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Heat transport through plasmonic interactions in closely spaced metallic nanoparticle chains

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Abstract. We report a numerical investigation on the near-field heat transfer through one-dimensional arrays of metallic nanoparticles closely spaced in a host material. Using the Landauer-Buttiker formalism we study the thermal behavior of chains in ballistic regime. It is shown that that the heat transport through plasmonic interactions can be several order of magnitude more efficient than in classical dipolar chains. This result could explain the anomalously high thermal conductivity observed in many nanoparticles based composite media such as nanofluids and could find broad practical applications in nanoscale thermal management.

1. Introduction

The enhancement of the thermal conductivity of thermal insulators such as cooling liquids and electronic insulators is a key issue in the fields of heat exchangers and of microelectronics. Embedding strongly interacting nano-objects seems a promising way to reach this goal. Indeed, these interactions lead to a very strong enhancement of exchanges close to the contact [1-2]. However so far the study of these exchanges has been limited to pairs of nanoparticles [3-5] separated by vacuum. In closely spaced nanoobjects lattices, the physics of heat and momentum transfers still remains to be described. Indeed, in such structures, the multipolar moments supported by each object are able to interact with its counterparts giving rise to new channels [6] for heat and momentum transport through the so called “collective plasmon modes” (CPM). In the present work, we study in nanoparticle chains how these CPM contribute to the heat transport when the separation distance between two neighboring particles is small in front of their characteristic size. The thermal behavior of these chains in ballistic regime is studied using the Landauer-Buttiker theory [7].

To start, let us consider a one-dimensional straight chain of polarizable metallic spherical particles of radius a dispersed in a homogeneous isotropic dielectric host material with a inter-particle distance d. An electric field $V_{ext}(n)$ applied on the charge distribution of the nth particle induces on it a multipolar moment $q_{lm}(n)$ of order (l,m) [8]. Taking into account the interactions due to the others particles we obtain [9-10]

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where $A_{lmn}^{nn'}$ is the coupling strength between the multipole moment of order $(l, m)$ at the position of the $n$th particle and the multipole moment of order $(l', m')$ at the $n'$th particle while $\alpha_{lmn}$ is the multipolar polarizability [10] of the $n$th particle. According to the theory of spectral representation introduced by [11] this coefficient is purely geometric and writes, for one-dimensional periodic lattices

$$A_{lmn}^{nn'} = \begin{cases} 0 & \text{if } n = n' \\
(-1)^n 4\pi \frac{(l + l')!}{[(2l + 1)(2l' + 1)]^{1/2}} \left[ \frac{1}{[(l + m)(l' + m)(l - m)(l' - m)]^{1/2}} \right] \\
\times \left( \frac{n' - n}{p' - p} \right)^{1/2} \frac{1}{d^{1/2}} \left( \frac{n' - n}{p' - p} \right)^{1/2} \delta_{nn'}^{nn'} & \text{if } n \neq n' \end{cases} \quad (2)$$

Seeking a solution of linear system (1) under the oscillating form $q_{lmn} = q_{lmn} \exp[j(\omega_{lm} t - k m)]$, where $\omega_{lm}$ is a complex pulsation we obtain, using the Drude model $\varepsilon_p(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega - j\gamma)}$ ($\omega_p$ is the plasma resonance pulsation of metal and $\gamma = \tau^{-1}$ its damping factor), the multipolar moments must satisfy

$$\sum_{n' \neq n} (\frac{s}{2l + 1} \delta_{lmn}^{nn'} + \frac{t}{4\pi} \frac{1}{l^{1/2}} l^{1/2} d^{1/2}) \sum_{n' \neq n} (-1)^r A_{lmn}^{nn'} \exp[j(n - n')k d] \bar{z}_{nn'}^{nn'} - \frac{1}{2l + 1} \delta_{lmn}^{nn'} = \frac{1}{4\pi} V_{lmn}^{nn'} \exp[jk d] \bar{z}_{lmn}^{nn'}$$

where we have set $\bar{z}_{lmn} = \omega_{lm}(\omega_{lm} - i\gamma)/\omega_p^2$, $s = l(l + \varepsilon_m) + 1$, $t = l(1 - \varepsilon_m)$ and $\bar{q}_{lm} = q_{lmn}/(l d^{2l+1})^{1/2}$, $\varepsilon_m$ being the dielectric permittivity of host medium. Here $\delta_{lmn}^{nn'}$ denotes the product of two Kronecker symbols relative to the couples $(l, l')$ and $(m, m')$, respectively. The calculation of eigenvalues $\bar{z}_{lmn}$ of this system allow us to determine the dispersion relation of resonant collective modes (eigenmodes) of the chain $\omega_{lm}(k) = \frac{1}{2} [-4\bar{z}_{lmn}(k) \omega_p^2 - \gamma^2 + j\gamma]$.

