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Effect of plasmonic coupling on photothermal behavior of random nanoparticles

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ABSTRACT

In this paper, the effect of plasmonic coupling on the photothermal behavior of a random distribution of silver nanoparticles is investigated. The spatial profiles of the temperature increase for illuminated nanoparticles have been computed by means of discrete dipole approximation method and thermal Green's function. Our results show that in a random assembly of nanoparticles the effects of plasmonic coupling with other nanoparticles and thermal accumulation lead to a photothermal behavior for nanoparticles which is different from the one with an isolated single nanoparticle. The separate contributions of plasmonic coupling and thermal accumulation effects to temperature increase of nanoparticles assembly have been determined qualitatively. Based on obtained results, for wavelengths far from the plasmonic resonance of a single nanoparticle, the plasmonic coupling between clustered nanoparticles can heat up nanoparticles to significant high temperature which it cannot be expected for the case where the plasmonic coupling is assumed to be ignored (where nanoparticles only can interact due to the thermal accumulation). On the other hand, at the plasmonic resonance wavelength of a single nanoparticle, plasmonic coupling between clustered NPs causes the temperature increase to be lower in comparison with the case which the clustered nanoparticles are assumed as an assembly of individual non-coupled nanoparticles. Our results help to have a better understanding of the physics of photo heating of random nanoparticles in biological applications.

1. Introduction

A noble metal nanoparticle (NP) under illumination at its plasmonic resonance, which can be tuned from the visible to the infrared frequency ranges, strongly absorbs the light energy. The absorbed energy is converted to heat which raises the temperature of nanoparticle and its immediate surrounding media [1–5].

For a long time, heat generation in metallic NPs, induced by light absorption, has been considered as the side effects in plasmonics applications that had to be minimized. Recently, metallic NPs have been used as nanoscale sources of heat which it emerges a new promising field that could be named thermo-plasmonics [1,6–8].

In recent years, thermo-plasmonics found a wide range of applications in nanotechnologies especially in biology and medicine such as photothermal cancer therapy [9,10], drug delivery [11], nanosurgery [12,13] and photothermal imaging [14].

Almost in all the applications, we encounter with a random distribution of many NPs. In the case of low concentration, where NPs are far enough from each other and the interaction between them can be ignored, the photothermal behavior of the distribution can be considered as the summation of the response of individual non-interacting NPs [15].

In general, when the NPs interact, the photothermal behavior of each NP in the distribution can be very different in comparison with a single isolated NP. In fact, in an assembly of NPs, two different effects determine the photothermal properties of each NP; the effect of plasmonic coupling with the other NPs and thermal accumulation effect.

While extensive studies made on plasmonic coupling for a pair of nanoparticles [16–22], there is much less effort being expended on the ensemble of random NPs.

The important question that arises about the effect of plasmonic coupling on the photothermal behavior of random NPs is that will plasmonic coupling work in favor of the application, because of the field enhancement? Or will it work against it, because it shifts the plasmonic resonance wavelength?

The main concern of this paper is to find the answer of this question. In this paper, we use the discrete dipole approximation (DDA) method and thermal Green's function [7,8] to investigate the effects of plasmonic coupling and thermal superposition on temperature increase of randomly distributed nanoparticles. To the best of our knowledge, this is

