Physica B 433 (2014) 67–71

Contents lists available at ScienceDirect

Physica B

journal homepage: www.elsevier.com/locate/physb

On thermal vacuum radiation of nanoparticles and their ensembles

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ARTICLE INFO

Article history:
Received 10 September 2013
Accepted 10 October 2013
Available online 17 October 2013

Keywords:
Radiative heat transfer
Black-body radiation
Nanoparticles
Fluctuation-electromagnetic interactions

ABSTRACT

Radiant emittance of dimers and ensembles of particles consisting of gold, graphite and silica glass nanoparticles in vacuum is studied numerically based on the fluctuation-electromagnetic theory. The presence of neighboring particles of the same temperature causes an oscillating character of radiant emittance (per one particle) depending on the particle size, interparticle distance and temperature. We conclude that an ensemble of particles could be a much more intense source of thermal radiation than an equivalent solid body with the same outer surface area. Alternatively, when the neighboring particles create a significant "screening effect" (silica), an ensemble of particles could be a very good heat protector.

1. Introduction

A role of paramount importance in nanoelectronics, nanophotonics, and photovoltaics is played by heat dissipation. Radiative heat exchange processes provide a convenient way of handling the excess of heat produced in these devices [1]. On the other hand, there is a great interest in studying the properties of high-temperature clusters [2] and dust particles in fusion devices [3] where the processes of radiative heat transfer between the particles and the surrounding vacuum are essential in the energy balance. However, in describing thermal vacuum radiation of small particles of submicron dimensions the Planck's law of blackbody radiation is not sufficient.

As is well known, blackbody radiation mediates heat exchange between bodies placed in vacuum and separated by large distance d compared to the thermal wavelength $\lambda_T = 2\pi\hbar c/k_B T$. For the two parallel plates of area S that leads to the radiative heat transfer rate (RHT) corresponding to the Stefan–Boltzmann law $W_{SB} = \frac{\pi^2}{60}\frac{k_B^4}{\hbar^3 c^2} S T^4$ independent of d. However, at $d < \lambda_T$ the rate is enhanced greatly due to the involvement of evanescent waves [4,5] and satisfies a $1/d^2$ dependence at small distances d, as shown in the experiments [6,7].

In the case of two spherical particles of radius r separated by a small vacuum gap of width d, a leading near-field contribution to RHT obeys the law $1/(2r+d)^6$ [8–10]. In Ref. [11], the authors have reconsidered this problem within the assumption of dipolar response by taking into account radiative corrections and electromagnetic crossed terms mixing the electric and magnetic particle responses. In particular, it was shown that crossed terms could play a dominant role in heterogeneous structures, and become less important in homogeneous ones when calculating RHT between the two particles of different temperature.

In this regard, no attention to date was paid to thermal radiation emitted into the surrounding vacuum provided that neighboring particles have the same temperature. Since the near-field interactions can be significant, the presence of another particle (or an ensemble of particles) affects thermal radiation of each particle when the ensemble and vacuum environment are characterized by different temperatures. An intriguing question is whether the energy radiated by a particle be higher than the corresponding black-body limit or not, and how the answer depends on material properties, the size and spatial configuration of particles. It is worth noting that the finite-size effect of thermal radiation can be traced even within the simplest quantum approach, since there exists a non-zero minimum for the radiation frequency due to the finite size of the body [12]. Thermal radiation of nanoparticles was also considered in Ref. [13] using a simplified model approach.

In this work, based on the fluctuation-electromagnetic theory, we calculate RHT (W) for a spherical particle of radius r and temperature T_1 which is a part of dimer or an ensemble of particles at temperature T_1 embedded in the environment at temperature T_2. We use the assumption of dipolar response within the Mie theory [14] and tabulated dielectric characteristics of materials from Ref. [15]. Despite that net RHT between the particles is absent, each of them emits (absorbs) thermal photons into (from) vacuum background and the rate of this process depends on a spatial configuration of particles embedded in vacuum. In the simplest case of two particles (dimer) separated by a center-to-center distance $R \geq 2r$, the cooperative effect and RHT between the particles and vacuum environment prove to be the functions of R.

