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The heat transport mediated by near-field interactions in networks of plasmonic nanostructures is shown to be analogous to a generalized random walk process. The existence of superdiffusive regimes is demonstrated both in linear ordered chains and in three-dimensional random networks by analyzing the asymptotic behavior of the corresponding probability distribution function. We show that the spread of heat in these networks is described by a type of Lévy flight. The presence of such anomalous heat-transport regimes in plasmonic networks opens the way to the design of a new generation of composite materials able to transport heat faster than the normal diffusion process in solids.

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It is commonly admitted that heat conduction in a bulk solid is governed by a normal diffusion process. Heat carriers (phonons or electrons) move through the atomic lattice of material following a random walk [1] with a step length probability density which is a Gaussian. The heat spatial spreading from regions of high temperature to regions of low temperature is therefore intrinsically limited both by the speed of heat carriers and by the distance covered by them between two successive collision events. To go beyond this transport mechanism and accelerate the heat propagation within the medium, we propose here to add a supplementary channel for heat exchanges with long-range interactions such as those that exist in generalized random walks (GRW), processes where the step length probability is broadband. Lévy flights [2,3] are probably the most famous class of GRW in which extremely long jumps can occur as well as very short ones. The existence of photonic Lévy flights has been recently demonstrated [4] in self-similar materials, the so-called Lévy glasses. In those media, appropriately engineered so that photons perform random jumps with a probability distribution of step lengths which decays algebraically, the transport of propagative photons becomes superdiffusive. However, the magnitude of heat flux which can be transported with radiative photons is limited by the famous Stefan-Boltzmann law [5] and is several orders of magnitude smaller than the flux carried by conduction in solids. The situation radically changes when these photons become nonradiative. As predicted by Polder and Van Hove [6] 40 years ago and experimentally verified during the last few years [7–10], when two media out of thermal equilibrium are separated by a small distance (compared with their thermal wavelength), they exchange energy mainly by photon tunneling. In such a situation, the heat flux transported from one medium to the other one can surpass by several orders of magnitude the flux exchanged between two blackbodies [11,12] in the far field. In two recent works [13,14], we have established that a similar exalted heat transport can also exist at larger distances, thanks to many-body interactions. In this Letter, we investigate in detail how heat is transported throughout different plasmonic nanostructure networks which are either ordered or disordered. By analyzing the transport process through these structures as a GRW of a passive tracer in a medium, we demonstrate the existence of anomalous (superdiffusive) regimes driven by the collective near-field interactions.

To start this analysis, let us consider a three-dimensional network of spherical particles of radius R_i at temperature T_i, distributed inside an environment at temperature T_{env}. When the mean separation distance between two arbitrary particles is larger than their respective diameters and their size is small enough compared with the thermal wavelengths $\lambda_{T_i} = c\hbar/(k_B T_i)$, then this network can be modeled by a set of pointlike dipoles in mutual interaction and coupled to the surrounding bath. The time evolution of particle temperatures is governed by the following energy balance:

$$C_i \frac{\partial T_i}{\partial t} = \sum_{j \neq i} \mathcal{P}_{i \leftrightarrow j} + \mathcal{P}_{i \leftrightarrow B} + S_i, \quad (1)$$

where C_i represent the nanoparticle heat capacity, respectively, while $\mathcal{P}_{i \leftrightarrow j}$, $\mathcal{P}_{i \leftrightarrow B}$, and S_i denote the net power exchanged between two arbitrary particles, the power exchanged between a particle and the thermal bath, and the power received by a particle from an external source, respectively. Using the Landauer formalism introduced in Refs. [15,16] and extended in Ref. [13] to the N-body heat-transport problem, it can be shown that

$$\mathcal{P}_{i \leftrightarrow j} = 3 \int_0^\infty \frac{d\omega}{2\pi} [\Theta(\omega, T_i) - \Theta(\omega, T_j)] \mathcal{T}_{i,j}(\omega), \quad (2)$$

where

$$\mathcal{T}_{i,j}(\omega) = \frac{4}{3} \frac{\omega^4}{c^4} \mathrm{Im}(\alpha_i) \mathrm{Im}(\alpha_j) \mathrm{Tr}[\mathbb{G}_{ij} \mathbb{G}_{ij}^\dagger] \quad (3)$$

