Radiative heat transfer between metallic nanoparticles

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In this letter, we study the radiative heat transfer between two nanoparticles in the near and far fields. We find that the heat transfer is dominated by the electric dipole-dipole interaction for identical dielectric particles and by the magnetic dipole-dipole interaction for identical metallic nanoparticles. We introduce polarizability formulas valid for arbitrary values of the skin depth. While the heat transfer mechanism is different for metallic and dielectric nanoparticles, we show that the distance dependence is the same. However, the dependence of the heat flux on the particle radius is different. © 2008 American Institute of Physics. [DOI: 10.1063/1.2931062]

In a vacuum, the heat flux between two bodies is only due to radiative heat transfer. It was predicted four decades ago1 that the radiative heat flux exchanged by two bodies increases dramatically when the distance between them decreases down to distances on the order of the peak wavelength of Planck’s spectrum. The heat transfer between two nanoparticles has been theoretically predicted by modeling the nanoparticles by using electric dipoles.2–4 Experiments between macroscopic media5,6 or involving a nano-object7 have been reported. Different behaviors for polar materials and metallic ones were found for the interaction of a surface for polar or metallic particles. It decays as 1/d in the near field and as 1/d2 in the far field. We find that the radius dependence differs for metallic and dielectric nanoparticles.

The derivation of the radiative heat flux Φ = ∆P exchanged by two nanoparticles (see Fig. 1) is done in the framework of fluctuational electrodynamics.8,9 It is based on the absorption by a particle (P1) of the electromagnetic fields generated by the random currents in the other particle (P2) and vice versa.

The power absorbed by P1 reads

\[ P = \int d^3 \vec{r} \left< \vec{j}(\vec{r}, t) \cdot \vec{E}_{inc}(\vec{r}, t) \right> \]

where \( \vec{j} \) is the electric current density in P1 and \( \vec{E}_{inc} \) is the electric field at point \( \vec{r} \) in this particle. We consider a particle with radius \( R \) such that \( R \ll \lambda \), where the vacuum wavelength is the electric field at point \( \vec{r} \) in this particle. We consider a particle with radius \( R \) such that \( R \ll \lambda \), where the vacuum wavelength is the electric field at point \( \vec{r} \) in this particle. We consider a particle with radius \( R \) such that \( R \ll \lambda \). 

The power absorbed by a particle illuminated by a plane wave is the product of the incident flux by the particle absorption cross section. The latter reads11

\[ C_{abs} = C_{ext} - C_{sca}. \]

A first order expansion in \( x = kR \) yields

\[ C_{abs} = \frac{6 \pi}{k^2} \text{Re}(a_1 + b_1). \] (1)

Note that this expansion is done for arbitrary values of \( y = \sqrt{\varepsilon(\omega)}/\lambda \). \( a_1 \) and \( b_1 \) are the first Mie coefficients. They are related to the electric and magnetic dipolar moment, respectively.11,12 The power absorbed by the particle can be cast in the form

\[ P = \left< \frac{\partial \vec{p}}{\partial t} \cdot \vec{E}_{inc} - \vec{m} \cdot \frac{\partial \vec{B}_{inc}}{\partial t} \right>, \] (2)

where \( \vec{E}_{inc} \) and \( \vec{B}_{inc} = \vec{B}_{inc}/\mu_0 \) are the incident electric and magnetic fields and \( \vec{p} \) and \( \vec{m} \) are the electric and magnetic dipolar moments of the particle. The dipolar approximation is valid provided that \( \lambda \gg R \) and \( d \gg R \), where \( d \) is the distance between the center of the nanoparticles. In practice, a distance on the order of a few radii \( (8) \) appears to be sufficient.9 For smaller distances higher multipoles must be included.4,13,14

For metallic nanoparticles, it has been shown8,15 that the magnetic dipole moment gives a significant contribution to absorption in the near field. These works focus on very small nanoparticles \((R \ll \delta) \) (Ref. 8) or larger ones \((R \gg \delta) \). As the particle sizes are generally not smaller or larger than the skin depth in the full contributing spectrum, an extended formula valid for arbitrary-large skin depths is needed. Provided that \( R \ll \lambda \), we derive from Mie’s solution the following magnetic polarizability:

\[ C_{mag} = \frac{4 \pi}{k^2} \text{Re}(a_2 + b_2). \]

FIG. 1. (Color online) Two nanoparticles with radii \( R_1 \) and \( R_2 \) at an inter-

particular distance \( d \). The skin depths \( \delta_1 \) and \( \delta_2 \) are the fundamental quantities for metallic particles.

