Three-body radiation dynamics in systems with anisotropic nanoparticles

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Abstract – The time evolution of temperatures of anisotropic nanoparticles in two- and three-body systems are simulated for various relative orientations. Nanoparticles are immersed in a thermal bath at constant temperature. It is shown that in two-body systems, the relative orientation of nanoparticles could drastically affect the dynamics of temperature evolution and thermalization time scale. Moreover, in some configurations, the temperature difference in initial state has a major effect on the dynamics of temperatures. In three-body systems, the orientation of the third nanoparticle influences the temperature dynamics, which allows one to control the thermalization time scales between anisotropic nanoparticles. Also, in addition to the previously known contribution of the smallest distance between isotropic nanoparticles on the thermalization time scales, it is shown that the nanoparticles’ orientations are more important in some particular arrangements.

It is well known that the radiative heat transfer intensity can be sufficiently enhanced when the separation between objects is much smaller than the characteristic wavelength of thermal emission. This phenomenon is demonstrated experimentally for plane-plane and sphere-plane geometry and it is shown that the heat flow at nanoscale distances is greater than the black-body radiation law posed by Stefan Boltzmann [1–4]. Several theories have been proposed to explain the heat transfer between simple particle geometries at nanoscale distances and have been widely used for two-body systems including parallel slabs [5–7], a particle in front of a planar surface [2,8,9], two spheres [10–13], and two anisotropic objects [14–16]. There are series of theoretical studies in recent years concerning the many-body effects in the radiative heat transfer [17–22]. It is shown in many publications that the heat flux exchange between two objects can be remarkably enhanced in a three-body system with respect to a two-body system, thanks to many-body interactions. In ref. [22], Ben-Abdallah et al. have shown a configurational resonance in heat exchange between spherical nanoparticles due to many-body interactions. Similarly, the exaltation of the heat transfer in a three-body system of parallel slabs is reported in [23].

There are very few studies on the heat transfer between anisotropic materials. The effect of shape, orientation and polarizability tensor in the heat transfer between anisotropic objects allows for a large freedom of tunability. Huth et al. studied the shape dependence of heat transfer between a spheroidal nanoparticle and a planar surface for various distances [9]. Compared to the sphere-plane geometry, they have shown that it is possible to control the heat transport in the system by means of the shape. Biehs et al. showed that the heat exchange between two anisotropic media can be significantly larger than those traditionally measured between two isotropic materials [14]. The strong dependence of heat exchange between two anisotropic nanoparticles on their shapes and relative orientations has been recently reported in refs. [16,24] and it has been shown the heat exchange in a system of anisotropic nanoparticles can be tuned drastically depending on relative orientations and polarizability tensors.

Most of the theoretical efforts in the investigation of radiative heat transfer have been restricted to the steady-state regime. The issue of radiative thermalization and dynamics of temperatures has been addressed in few papers. Tschkin et al. have investigated the radiative cooling of a spherical nanoparticle close to an interface [25]. They have shown that the thermal relaxation time of the nanoparticle varies rapidly for distances smaller than the thermal wavelength and is very sensitive to the initial temperature differences. Dyakov et al.
studied the propagation of thermal radiation in a two-layered structure. In ref. [26], they have shown that the thermalization time scale between the plates strongly depends on the separation distance. The dynamics of heat transfer between isotropic nanoparticles is well treated by Messina and his co-workers with consideration of many-body effects [27]. Using a purely fluctuational electrodynamics approach, they studied the time evolution of temperatures of spherical nanoparticles with respect to several geometrical arrangements. It was suggested that many-body interactions might lead to rapid thermalization. They concluded that the smallest distance between nanoparticles in a system determines the time scale on which the heat flux is exchanged between the particles. 

In this letter, we study the dynamics of heat exchange between anisotropic nanoparticles and visualize the dependence of temperature dynamics on the nanoparticles relative orientation. To do this, we use the fluctuational electrodynamics theorem which relates the power spectral density of fluctuating charge density to the local temperature- and frequency-dependent relative dielectric response of nanoparticles [28]. Using this theorem and properties of the dyadic Green’s function as well as polarizability tensors, allows studying the phenomenon of heat transfer between anisotropic nanoparticles at different temperatures. For the sake of simplicity, we restricted ourselves to nanoparticles with spheroidal shape only, which are made of silicon carbide (SiC). We show that the relative orientation of nanoparticles could drastically affect the dynamics of temperature evolution and thermalization time scale. We also demonstrate that in some configurations, the temperature difference in initial state has a major effect on the dynamics of temperatures. Moreover, we show that in three-body systems, the orientation of the third nanoparticle influences the dynamics of temperatures in comparison to the two-body systems. 

