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Accurate thermoplasmonic simulation of metallic nanoparticles

Da-Miao Yu, Yan-Nan Liu, Fa-Lin Tian, Xiao-Min Pan*, Xin-Qing Sheng

The Center for Electromagnetic Simulation, School of Information and Electronics, Beijing Institute of Technology, 100081 Beijing, PR China

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ABSTRACT

Thermoplasmonics leads to enhanced heat generation due to the localized surface plasmon resonances. The measurement of heat generation is fundamentally a complicated task, which necessitates the development of theoretical simulation techniques. In this paper, an efficient and accurate numerical scheme is proposed for applications with complex metallic nanostructures. Light absorption and temperature increase are, respectively, obtained by solving the volume integral equation (VIE) and the steady-state heat diffusion equation through the method of moments (MoM). Previously, methods based on surface integral equations (SIEs) were utilized to obtain light absorption. However, computing light absorption from the equivalent current is as expensive as $O(N_sN_v)$, where N_s and N_v, respectively, denote the number of surface and volumetric unknowns. Our approach reduces the cost to $O(N_v)$ by using VIE. The accuracy, efficiency and capability of the proposed scheme are validated by multiple simulations. The simulations show that our proposed method is more efficient than the approach based on SIEs under comparable accuracy, especially for the case where many incidents are of interest. The simulations also indicate that the temperature profile can be tuned by several factors, such as the geometry configuration of array, beam direction, and light wavelength.

1. Introduction

Heat caused by light illumination on metallic nanostructures can be greatly enhanced by thermoplasmonics. Numerous efforts have been recently dedicated to the development of experimental and numerical methods to reveal the underlying physics and improve application designs [1–13]. Accurate temperature control is important in many applications, e.g., photothermal therapy [14–17], photothermal drug delivery [18–20], imaging and microscopy [21–24], hot printing [25] and nanoantennas design [26–28]. However, probing temperature at the nanoscale is fundamentally a complicated task [1]. This highlights the requirement for an accurate and efficient prediction of the temperature increase. To theoretically study the thermoplasmonics of nanostructures, numerical frameworks coupling electromagnetic waves must be combined with that of thermodynamics because light absorption and heat dissipation are competing mechanisms that govern the actual temperature distribution in metallic nanostructures. Light absorption relates to the solution of Maxwell's equations and heat dissipation to that of the Poisson equation if we restrict the heat study to the steady-state regime.

Generally, the capability of the simulation is limited by the cost of calculating light absorption, $b(\mathbf{r})$. Before computing for $b(\mathbf{r})$, Maxwell's equations should be solved first. Available simulation techniques include Finite-Difference Time-Domain (FDTD) [29,30], Finite-Difference Frequency-Domain (FDFD) [31], and the methods based on integral equations (IEs) [32–38]. The most popular and efficient approaches were critically compared in [37] with respect

* Corresponding author.
E-mail address: xmpan@bit.edu.cn (X.-M. Pan).

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to their capabilities for practical cases. It has been shown that methods based on surface IEs (SIEs) – named after boundary element method (BEM) in many studies – are superior to those based on volume IEs (VIEs) in terms of efficiency. However, in this work, the volume integral equation (VIE) is still chosen to solve Maxwell's equations for complicated and relatively large-scale applications. In contrast to the widely used discrete dipole approximation (DDA) and its efficient variations [37,39–41], the method of moments (MoM) is used to discretize the VIE. The resultant approach is termed MoM-VIE for conciseness.