### 2. Multipolar thermal conductance in ballistic regime

In order to estimate the multipolar thermal conductance of a chain we use an approach analogous to that used in the Landauer theory of electronic transport. Two heat reservoirs which are maintained at neighborhood temperatures $T$ and $T + \delta T$ (the temperature difference $\delta T << \frac{2T + \delta T}{2}$) are linked together with the chain we want to characterize. Assuming a perfect coupling between the reservoirs and the plasmons of the chain, the thermal conductance associated to CPM is given by [7]

$$G = \frac{\hbar}{2\pi k_B T^2} \sum_{l,m} \int_{-l/2}^{l/2} \omega_{lm}(k) v_{ph}(k) \frac{e^{\beta \omega_{lm}}}{(e^{\beta \omega_{lm}} - 1)^2} dk, \quad (4)$$
where \( k \) stands for the wave vector, \( \omega_{lm}(k) \) is the pulsation of \( m \)th mode and 
\[ f_p(\omega) = [\exp(\beta\hbar\omega) - 1]^{-1} \]
the Bose-Einstein distribution function. In figure 1, we have plotted the dispersion relation of collective plasmons supported by a chain of copper nanoparticles in a lossless and non-dispersing host material when the nanoparticles (10 nm radius) are in contact in a positive and a negative host dielectric. The plasma frequency, is given by the usual relation
\[ \omega_p = (\rho_e e^2 / \varepsilon_0 m^*)^{1/2} \]
where \( \rho_e \), \( e \), \( \varepsilon_0 \), and \( m^* \) denote the electronic density, the charge of electrons, the permittivity of vacuum and the effective mass of electrons. The physical properties of copper used to calculate the transport properties were \( \rho_e = 8.5 \times 10^{28} \text{ m}^{-3} \) [12], \( \gamma = 1.38 \times 10^{13} \text{ s}^{-1} \) [13], \( m^* = 1.42 \times m_e \) [14] (\( m_e \) being the mass of free electrons). In positive dielectric host the \( G(d) \) curves displayed in figure 2 let clearly appear that the higher multipoles (\( l > 1 \)) participate strongly to the energy transport at very short distance.

We see on figure 1 that for nanoparticles close to the contact (only the contact is plotted on figure 1), the branches of multipoles in a positive host dielectric are as much distorted than the dipolar ones (figure 1-a.1). On figure 2 we see that this lead to an enhancement of \( G \) close to the contact. In negative host dielectric (see figures 1-b.1 and 2-b.2) the optical behavior of the chain radically changes. Indeed, contrary to the previous configurations, some of multipoles have a purely imaginary pulsation (figure 1-b.2) at least over a part of the Brillouin zone. Then, these modes do not participate anymore to the ballistic transport through the chain and they limit the thermal conductance of the chain to very low values. As a direct consequence of the presence of ‘non-ballistic’ modes, we have found (not plotted here) that the thermal conductance drastically falls down to zero.

3. Conclusion
In a positive dielectric host, we have shown that the multipole interactions enhance, by several order of magnitude, the thermal conductance of the chain near to the contact. This could explain the anomalously high thermal conductivity recently observed in many nanoparticles colloidal suspensions [15]. In contrary, in negative dielectric matrix, some of modes become not propagative and do not participate anymore to the heat exchanges so that the plasmonic thermal conductance becomes extremely low.
**Figure 2.** Plasmonic thermal conductance at 900 K of linear chains of 10 copper particles 10 nm radius vs the separation distance (a) and vs the dielectric constant of host material $\varepsilon_m$ (b) when $l \leq 5$. The inset in (a) is the geometric configuration studied. Regularly spaced nanoparticle chains are connected to two reservoirs kept at neighborhood temperatures. The separation distance $d$ between particles is comparable or smaller than the particle diameter $2a$.