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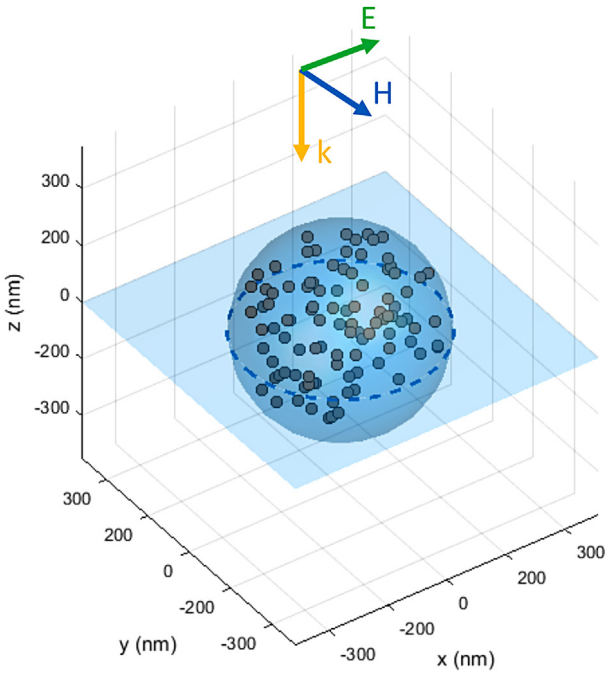

Fig. 1. Schematic drawing of the structure under study, where silver nanoparticles have been distributed randomly inside a spherical region in water and a $\hat{x}$-polarized light illuminates them.

the first study that determines quantitatively the separate contributions of each of these two effects to the photothermal behavior of illuminated NPs assembly.

Here, we have considered silver NPs because silver NPs, due to their stronger and sharper plasmon resonance, recently have been intensively used in diagnosis and the treatment of cancer and as the drug carriers [4,5,23–27]. As an experimentally relevant example of the structure that we have investigated in this paper, one can consider the application of NPs as nano-source of heat in aqueous medium such as in medicine and biology.

2. Theoretical model

Fig. 1, shows the sketch of the considered structure in this paper. We have considered $N = 1000$ silver spherical nanoparticles with diameter of $d = 10$ nm, distributed randomly inside a spherical region with the radius of $R = 250$ nm. Nanoparticles have been surrounded by a homogeneous dielectric medium. The surrounding medium is considered as water with the permittivity of $\varepsilon_m = 1.77$ and thermal conductivity of $\kappa_m = 0.6\ \mathrm{Wm^{-1}K^{-1}}$. A linear $\hat{x}$-polarized monochromatic light with the intensity of $I_0 = 0.5\ \mathrm{mW/\mu m^2}$ illuminates the particles in $-\hat{z}$ direction.

Investigating the thermo-plasmonic behavior of metallic nanostructures necessitates numerical approaches coupling optics and thermodynamics. The boundary element method (BEM) [28,29], discrete dipole approximation (DDA) and Green dyadic tensor (GDT) [30–33] are the methods that have recently been extended to compute the steady-state temperature in metallic nanostructures under continuous wave illumination [1,8,34].

Here, to compute the light scattering and absorption by nanoparticles, we have used the DDA method and to compute the spatial distribution of temperature in steady state regime, the thermal Green's function method has been used.

2.1. Discrete Dipole Approximation (DDA) method

The DDA is a suitable method to investigate the optical properties of illuminated NPs. In this method, an assembly of N identical sub wavelength-sized spherical NPs are considered. Nanoparticles have been distributed in a homogeneous medium with permittivity of ε_m.

When NPs illuminated by the incident monochromatic electric field, $\boldsymbol{E}^{inc}(\boldsymbol{r}) = \boldsymbol{E}_0 \exp(-i\omega t + i\boldsymbol{k}.\boldsymbol{r})$, the dipole moment of ith nanoparticle located at $\boldsymbol{r}_i$ is,

$$\boldsymbol{P}_i = \varepsilon_0 \varepsilon_m \alpha_i(\omega)\, \boldsymbol{E}_i^{ext} \tag{1}$$

where $\alpha_i(\omega)$ is the polarizability of the ith particle and $\boldsymbol{E}_i^{ext} = \boldsymbol{E}^{ext}(\boldsymbol{r}_i)$, is the external electric field amplitude experienced by the ith particle which has two origins: the incident field $\boldsymbol{E}^{inc}(\boldsymbol{r}_i)$ and the field scattered by the $N-1$ neighbor particles.