Before we go through the dynamics of heat transfer between anisotropic particles, let us focus on the problem of radiative heat exchange in a system of anisotropic nanoparticles without considering the temporal evolution of temperatures. Consider an energy exchange problem between N anisotropic nanoparticles identified for convenience by indexes \( i = 1, 2, \cdots, N \) with positions \( \mathbf{r}_i \), polarizability tensors \( \hat{\alpha}_i \), and temperatures \( T_i \). In order to take into account the radiation impinging on such a system from the outside, we assumed that this collection is immersed in a thermal bath at a constant temperature \( T_b \). Based on the assumption that nanoparticles behave as fluctuating dipoles at different temperatures, the fluctuation-dissipation theorem provides the energy which is dissipated in each nanoparticle. The nanoparticles themselves interact through the radiation fields and on the other side with the thermal bath and the total radiation power dissipated in each particle is [24]

\[
\mathcal{P}_i = \mathcal{F}_i(T_i) + \sum_{j \neq i} \mathcal{F}_{i,j}(T_j) + \sum_{j,j'} \mathcal{F}_{i,j,j'}(T_b)
\]

with

\[
\mathcal{F}_i(T_i) = \text{Im} \int_0^\infty \frac{d\omega}{\pi} \text{Tr} \left[ \hat{A}_i \text{Im}(\hat{\chi}_i) \hat{C}_i^\dagger \Theta(\omega, T_i) \right],
\]

\[
\mathcal{F}_{i,j}(T_j) = \text{Im} \int_0^\infty \frac{d\omega}{\pi} \text{Tr} \left[ \hat{A}_{ij} \text{Im}(\hat{\chi}_j) \hat{C}_{ij}^\dagger \Theta(\omega, T_j) \right],
\]

\[
\mathcal{F}_{i,j,j'}(T_b) = \text{Im} \int_0^\infty \frac{d\omega}{\pi} \text{Tr} \left[ \hat{B}_{ij} \text{Im}(\hat{G}_{j,j'}) \hat{D}_{ijj'}^\dagger \Theta(\omega, T_b) \right],
\]

where \( \mathcal{F}_i(T_i) \) denotes the so-called self-emission by the \( i \)-th particle corresponding to the heat lost by its radiation in the presence of other particles in the system, \( \mathcal{F}_{i,j}(T_j) \) is the radiative heating of the \( i \)-th particle due to radiation of the \( j \)-th one which contains both the direct radiant energy and those that reflected back and forth between particles several times, and finally \( \mathcal{F}_{i,j,j'}(T_b) \) is a portion of power which is absorbed in the \( i \)-th particle due to the radiation of the thermal bath. In these equations, \( \Theta(\omega, T) = h \omega [1 + 2n(\omega, T)]/2 \) and \( n(\omega, T) \) is the Bose-Einstein energy distribution function and we have introduced \( \hat{\chi} = \hat{\alpha} + k^2 \hat{\alpha} \hat{G} \hat{\alpha} \). Here \( G_{ij} = G_{ji} \) are the electric dyadic Green functions, \( \hat{G}_0 = \frac{i}{\omega} \hat{1} \), and \( k = \omega/c \). The physical properties of nanoparticles and many-body interaction effects are contained in the coupling matrices \( \hat{A} \), \( \hat{B} \), \( \hat{C} \) and \( \hat{D} \). For the special case of three anisotropic nanoparticles, we have

\[
\hat{A} = \begin{bmatrix}
1 & -k^2 \hat{\alpha}_1 \hat{G}_{12} & -k^2 \hat{\alpha}_1 \hat{G}_{13} \\
-k^2 \hat{\alpha}_2 \hat{G}_{21} & 1 & -k^2 \hat{\alpha}_2 \hat{G}_{23} \\
-k^2 \hat{\alpha}_3 \hat{G}_{31} & -k^2 \hat{\alpha}_3 \hat{G}_{32} & 1
\end{bmatrix},
\]

\[
\hat{B} = k^2 \begin{bmatrix}
\hat{\alpha}_1 & 0 & 0 \\
0 & \hat{\alpha}_2 & 0 \\
0 & 0 & \hat{\alpha}_3
\end{bmatrix},
\]

\[
\hat{C} = k^2 \begin{bmatrix}
\hat{G}_0 & \hat{G}_{12} & \hat{G}_{13} \\
\hat{G}_{21} & \hat{G}_0 & \hat{G}_{23} \\
\hat{G}_{31} & \hat{G}_{32} & \hat{G}_0
\end{bmatrix} \hat{A},
\]

\[
\hat{D} = \hat{1} + \begin{bmatrix}
\hat{G}_0 & \hat{G}_{12} & \hat{G}_{13} \\
\hat{G}_{21} & \hat{G}_0 & \hat{G}_{23} \\
\hat{G}_{31} & \hat{G}_{32} & \hat{G}_0
\end{bmatrix} \hat{B}.
\]