The reasons for our choice are as follows. Firstly, the computational cost of $b(\mathbf{r})$ is essentially dominated by the evaluation of electric fields. MoM-VIE takes the field (or flux) as unknowns. Consequently, it can provide the required fields directly after the unknowns are solved. By contrast, SIEs use surface equivalent current as unknowns. An expensive integration operation has to be utilized to compute the fields from the equivalent current. The comparison in [37] was conducted with respect to the solution of Maxwell's equations, which excludes the computation of $b(\mathbf{r})$ from the solved unknowns. Actually, the latter can be a bottleneck in SIEs. Let N_s and N_v, respectively, denote the number of unknowns in an SIE solution and that in a VIE case for the same (homogeneous dielectric) structure; the cost is about $O(N_sN_v)$ in the SIE case because that for a given point is $O(N_s)$. Here, we utilize the fact that the number of sampling points should be proportional to N_v to achieve an acceptable spatial resolution of $b(\mathbf{r})$. In contrast, the cost is only $O(N_v)$ in the VIE case under a comparable accuracy. Secondly, because N_s or N_v is generally large, an iterative solver is always used. According to the study in [42], the PMCHW (Poggio, Miller, Chang, Harrington, Wu) formulation is the most accurate and reliable one among all the commonly used SIEs in plasmonic simulations. However, the associated approach often suffers from the slow convergence of the iterative solution [42]. On the contrary, our approach, MoM-VIE, always converges very fast. Thirdly, some other factors should be taken into account for the efficiency issue. For example, the solution of MoM-VIE and that of SIEs can be both significantly accelerated by using the fast multipole algorithm (FMA) [43,**?**]. The matrix–vector multiplication in MoM-VIE is about $\frac{N_v \log N_v}{N_s \log N_s}$ times slower than that in an SIE solver for the same problem with the FMA acceleration. Typically, $N_s \approx V^{2/3}$ and $N_v \approx V$, where V denotes the volume of the particle [37]. As a consequence, $\frac{N_v \log N_v}{N_s \log N_s} = 1.5V^{1/3}$. Theoretically, the larger the surface–volume ratio (which can be indicated by N_s/N_v), the stronger the light absorption could be. In turn, the larger temperature increase can be obtained. The representative applications are often with a large N_s/N_v. MoM-VIE takes the additional advantage for such cases. The last, but not the least, VIEs are inherently capable of heterogeneous dielectric structures [43] in contrast to SIEs [**?**,45–47]. To achieve a more visible view, we summarize the comparison of SIEs and VIEs in Table 1. It should be noted that, for particle ensembles, the cost of elevating $b(\mathbf{r})$ in SIEs is less than $O(N_sN_v)$, because $b(\mathbf{r})$ within a given particle can be computed from the sources on the particle's boundary. As discussed in several studies [48–52], fluctuating current sources due to thermal agitation will influence the spatial distribution of the heat source density when particles/structures are closely positioned. In this work, these stochastic thermally fluctuating current sources are not taken into consideration.

In the following section, we briefly describe the theoretical background, including the Poisson equation and the employed VIE. The discussion is then presented on the MoM discretization of the two types of equations. Simulation results are discussed next to validate the proposed scheme, including the studies on accuracy, efficiency and capability. The controllability of the thermoplasmonics is discussed along with the simulation study.

2. Theoretical background

Upon light illumination, electronic resonances known as localized surface plasmon (LSP) resonances can be excited for metallic nanostructures. The associated heat generation is governed by the light absorption (or Joule heating) with the heat source density (in W/m^3):

$$b(\mathbf{r}) = \frac{\omega}{2}\varepsilon_0 \mathrm{Im}(\varepsilon_r(\mathbf{r}))|\mathbf{E}(\mathbf{r})|^2, \tag{1}$$

where $\mathbf{r}$ is the position vector in terms of a prescribed coordinate system, ω is the frequency of the incident light, ε_0 is the permittivity of the background space and Im $(\varepsilon_r(\mathbf{r}))$ is the imaginary part of the relative permittivity of the nanostructure.

2.1. Steady-state heat equation

For the continuous-wave (CW) illumination, the heat dissipation resulting from light absorption is depicted by the heat diffusion equation in the steady-state regime. If we suppose that the nanostructure in question is embedded in an infinite homogeneous background space, then the pertinent equation is the Poisson equation of the form [1],

$$\nabla \cdot [\kappa(\mathbf{r})\nabla T(\mathbf{r})] = -b(\mathbf{r}), \tag{2}$$

where $T(\mathbf{r})$ is the temperature at point $\mathbf{r}$ and $\kappa(\mathbf{r})$ is the thermal conductivity. To simplify the problem, we assume that the nanostructure is thermally homogeneous. At the material interfaces, the boundary condition should be satisfied, which, in our applications, corresponds to the continuity of temperature T and that of its normal flux $\frac{\partial T}{\partial n}$. More precisely, the boundary condition is of the form

$$\begin{cases} T^+ = T^- \\ \kappa_e \dfrac{\partial T^+}{\partial n} = \kappa_i \dfrac{\partial T^-}{\partial n} \end{cases}, \tag{3}$$

where κ_e denotes the thermal conductivity of the exterior background space and κ_i denotes that of the nanostructure; $T^{\pm} = T(\mathbf{r})\big|_{\mathbf{r} \in \Gamma^{\pm}}$, where Γ^+ is the exterior boundary surface and Γ^- is the interior one. The combination of the Poisson equation (2) and the boundary