$$\boldsymbol{E}_i^{ext} = \boldsymbol{E}_i^{inc} + \frac{k_0^2}{\varepsilon_0} \sum_{j \neq i}^{N} \boldsymbol{G}_{ij} \cdot \boldsymbol{P}_j. \tag{2}$$

Here $\boldsymbol{E}_i^{inc} = \boldsymbol{E}^{inc}(\boldsymbol{r}_i)$ and $\boldsymbol{G}_{ij} = \boldsymbol{G}(r_i, r_j)$ is the electric Green's tensor defined as [35]:

$$\boldsymbol{G}(\boldsymbol{r}_i, \boldsymbol{r}_j) = \frac{1}{4\pi} \left(1 + \frac{\nabla\nabla}{k^2}\right) \frac{e^{ik|\boldsymbol{r}_i - \boldsymbol{r}_j|}}{|\boldsymbol{r}_i - \boldsymbol{r}_j|}. \tag{3}$$

In the dipolar approximation, the polarizability $\alpha_i(\omega)$ of the particle reads [8],

$$\alpha_i(\omega) = \frac{\alpha_0(\omega)}{1 - (2/3)\, ik^3 \alpha_0(\omega)} \tag{4}$$

where $\alpha_0(\omega)$ is the standard Clausius–Mossotti polarizability,

$$\alpha_0(\omega) = 4\pi a^3 \frac{\varepsilon(\omega) - \varepsilon_m}{\varepsilon(\omega) + 2\varepsilon_m}. \tag{5}$$

Here a and $\varepsilon(\omega)$ are the radius and electric permittivity of the nanoparticles, respectively. By substituting $\boldsymbol{P}_j$ from Eq. (1) into Eq. (2), we can obtain

$$\boldsymbol{E}_i^{inc} = \sum_{j=1}^{N} \boldsymbol{A}_{ij} \cdot \boldsymbol{E}_j^{ext}. \tag{6}$$

In Eq. (6), $\boldsymbol{A}_{ii} = \boldsymbol{I}$ ($\boldsymbol{I}$ is a 3×3 unitary matrix) and $\boldsymbol{A}_{ij}$ is defined as:

$$\boldsymbol{A}_{ij} = -k_0^2 \varepsilon_m \alpha_i \boldsymbol{G}_{ij}, \quad (j \neq i). \tag{7}$$

For a set of N nanoparticles that their positions are defined by $\boldsymbol{r}_i$, the N values of $\boldsymbol{E}_i^{inc}$ can be obtained directly. By inserting $\boldsymbol{E}_i^{inc}$ into Eq. (6) and solving the system of $3N$ linear equations numerically, the external electric field amplitude, $\boldsymbol{E}_i^{ext}$, at each particle position $\boldsymbol{r}_i$, can be calculated.

2.2. Thermal Green's function method

A direct consequence of light absorption by a metallic NP is the temperature increase of NP and its immediate medium because of heat diffusion. The heat power Q_i absorbed in ith NP is,

$$Q_i = \frac{1}{2} \sigma_{abs} nc\varepsilon_0 |\boldsymbol{E}_i^{ext}|^2 \tag{8}$$

where, σ_{abs} is the light absorption cross section of the particle and $n = \sqrt{\varepsilon_m}$ is the refractive index of the surrounding medium. The electric field distribution, $\boldsymbol{E}_i^{ext}$, can be obtained by means of DDA method and the absorption cross section can be obtained from $\alpha(\omega)$, [1].

$$\sigma_{abs} = k\mathrm{Im}(\alpha) - \frac{k^4}{6\pi} |\alpha|^2. \tag{9}$$

In a steady-state regime, the temperature profile, $T(\boldsymbol{r})$, throughout the system is solution of the Poisson's equation [7,8],

$$\nabla \cdot (\kappa(\boldsymbol{r}) \nabla T(\boldsymbol{r})) = -q(\boldsymbol{r}) \tag{10}$$