The frequency-dependent relative dielectric permittivity of SiC is well described by the Lorentz model [29]:

\[
\varepsilon(\omega) = \varepsilon_\infty \left( 1 + \frac{\omega_L^2 - \omega_T^2}{\omega_L^2 - \omega^2 - i \Gamma \omega} \right)
\]

with \( \omega_L = 969 \text{ cm}^{-1} \), \( \omega_T = 733 \text{ cm}^{-1} \), \( \Gamma = 4.76 \text{ cm}^{-1} \) and \( \varepsilon_\infty = 6.7 \). This relative permittivity is used to calculate the dressed polarizability tensors of nanoparticles. In the case of small isotropic non-spherical particles, the dressed polarizability tensor can be expressed as [30]

\[
\hat{\alpha}_i = \hat{\alpha}_b \left( \hat{1} - i \frac{k^3}{6\pi} \hat{\alpha}_b \right)^{-1},
\]
with
\[
\dot{\alpha}_b = v_i (\hat{\epsilon} - 1) \left\{ 1 + (\hat{L} - k^2 \hat{M})(\hat{\epsilon} - 1) \right\}^{-1},
\]
where \( \hat{L} \) and \( \hat{M} \) are the singular and non-singular part of the depolarization dyadic [31]. Let us stress that the polarizability tensor equation (5) is consistent with energy conservation (or, equivalently, the optical theorem). For the special case of small prolate spheroidal nanoparticles, the components of the polarizability tensor along and across the rotation axis of a spheroidal nanoparticle are given by
\[
\dot{\alpha}_{b,s} = \frac{4}{3} \pi R_b R_s^2 \frac{\epsilon(\omega) - 1}{1 + L_\beta \epsilon(\omega) - 1} \quad (\beta = b, s),
\]
where \( R_b \) and \( R_s \) are the major and minor axes of the spheroid and the depolarization factors take the form [32]:
\[
L_s = \frac{1}{2} (1 - L_b),
\]
\[
L_b = \frac{1 - e^2}{2e^3} \left[ \ln \left( \frac{1 + e}{1 - e} \right) - 2e \right],
\]
with \( e^2 = 1 - \frac{R_s^2}{R_b^2} \) for prolate spheroid \( (R_b > R_s) \). Moreover, we require that \( R_b \) be smaller than both the thermal wavelength and the intra-particle distances to substantiate the dipole approximation.

Now that we have established the formalism of heat exchange between anisotropic nanoparticles, we can present a framework for dynamics of temperatures. The time evolution of the temperatures in system is a solution of the nonlinear coupled system of differential equation:
\[
\rho_i c_i \frac{\partial T_i}{\partial t} = P_i (T_1, T_2, \ldots, T_N, T_b) \quad (i = 1, 2, \ldots, N),
\]
where \( \rho_i \) is the heat density of SiC is calculated as follows [33]:
\[
c = 1.267 + 0.049 \times 10^{-3} T - 1.227 \times 10^5 T^{-2} + 0.205 \times 10^8 T^{-3}.
\]
We note that the right-hand side of eq. (9) has \( N \) nonlinear terms with a strong nonlinearity in temperatures. We need the initial values of temperatures as well as the bath temperature to determine the solution uniquely. The solutions of this equation could be visualized as trajectories flowing through an \( N \)-dimensional phase space with coordinates \((T_1, \ldots, T_N)\).

As a preliminary step we have considered the temperature evolution in a two-body system. The system consists of two prolate spheroidal nanoparticles \((R_b = 4R_s = 80 \text{ nm})\) with a fixed center-to-center separation, \( d = 800 \text{ nm} \). The particles are heated up to initial temperatures \( T_1(0) \) and \( T_2(0) \) and immersed in a thermal bath at fixed temperature \( T_b \). The nanoparticles themselves interact with each other and also with the thermal bath. Our aim is to calculate the particles temperatures, \( T_1 \) and \( T_2 \), as a function of time. So, eq. (1) is used to calculate the total power dissipated in each particle and the dynamics of temperatures are calculated according to the energy balance equation, eq. (9). Since the thermalization rate in each particle due to thermal conduction by heat carriers is much larger than the heat transfer rate between the particle and the rest of the system via radiation, it ensures that the energy fluxes supported by heat carrier inside each particle is large enough to treat the temperatures as homogeneous inside each particle during the dynamical evolution of the system.