Numerical calculations were performed for gold, graphite and silica glass (SiO_2) particles with radii ranging from 0.1 to $1-3\ \mu m$

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http://dx.doi.org/10.1016/j.physb.2013.10.012

in the temperature range from 100 K up to the melting temperature of the corresponding bulk materials. We calculated a normalized RHT (blackness factor) W/W_{SB} depending on the particle radius and temperature. The sharp difference in the dependence of the blackness factor on temperature and radius was found for the particles of gold and graphite on the one hand, and SiO_2 particles on the other hand. In the former case, the maximum ratio of W/W_{SB} is observed for particles with a radius of $0.15-0.35\ \mu m$, increasing with temperature. For graphite particles, W/W_{SB} reaches 0.92 at $T=3000$ K, but for gold particles the highest blackness factor does not exceed 0.084 at $T=1337$ K. Interestingly, the blackness factor for SiO_2 particles reaches the maximum value of 0.55 at a much greater particle radius of $3.4\ \mu m$ and a much lower temperature of 223 K.

In the case of dimers, the dependence of W/W_{SB} on the distance R between the particles demonstrates an oscillating character. The presence of a neighboring particle may lead either to a decrease of RHT ("screening effect"), or to an increase of RHT ("mirror effect"), depending on the particle radius and temperature. The highest screening (up to 9%) is found for SiO_2 particles of radius $1\ \mu m$ at a temperature of 600 K, while the highest "mirror effect" does not exceed 3% for all types of the particles.

For an ensemble of N closely packed nanoparticles, their total RHT in vacuum rises proportionally to N, while the black body radiation from a solid body with the same outer radius rises as $N^{2/3}$. Therefore, at $N \gg 1$ the ensemble of nanoparticles should be a much more intense source (absorber) of thermal radiation than an equivalent solid body of the same size.

2. Theory

Fig. 1 shows the simplest system under study, namely the case of two identical isotropic spherical particles 1 and 2 in the vacuum environment. We assume that the particles are nonmagnetic and have the frequency-dependent electric and magnetic dipole polarizabilities $\alpha_{1,2}{}^{(e)}(\omega)$, $\alpha_{1,2}{}^{(m)}(\omega)$. Within the framework of fluctuation electrodynamics, the rate of heating (cooling) of each particle (particle 1 for definiteness) can be represented in the form [16]

$$\dot{Q} = \langle \dot{\mathbf{d}}_1(t)\mathbf{E}(\mathbf{r}_1,t)\rangle + \langle \dot{\mathbf{m}}_1(t)\mathbf{B}(\mathbf{r}_1,t)\rangle \\ = \int_{-\infty}^{+\infty} \frac{d\omega'}{2\pi}\frac{d\omega}{2\pi}(-\mathrm{i}\omega)\exp\left[-i(\omega+\omega')\right]\cdot \\ \left[\langle d_{1,i}(\omega)E_i(\mathbf{r}_1,\omega')\rangle + \langle m_{1,i}(\omega)B_i(\mathbf{r}_1,\omega')\rangle\right] \qquad (1)$$

Here $\mathbf{d}_1(t), \mathbf{m}_1(t)$ – electric (magnetic) dipole moments of particle 1, $\mathbf{E}(\mathbf{r}_1,t), \mathbf{B}(\mathbf{r}_1,t)$ – spontaneous electric and magnetic field at the location point $\mathbf{r}_1$ of particle 1, $d_{1,i}(\omega), m_{1,i}(\omega), E_i(\mathbf{r}_1,\omega'), B_i(\mathbf{r}_1,\omega')$ are the corresponding projections of the Fourier-transforms, points over variables indicate the time derivatives. It should be noted that all the aforementioned quantities include spontaneous and induced components of the dipole moments and fields. More details are given in Ref. [17], where we have calculated the heat transfer and other important characteristics in the system of two rotating particles placed in vacuum. The basic formula which we need follows from equation (30) in Ref. [17] assuming that $\Omega=0$ (rotation frequency) and making use the replacements

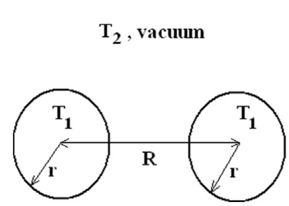

Fig. 1. A scheme of particle configuration.