denotes the monochromatic transmission coefficient between the dipoles i and j expressed in terms of the dyadic Green tensor $\mathbb{G}_{ij}$ between the location of two dipoles and the particle polarizability α while $\Theta(\omega, T) = \hbar\omega / [\exp((\hbar\omega / k_B T)) - 1]$ is the mean energy of a harmonic oscillator at temperature T. The electric polarizability is given by the simple Clausius-Mossotti form $\alpha = 4\pi R^3 (\varepsilon - 1)/(\varepsilon + 2)$; its imaginary part sharply peaks at the particle plasmon resonance. We have checked that the radiative correction [17] to this polarizability, which is proportional to $(k_0 R)^3$, is negligible for nanoparticles in the Wien frequency range under consideration. Note that we have dropped the magnetic dipole contribution right from the start, which turned out to be negligible for the considered systems. For calculating the dyadic Green function (GF) in a system of N particles, we use the coupled-dipole equation [18]

$$\mathbf{E}_{ij} = \mu_0 \omega^2 \mathbb{G}_0^{ij} \mathbf{p}_{j \neq i}^{\mathrm{fluc}} + \frac{\omega^2}{c^2} \sum_{k \neq i} \mathbb{G}_0^{ik} \alpha_k \mathbf{E}_{kj}, \quad (4)$$

for $i = 1, \ldots, N$, combined with the linear response of a dipole source

$$\mathbf{E}_{ij} = \omega^2 \mu_0 \mathbb{G}_{ij} \mathbf{p}_j^{\mathrm{fluc}}. \quad (5)$$

Equation (4) gives the field exciting the particle i and coming from particle j. It contains a direct contribution, associated with the fluctuating dipole source $\mathbf{p}_j^{\mathrm{fluc}}$, and the contributions induced by all other particles. Here, $\mathbb{G}_0^{ij} = (\exp(ikr_{ij})/4\pi r_{ij})[(1 + (ikr_{ij} - 1)/k^2 r_{ij}^2)\mathbb{1} + ((3 - 3ikr_{ij} - k^2 r_{ij}^2)/k^2 r_{ij}^2)\hat{\mathbf{r}}_{ij} \otimes \hat{\mathbf{r}}_{ij}]$ is the free space GF defined with the unit vector $\hat{\mathbf{r}}_{ij} \equiv \mathbf{r}_{ij}/r_{ij}$, $\mathbf{r}_{ij}$ being the vector linking the center of dipoles i and j, while $r_{ij} = |\mathbf{r}_{ij}|$ and $\mathbb{1}$ stands for the unit dyadic tensor. Note that in a transparent host material, $k = n(\omega/c)$, where n is its refractive index. In the presence of interfaces, the free space GF must be replaced by the appropriate tensor, which takes into account the reflection and transmission of waves across all diopters. The second term of the right-hand side of Eq. (1) is the power exchanged in the far field with the environment

$$\mathcal{P}_{i \leftrightarrow B} = \bar{C}_{\mathrm{abs};i} \sigma_B (T_{\mathrm{env}}^4 - T_i^4), \quad (6)$$

where $\bar{C}_{\mathrm{abs};i}$ is the thermally averaged dressed absorption cross section of the ith particle [19] and σ_B is the Stefan-Boltzmann constant. In general, this coupling to the environment depends on the geometrical setup and considered time scale [20] and cannot be neglected *a priori*. But, for the geometry studied here, the power exchanged between the particles through near-field interactions is much more significant than the power exchanged with the environment at the beginning of the thermal relaxation process [21], so that $\mathcal{P}_{i \leftrightarrow B}$ can be neglected with respect to the other terms in Eq. (1).