In order to show the relative orientation effect on the dynamics of heat transfer, particles are oriented in the system with one of the main arrangements, namely the tip-tip, side-side, tip-side (side-tip), and cross configurations. Figure 1 shows solution to eq. (9) for these types of configurations. The initial temperatures are \( T_1 = 350 \text{ K} \) and \( T_2 = 300 \text{ K} \) and particles are separated \( d = 800 \text{ nm} \) apart in a thermal bath at \( T_b = 300 \text{ K} \). The fixed point of eq. (9) corresponds to the thermal equilibrium state of the system. In the presence of thermal bath (at constant temperature) and in the absence of any other external sources, the particles would thermalize to the temperature of the bath. On the other hand, there is a fixed point at the bath temperature, regardless of the nanoparticles characteristics or initial temperatures. We would expect no change in temperatures in thermal equilibrium and from fig. 1 it is clear that it takes approximately 0.1 second for particles to thermalize with the bath (i.e., \( T_{1,2} \rightarrow 300 \text{ K} \)) which is independent from the relative orientation of the nanoparticles. However, it can be seen that there exist two types of thermalization time scales in the curves: one corresponds to the thermalization between two particles.
(smaller time scale) and the other to the thermalization of particles with the bath (larger time scale). The former is mainly due to the intra-particle heat exchange while the latter is caused by the radiative heating of nanoparticles by the bath.

In the case of tip-tip configuration, the first thermalization is \( \sim 1 \) order of magnitude faster than the second thermalization. For the side-side configuration, the early thermalization time scale increased slightly and the first thermalization temperature tends to \( T_b \). This is because the coupling between particles in this configuration is small in comparison with the tip-tip case and the heat transported from the particle at larger temperature to the colder one could slightly compensate the decrease of the temperature of the colder one due to its thermal emission. As the relative orientation changes to cross or tip-side configurations, the heat exchange between particles drops several orders of magnitude [24] and it seems that particles are thermalize by the bath independently.

The phase portraits for different configurations of particles in the two-body system are shown in fig. 2. Each of these configurations has a stable fixed point at \( T^* = T_b \) (solid blue dot) and all trajectories approach it as \( t \to \infty \). However, the direction of approach depends on the initial temperature difference as well as the relative orientation of the particles. Moreover, the thickness of the trajectories shows the thermalization rate which is faster for large temperature differences and the phase point slows down as it reaches the proximity of the thermalization point. If we approach this point monotonically as \( t \to \infty \) then the trajectories approach the fixed point as \( t \to \infty \).

Let us first consider the thermal evolution problem in a system where particles 1 and 2 form a tip-tip configuration. Figures 3(a) and (b) represent the time evolution of the temperatures calculated for two particular arrangements, namely, tip-tip-tip and tip-side-tip configuration. In fig. 3(a), nanoparticles are aligned along the longer spheroid axis and the intra-particle couplings are very weak. Surprisingly, it can be seen that the presence of the third particle reduces the time scale for thermalization between 1 and 2 of about an order of magnitude in comparison with the two-body system. Moreover, the heat exchange between 1 and 3 is enough to let the third particle thermalize by the first one even slightly faster than the time scale of thermalization of 1 and 2. This is explained by the fact that the coupling between 1 and 3 is larger than the coupling between 1 and 2 due to separation distances.

The approach to the fixed point is tangent to the temperature. The trajectory reaches this line is the intra-particle thermalization regime, then slowly ooze along this curve as it reaches the proximity of the thermalization point. In the case of tip-tip configuration, as shown in fig. 2(a), all trajectories slam straight onto the slower direction (here, the \( T_1 = T_2 \) direction) which corresponds to intra-particle thermalization regime, then slowly ooze along this curve until they reach a fixed point. The point at which the trajectory reaches this line is the intra-particle thermalization temperature. The approach to the fixed point is tangent to the \( T_1 = T_2 \) direction, along which the thermalization by bath occurs. On the other hand, if we look backwards along a trajectory \( (t \to -\infty) \), then the trajectories all become parallel to the faster decaying direction (here, the \( T_1 + T_2 - 2T_b = 0 \) direction). As the relative orientation changes to side-side configuration, the coupling between nanoparticles decreases an order of magnitude in comparison to tip-tip configuration [24]. As a result, the time scale for thermalization of the particles with each other increases and the decay to the slower direction decreased. The trajectories show faster decay along the \( T_1 = T_2 \) direction than the previous configuration and approaches the fixed point monotonically as \( t \to \infty \) (see fig. 2(b)).