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\[ \alpha_E(\omega) = -2\pi R^3 \left[ 1 - \frac{x^2}{10} + \left( -\frac{3}{y^2} + \frac{3}{y \cot y} \right) \frac{1 - x^2}{6} \right]. \]  

(3)

In the limit \( R \ll \delta \), Eq. (3) yields the magnetic dipolar moment valid for very small particles

\[ \alpha_H(\omega) = \frac{2\pi}{15} R^3 (k R)^2 [\varepsilon(\omega) - 1]. \]  

(4)

The electric polarizability takes the simple Clausius–Mossotti form \( \alpha_E(\omega) = 4\pi R^3 [\varepsilon(\omega) - 1]/[\varepsilon(\omega) + 2] \) if the skin depth is much larger than the radius. For dielectric nanometer scale particles, this is often a very good approximation. Yet, the skin depth effect may play a role. It is then useful to work with a more general form derived from the Mie solution

\[ \alpha_E(\omega) = 2\pi R^3 \left[ 2(\sin y - y \cos y - x^2 \left( -\frac{\sin y}{y^2} + \frac{\cos y}{y} + \sin y \right) ) \right] / \left[ (\sin y - y \cos y + x^2 \left( -\frac{\sin y}{y^2} + \frac{\cos y}{y} + \sin y \right) ) \right]. \]  

(5)

Note that the famous Clausius–Mossotti formula is recovered in the limit \( R \ll \delta \). We have represented in Fig. 2 the imaginary part of the electric and magnetic polarizabilities for different values of the radius of a gold nanoparticle. It is seen that the simplest forms are valid for any frequency when the radius is smaller than 20 nm.

Equation (2) leads to

\[ P = \int_{\omega=0}^{+\infty} \frac{d\omega}{2\pi} (\omega \text{Im} \alpha_E(\omega)) e_0(\mathcal{E}_m|^2) + \text{Im} \alpha_H(\omega) \mu_0(\mathcal{H}_m|^2), \]  

(6)

where \( e_0 \) and \( \mu_0 \) are the free space permittivity and permeability. The incident fields are generated by the fluctuating dipoles. The fluctuation-dissipation theorem\(^9,^{16,17}\) yields

\[ \langle p_m(\vec{r}, \omega) p_m^*(\vec{r}, \omega') \rangle = \frac{4\pi e_0}{\omega} \text{Im}(\alpha_E) \Theta(\omega, T) \delta(\omega - \omega') \delta_{m\ell}, \]  

(7)

where \( \Theta(\omega, T) = \frac{\hbar \omega}{[\exp(\hbar \omega/k_B T) - 1]} \) is the mean energy of an oscillator \((\hbar \text{ and } k_B \text{ are the Planck and Boltzmann constants})\). In a nonmagnetic medium \((\mu_1)\), the magnetic dipoles satisfy

\[ \langle m_m(\vec{r}, \omega) m_m^*(\vec{r}, \omega') \rangle = \frac{4\pi}{\omega} \text{Im}(\alpha_H) \Theta(\omega, T) \delta(\omega - \omega') \delta_{m\ell}. \]  

(8)

By using a Green’s function technique\(^9,^{20}\) relating the source dipoles to the emitted fields, we obtain

\[ \Phi = \sum_{m=H \text{ or } E} \int_{0}^{+\infty} \frac{1}{4\pi} [\Theta(\omega, T_1) - \Theta(\omega, T_2)] \text{Im}(\alpha_E(\omega)) \text{Im}(\alpha_E(\omega)) [\mathcal{E}_m(\omega)] \left[ \frac{3}{(kd)^6} + \frac{1}{(kd)^4} + \frac{1}{(kd)^2} \right] d\omega. \]  

(9)

We checked that multiple scattering is negligible for sufficiently small particles. It is not taken into account in Eq. (9).

The final result has the same structure for the electric and magnetic dipolar moments. It shows that the heat flux dependence on distance is the same for polar and metallic materials. This is in contrast with other studies with larger bodies, e.g., parallel surfaces\(^{21}\) or nanoparticle/surface\(^8,^{15}\).

We now focus on particles with the same radii. Figure 3 shows the heat transfer between two dielectric (SiC) and two metallic nanoparticles. It is seen that the electric dipole yields the dominant contribution for dielectric particles whereas the magnetic contribution dominates the heat transfer for gold nanoparticles, for the whole range of distances. Heat transfer between metallic nanoparticles is thus mainly due to the Joule dissipation of eddy currents. A quasistatic approximation neglecting magnetic fields is valid for dielectric particles but not for metallic particles. It is also worth noting that the \( 1/\ell^4 \) term is almost negligible. The near-field term decays as \( 1/\ell^6 \) and the far-field one as \( 1/\ell^2 \). In the first approximation, one can retain the sum of these two contributions.

We now discuss the dependence of the heat transfer on the particle radius. The flux varies as the square of the polarizability so that it varies as \( R^2 \) for dielectric particles that have the same radii. The dependence is different for the magnetic contribution that dominates the flux for metallic materials.
particles. At first sight, the polarizability varies as $R^3$ from Eq. (4). However, for the very small particles, the confinement effect introduces a further dependence of the dielectric constant on $R$ (see Ref. 22). We have $\text{Im}(\varepsilon) \approx R\alpha_p/(\omega\nu)$. It follows that the flux varies as $R^{12}$. This dependence becomes weaker when the radius increases.

In summary, we have given here a simple and general formula for the near-field radiative heat transfer between two nanoparticles, valid for $\lambda \gg R$, $d \gg R$, and any value of the skin depth. We have shown that heat transfer is dominated by magnetic fields for metallic nanoparticles and by electric fields for dielectric nanoparticles. Research on heat transfer properties of composite media including nanoparticles should benefit from this work.

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