In the following, we consider a situation near thermal equilibrium and expand the power exchange between dipoles [Eq. (2)]:

$$\mathcal{P}_{i \leftrightarrow j} = G(|\mathbf{r}_i - \mathbf{r}_j|)(T_j - T_i), \quad (7)$$

where we have introduced the thermal conductance at temperature T_i between dipoles i and j

$$G(|\mathbf{r}_i - \mathbf{r}_j|) \equiv \frac{\partial \mathcal{P}_{i \leftrightarrow j}(T_i)}{\partial T} = 3 \int_0^\infty \frac{d\omega}{2\pi} \frac{\partial \Theta(\omega, T_i)}{\partial T} \mathcal{T}_{i,j}(\omega). \quad (8)$$

Using this expression, the energy balance equation (1) can be recast into a Chapman-Kolmogorov master equation

$$\frac{\partial T_i}{\partial t} = \int_{R^d} p(\mathbf{r}_i, \mathbf{r}) \frac{T(\mathbf{r}, t)}{\tau(\mathbf{r})} d\mathbf{r} - \frac{T(\mathbf{r}_i, t)}{\tau(\mathbf{r}_i)} + \hat{S}_i, \quad (9)$$

which formally describes a system which is driven by a Markov process. The temperature distribution $T(\mathbf{r}, t)$ evolves in the same way as a generalized random walk, where jumps between positions $\mathbf{r}$ and $\mathbf{r}'$ occur with a probability distribution function (PDF) of step length proportional to $p(\mathbf{r}, \mathbf{r}') = (1/C\Delta V)\tau(\mathbf{r}) G(\mathbf{r} - \mathbf{r}')$ at a rate $\tau^{-1}(\mathbf{r}) = (1/C\Delta V) \int dr' G(\mathbf{r} - \mathbf{r}')$. The generalized walkers are injected at a rate $\hat{S}(\mathbf{r}) = S(\mathbf{r})/C$. [Here, C is the heat capacity per particle from Eq. (1), and $\Delta V = \bar{l}^3$ is the volume per particle in the system, $\bar{l}$ being the average distance.]

To analyze the transport of heat throughout the network, we investigate the distribution of step lengths $x = |\mathbf{r} - \mathbf{r}'|$. If the PDF $p(x)$ is Gaussian, all its moments $M^{(n)} = \int x^n p(x, t) dx$ are finite so that the regime of transport is diffusive on large scales. On the other hand, if it decays algebraically, so that at least one of its moments is divergent, the heat-transport regime becomes superdiffusive.

We first consider the case of linear chains ($d = 1$) of nanoparticles periodically dispersed in a transparent medium. Electromagnetic energy transport along linear chains

of metallic nanoparticles has been intensively studied in the past (see, for example, Refs. [22–24] and references therein). However, so far, very few studies have been devoted to heat transfer in these plasmonic systems [25]. The thermal conductance between a central particle and another one at distance Δx is plotted in Fig. 1 for different filling factors $(2R/h)$. We observe, for any density, that the large-distance tail of $G(x)$ asymptotically decays as $G = \mathcal{O}(1/x^2)$. By neglecting finite-size effects and remarking that τ is almost uniform [i.e., $\tau(x) \sim \tau_0$], $p(x, x)$ is a function of $x - x'$ only and the spatial Fourier transform of Eq. (9) gives

$$\frac{\partial \tilde{T}(k,t)}{\partial t} = \frac{1}{\tau_0}[\tilde{p}(k) - 1]\tilde{T}(k,t) + \tilde{\hat{S}}_i. \tag{10}$$

In the small wave number approximation (i.e., hydrodynamic limit), we get, after expanding $\tilde{p}(k)$ around $k = 0$ and coming back to the real space, the 1D Fokker-Planck equation which describes the transport process

$$\frac{\partial T(x,t)}{\partial t} = AT(x,t) - \mathcal{V}\frac{\partial T(x,t)}{\partial x} + D\frac{\partial^2 T(x,t)}{\partial^2 x} + \hat{S}, \tag{11}$$

where $A = (1/\tau_0)[\tilde{p}(0) - 1]$, $\mathcal{V} = -(1/\tau_0)(d\tilde{p}(0)/dk)$, and $D = (1/\tau_0)(d^2\tilde{p}(0)/dk^2)$ denote the transport coefficient, the drift term, and the diffusion coefficient, respectively, inside the chains. By noting that D is proportional to the second moment of the PDF, we immediately see