Figs. 2(c) and (d) represent the time evolution of the temperatures calculated for two particular arrangements, namely, cross and tip-side-tip configuration. The thickness of the trajectories shows the thermalization rate.

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Fig. 3: (Colour on-line) Dynamics of temperatures in a three-body system. Particles 1 and 2 are oriented in a tip-tip configuration and a third particle is placed between them to form a (a) tip-tip-tip and (b) tip-side-tip configuration. The separation between 1 and 2 is \( d = 800 \text{ nm} \) as in the two-body case and the initial temperatures are \( T_1 = 350 \text{ K} \), \( T_2 = 300 \text{ K} \) and \( T_3 = 300 \text{ K} \), respectively. The collection is immersed in a thermal bath at \( T_b = 300 \text{ K} \). The red (solid) line corresponds to the hotter particle (the first particle), the blue (dashed) line is for the cold one (the second particle) and the green (long-dashed) line is for the third particle.

After such an intra-particle thermalization, it takes a long time for the phase point \((T_1 = T_2 = T_3)\) to pass through such a bottleneck state before making a transition to the final thermalization state of the system. Since the coupling with the thermal bath does not change significantly by the presence of the third particle, the time it takes for the system to reach the steady state is of the same order in the two-body system. As the third particle rotates to form a tip-side-tip configuration, fig. 3(b), the coupling between 1 and 2 drops an order of magnitude in comparison with the previous case [24]. The tip-side coupling between 3 and 1 (or 2) is not large enough and it has a trivial role in the dynamics of temperatures of particle 1 and 2. As a result, the temperature evolutions of particles 1 and 2 are similar to the two-body case (see fig. 1(b)). Moreover, particle 3 is not affected by particle 1 as much as particle 2, even though it is closer to it and has the same initial temperature as particle 2. Basically, this corresponds to the nature of couplings between anisotropic nanoparticles. On the other hand, while the smallest distances between particles determines the thermalization time scales in systems with isotropic particles [27], the orientations (or more generally, the polarizability tensors) play a crucial role when we are dealing with anisotropic particles.

There exist three particular orientations in adding a third particle to a two-body system with side-side configuration. Figure 4(a) shows the temperature evolution for side-side-side configuration. Since the relative orientation of each couple of particles in the system are identical (side-side form), the intra-particle distances determines the time scales on which the heat flux is exchanged between the particles. In this case, particle 3 is closer to particle 1 and as a result it is heated faster than particle 2. Once again, the shift in the time scales can be attributed to the presence of the third particle which acts as a bridge for indirect heat transfer between particles 1 to 2. The results of the temperature evolution of particles for other orientation of the third particle are shown in figs. 4(b) and (c), for side-tip-side and cross-cross configurations, respectively. The orientation of the third particle does not affect the intra-particle coupling between particles 1 and 2 significantly. Moreover, particle 3 is heated slightly more by particle 1 in the cross-cross configuration before the final thermalization with the thermal bath.

Finally, we consider a system in which particles 1 and 2 are arranged in a tip-side configuration. By adding a third particle, with one of the three mentioned orientations, three possible configurations are formed (see figs. 5(a)–(c)). It can be seen in these figures that the...
temperature evolution of particle 2 is not influenced by the presence of particle 3. However the intra-particle thermalization between particles 1 and 3 is very sensitive to the orientation of the third particle and specially pronounced in a tip-tip-side configuration shown in fig. 5(b). On the other side, the difference in the thermal dynamics of particle 3 in figs. 5(a) and (c) is mainly related to the many-body effect, that is the energy transferred from particle 1 to 3 through scattering from particle 2. The more interesting case is a configuration in which the third particle rotates by an angle of 45 degrees in the plane of particles 1 and 2, namely the tip-oblique-side configuration as shown in fig. 5(d). Upon inspection of low coupling between particles 1 and 2 observed in previous configurations, it can be seen that the temperature evolution is largely decided by three-body interactions. Here, the third particle appears to remove energy from the first particle and shares almost all of it with the second one.

In conclusion, we have investigated the dynamics of temperatures in two- (and three-)body systems of anisotropic nanoparticles. Depending on the relative orientation of nanoparticles and initial temperatures, the dynamics of heat transfer presents several interesting degrees of freedom. Although we have considered the heat transfer dynamics for spheroidal particles, our study reveals cryptic aspects of the dynamics of heat transfer between arbitrary shaped (and/or polarizability tensor) objects. We believe that our findings are essential for understanding the heat transfer in anisotropic systems, which is of interest for controlling the thermalization time scales and the design of new materials capable of tailoring heat propagation in nanoscale devices.

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