$T_3 \to T_2, T_2 \to T_1$:

$$\dot{Q} = -\frac{2\hbar}{\pi c^3}\int_0^\infty d\omega\omega^4 \mathrm{Im}\,\alpha_1{}^{(e)}(\omega)\left[\coth\frac{\hbar\omega}{2k_BT_1} - \coth\frac{\hbar\omega}{2k_BT_2}\right] \\ -\frac{2\hbar}{\pi}\int_0^\infty d\omega\omega\left(\frac{\omega^2}{\hbar c^2}\right)^2\left\{\mathrm{Im}\,\alpha_1{}^{(e)}(\omega)\mathrm{Im}\,D_{ik}(\omega,\mathbf{R})\mathrm{Re}\left[\alpha_2{}^{(e)}(\omega)D_{ik}\right.\right. \\ \left.(\omega,\mathbf{R})\right]\left[\coth\frac{\hbar\omega}{2k_BT_1} - \coth\frac{\hbar\omega}{2k_BT_2}\right]\Bigg\} + (e\to m) + (e,m) \qquad (2)$$

$$D_{ik}(\omega,\mathbf{R}) = \left(-\frac{\hbar c^2}{\omega^2}\right)\exp\left(i\omega R/c\right)\left[\left(\frac{\omega^2}{c^2R} + \frac{i\omega}{cR^2} - \frac{1}{R^3}\right)(\delta_{ik} - n_in_k) \right. \\ \left. + 2\left(\frac{1}{R^3} - \frac{i\omega}{cR^2}\right)n_in_k\right] \qquad (3)$$

where $D_{ik}(\omega,\mathbf{R})$ – components of the retarded Green's function of photons in vacuum ($\mathbf{n}=\mathbf{R}/R$, $i,k=x,y,z$), the term $(e\to m)$ is identical to the first two terms and describes magnetic polarization effects, while the last term (e,m) corresponds to the crossed magnetic–electric polarization contributions. As was shown in Ref. [11], the crossed terms can be significant in heterogeneous metal–dielectric systems. Here we consider only the case of metal–metal and dielectric–dielectric combinations neglecting these crossed terms. Moreover, Eq. (2) does not include the processes of multiple scattering, since they are also negligibly small [11]. If the particles have different temperatures, Eq. (2) involves an extra term describing direct heat exchange between the particles [8–11]. In addition, it is worth noting that the first term in Eq. (2) corresponds to the limiting case $R\to\infty$ or if we neglect the cooperative effect of another particle [10], which is described by the second term of Eq. (2).

After substituting Eq. (3) in Eq. (2) the second term of Eq. (2) takes the form

$$\dot{Q}^{(2)} = -\frac{2\hbar}{\pi}\int_0^\infty d\omega\omega\mathrm{Im}\,\alpha_1{}^{(e)}(\omega)\left[\mathrm{Re}\alpha_2{}^{(e)}(\omega)f_1(\omega R/c)\right. \\ \left.-\mathrm{Im}\,\alpha_2{}^{(e)}(\omega)f_2(\omega R/c)\right]\left[\coth\frac{\hbar\omega}{2k_BT_1} - \coth\frac{\hbar\omega}{2k_BT_2}\right] + (e\to m) \qquad (4)$$

$$f_1(x) = 2(x^3 - 3x)\cos(2x) + (x^4 - 5x^2 + 3)\sin(2x) \qquad (5)$$

$$f_2(x) = 2x^4 - 4x^2 + 6 - 2(x^4 - 5x^3 + 3)\cos\,(x)^2 + 2(x^3 - 3x)\sin(2x) \qquad (6)$$

Therefore, radiant emittance W of particle 1 is described by a sum of the first term in Eq. (2), involving direct RHT (referred to as $\dot{Q}^{(1)}$) between the particle and vacuum, and $\dot{Q}^{(2)}$ – indirect RHT due to the presence of the second particle (the second term in Eq. (2)). Assuming that $\dot{Q}^{(1)} < 0$, the sign of $\dot{Q}^{(2)}$ corresponds to the "mirror effect" ($\dot{Q}^{(2)} < 0$) or "screening effect" ($\dot{Q}^{(2)} > 0$) at a given R.

For an ensemble of N equidistant closely packed particles of the same radius r and temperature T_1, neglecting by the second and higher order correlations, as well as the surface effects, a total radiation power of the ensemble is given by

$$W_N \approx N\left(\dot{Q}^{(1)} + 12Q^{(2)}\right) \qquad (7)$$

In numerical calculations, it is convenient to introduce the blackness parameter $b = W/W_{SB}$ or $b_N = W_N/W_{SB}$, relating to a single particle or to an ensemble of N particles, where the black body radiant emittance W_{SB} is given by

$$W_{SB} = \frac{\pi^3}{15}\frac{k_B{}^3}{\hbar^3c^2}R_0{}^2T^4 \qquad (8)$$

with allowance for $R_0 = r$ in the former case and $R_0 = RN^{1/3}$, $R \ge 2r$ in the latter case.