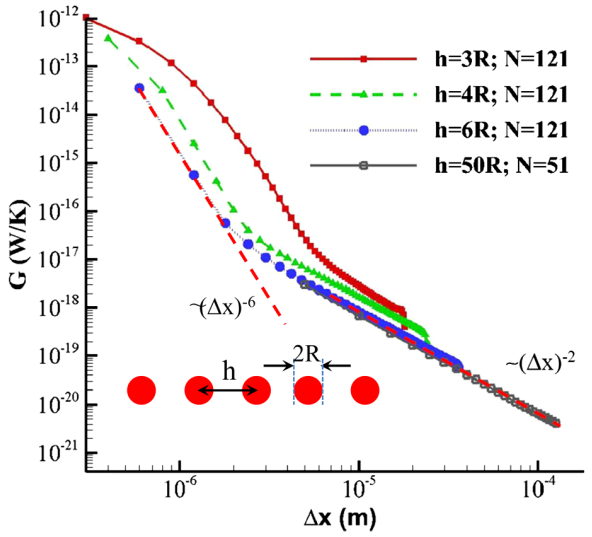

FIG. 1 (color online). Thermal conductance G in log-log scale for a chain of SiC spherical particles with different interparticle distances h and different particle numbers N as a function of the separation distance $\Delta x = |\mathbf{r} - \mathbf{r}'|$ at temperature $T = 300$ K. All particles are identical (radius of $R = 100$ nm), and their electric polarizability is given by the simple Clausius-Mossotti form $\alpha = 4\pi R^3[(\epsilon - 1)/(\epsilon + 2)]$ [17]. The dielectric permittivity of the particles is described by a Drude-Lorentz model [32]. The maximum distance for a given system is at half the chain length.

according to the asymptotic behavior of the conductance tail (see Fig. 1) that the diffusion coefficient is diverging, demonstrating so the superdiffusive behavior of heat transport in 1D dipolar chains. Besides, we clearly observe on these curves the transition between the region where the electrostatic regime ($\Delta x \ll \lambda_T$) dominates and the region where interactions take place at distances longer than the wavelength. In the first case, we distinguish two different behaviors. When $h = 6R$, the thermal conductance follows a power law in the $G \sim (\Delta x)^{-6}$ analog to what is usually observed between two isolated dipoles [26,27]. In denser chains, the dependence of the conductance on the interparticle distance is no longer the same. We see even in Fig. 1 that the thermal conductance tends to saturate at close separation distances in chains where extreme near-field interactions take place (i.e., $h = 3R$). This saturation could result from collective effects due to the multiple interactions between the particles [25,28], in particular, from strong dielectric screening of the electrostatic fields due to the high density. However, this behavior still remains an open problem today. At long distances (compared with λ_T), we see that $G \sim O(1/x^2)$ for any chain. In the diluted chain ($h = 50R$ in Fig. 1), all dipoles can be considered as isolated and they exchange heat in the far field mainly with their nearest neighbors [29]. As for the field magnitude (and that of the dyadic Green's tensor), it evolves from each dipole as $1/x$, so that the thermal conductance follows a $1/x^2$ power law.

Now, let us discuss the heat-transport process mediated by the near-field interactions in three-dimensional disordered networks made of N identical nanoparticles of identical radius R randomly distributed within a fictitious cubic box of side a as depicted in the inset of Fig. 2. Each realization is generated with a uniform distribution probability, and a minimum distance $r_{\min} \sim 2R$ is imposed between two adjacent particles in order to keep the dipolar approximation valid. Moreover, in any generated realization, a nanoparticle P_0 occupies the center of the simulation box and is used as a reference for the calculation of thermal conductance. The mean conductance $\langle G(x)\rangle$ is calculated by a double average: over a thin spherical shell of radius x centered at P_0 and over realizations of the dipolar network. The results are plotted in Fig. 2 vs distance x for different filling factors $f = NV/a^3$. Inspection of this figure shows that $\langle G(x)\rangle$ decays in power law as ζ/x^γ with an exponent γ which depends only on the filling factor f. According to this, the energy balance equation (9) (its statistical averaging) can be recast into a fractional diffusion equation

$$C_i\frac{\partial T_i}{\partial t} = -\kappa(-\Delta)^{\alpha/2}T(\mathbf{r}_i) + S_i, \tag{12}$$

with $\alpha = \gamma - d$. Here, $(-\Delta)^{\alpha/2}$ is the fractional Laplacian [30,31]

