. Rev. Lett. 117, 134303 (2016).
[28] L. X. Zhu, Y. Guo, and S. H. Fan, Phys. Rev. B 97, 094302 (2018).
[29] M. Krüger, G. Bimonte, T. Emig, and M. Kardar, Phys. Rev. B 86, 115423 (2012).
[30] B. Müller, R. Incardone, M. Antezza, T. Emig, and M. Krüger, Phys. Rev. B 95, 085413 (2017).
[31] B. Czapla and A. Narayanaswamy, J. Quant. Spectrosc. Radiat. Transfer 227, 4 (2019).
[32] A. Narayanaswamy and G. Chen, Phys. Rev. B 77, 075125 (2008).
[33] P. Ben-Abdallah, K. Joulain, J. Drevillon, and C. Le Goff, Phys. Rev. B 77, 075417 (2008).
[34] J. Ordonez-Miranda, L. Tranchant, S. Gluchko, and S. Volz, Phys. Rev. B 92, 115409 (2015).
[35] E. J. Tervo, O. S. Adewuyi, J. S. Hammonds, and B. A. Cola, Mater. Horiz. 3, 434 (2016).
[36] C. Kathmann, R. Messina, P. Ben-Abdallah, and S.-A. Biehs, Phys. Rev. B 98, 115434 (2018).
[37] E. J. Tervo, B. A. Cola, and Z. M. Zhang, J. Quant. Spectrosc. Radiat. Transf. 246, 106947 (2020).
[38] E. J. Tervo, M. Francoeur, B. Cola, and Z. M. Zhang, Phys. Rev. B 100, 205422 (2019).
[39] G. W. Mulholland, C. F. Bohren, and K. A. Fuller, Langmuir 10, 2533 (1994).
[40] C. F. Bohren and D. R. Huffman, *Absorption and Scattering of Light by Small Particles* (John Wiley & Sons, New York, 1983).
[41] P. Ben-Abdallah, Phys. Rev. Lett. 123, 264301 (2019).