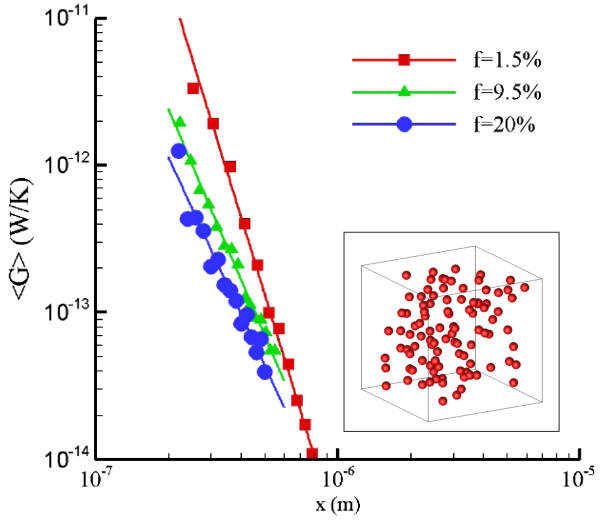

FIG. 2 (color online). Averaged thermal conductance $\langle G \rangle$ in log-log scale for clusters of SiC spherical particles as a function of the separation distance x for a different filling factor f and at temperature $T = 300$ K. The statistical averaging is performed with $m = 250$ realizations generated with a uniform random distribution probability. The inset shows an example of a network with a volumic fraction $f = 1.5\%$ generated with $N = 100$.

$$(-\Delta)^{\alpha/2} T(\mathbf{r}) = c_{d;\alpha} P \int_{R^d} \frac{T(\mathbf{r}) - T(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|^{d+\alpha}} d\mathbf{r}', \quad (13)$$

with $c_{d;\alpha} = [2^{-\alpha}\pi^{1+d/2}/(\Gamma(1 + \alpha/2)\Gamma((d + \alpha)/2) \times \sin(\alpha\pi/2))]$ and where P denotes the principal part. In Eq. (12), $\kappa = \zeta/(\Delta V c_{d;\alpha})$ is the fractional diffusion coefficient inside the plasmonic structure. The smaller the parameter α in the fractional Laplacian, the larger is the range of interactions through the medium. When $\alpha = 2$, we recover the classical Laplacian, and the heat transport becomes diffusive. If $\alpha > 2$, the transport is subdiffusive, while for $\alpha < 2$, it is superdiffusive. The long-range (nonlocal) interactions through the network are responsible for the existence of anomalous heat transport. At low f, the exponent γ is close to 5 (i.e., α is close to 2 but still smaller). Even at low filling, the exponent γ is slightly smaller than 5, so that the heat transport in a 3D network is already superdiffusive. This regime corresponds to plasmonic networks where the mean separation distance between the nearest particles is $\bar{l} > 6.6R$. In this situation, heat exchange is not limited to the closest neighbors and collective effects continue to play a role despite the medium being quite dilute. At higher densities, α decreases, showing that the heat transport becomes more and more nonlocal. At $f = 20\%$, i.e., in networks where extreme near-field interactions occur ($\bar{l} \simeq 2.7R$), we have $\alpha \simeq 0.64$, so that the heat transport becomes unambiguously superdiffusive.

We have demonstrated that the heat transport mediated by photon tunneling in plasmonic networks can be extremely superdiffusive. This nonlocal heat-transport mechanism allows us to go beyond the standard diffusion limit in solids. The ability to design nanocomposite materials able to transport heat faster than with phonons in solids opens new perspectives. It could find broad applications in different fields of material sciences that require ultrafast thermal management. Many fascinating questions on the links between the spatial structuration of plasmonic structure networks and the transport of heat through them remain open. For instance, the role played by the disorder and the presence of localized and delocalized modes is one of them. Also, the phonon-photon coupling within the plasmonic structures embedded in solids is a fundamental issue because it affects the transition between the superdiffusive regime and the classical diffusive transport.

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