Study of thermoplasmonic properties of gold nanodimer in visible-infrared region of electromagnetic spectrum

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Abstract
In the present study, the heat generation in gold nanodimer when irradiated at their localized surface plasmon resonances is investigated numerically. The theoretical calculations are performed employing the first principal approach to obtain the absorption cross-section of gold nanodimer for different parameter ranges. The heating mechanism is enumerated in terms of its temperature by solving the steady-state heat transfer equation which depends on the absorption cross-section and surface plasmon resonance wavelength. These surface plasmon resonances are quite sensitive to the distance between the dimer and have been tuned from visible to IR range by managing the distance between spheres from 0 to 6 nm. The computation of normalized electric field distribution of gold nanodimer under the plasmon resonance has been mapped using boundary element method (BEM) which enables visualization of the local hot spot that plays a significant role in optical heating applications. The work furnishes the basic understanding of the heating mechanism of gold nanodimer which can find application as plasmonic nanoheaters in several branches of science in visible and near-infrared regions of the electromagnetic spectrum.

1. Introduction
Recently, it has been observed in several studies that metallic nanoparticles (NPs) which support surface plasmon resonances exhibit vast potential in optical heating applications. The heat generation via metal nanostructures induced by light absorption has long been considered as a negative effect that needs to be eliminated [1–6]. To change this notion in the context of metal, several efforts have been carried out in recent years which demonstrated that metal nanostructure can be used as a nanoheaters and opens up new avenues in the field of nanotechnology research [7–10]. This new field, which encompasses studying the combined effect of nano-optics and thermodynamics has been termed thermoplasmonics [11–13]. Several potential applications of the phenomenon of thermoplasmonics in nanotechnology have been highlighted [14–16]. These include diverse fields like nanofluids, phononics, photothermal cancer therapy, and imaging [17–19]. In photothermal tumor therapy, the laser impinges on the metallic nanoparticles which produce heat that stops the cellular activity either by shrinking the size of the tumor or decreasing the spread of cancerous cells [20–22]. The increase in temperature of metallic nanoparticles is caused by the absorption of light due to local surface plasmon resonance (SPR) which produces a large electromagnetic field at the vicinity of the nanoparticle surface. The physics of surface plasmon resonances and their tunability in a broader range of electromagnetic spectrum finds applications in almost every field of science and technology wherein light–matter interactions are observed. Further, several efforts have been attempted by the nanotechnology research community to optimize the morphology of a plasmonic nanoparticle and its SPR resonance for various applications. On the other hand, several researchers have also been carried out to elucidate the thermo-optics phenomena of metallic nanoparticles for complex nanogeometries [23–28].

But little attention has been paid in the literature to theoretically explain the local heating mechanism in presence of plasmonic nanogeometries. In all such works, it was reported that the surface plasmon resonances...
can be tuned in a different part of the electromagnetic spectrum corresponding to different shapes like nanostars, Nanotriangles, spheres, etc which act as nano heat sources [19, 29, 30]. Baffou, Oliveros et al had reported the thermoplasmonic properties and tunability of surface plasmon resonances in the visible range (400–800 nm) of the electromagnetic spectrum by using different shapes of nanoparticles [31, 32]. However, no systematic theoretical/experimental study has been carried out that explains the thermo-optics and SPR tunability corresponding to one particular nanogeometry using the first principle approach. To address these lacunae, in the present paper we simulate the metal nanodimer to tune the SPR resonances from visible to IR region of the electromagnetic spectrum using a particular structure i.e., sphere nanodimer. The main thrust of this work is to reveal the extent of tunability of the surface plasmon resonances of spherical dimer and hence analyze the heating effect of dimer corresponding to each SPR wavelength in terms of field distribution.

In this study, we employ the electrostatic approach to calculate the absorption cross-section of the metal nanodimer and investigate the behavior of surface plasmon resonance in the visible and near-infrared regions of the electromagnetic spectrum. The heat generated by the metal nanodimer surrounded by water media depends on the quantity of light absorbed into it. When light is absorbed by the nanoparticle, it is heated up leading to the nanodimer behaving as a nano source of heat. The temperature profile of this nanodimer which acts as the nano heat source is obtained by solving the steady-state heat equation for the metal nanodimer. The absorption cross-section terms in the steady-state equations account for the role of the nanodimer [33–35].

All the parameters investigated in this study reveal that how to tune the surface plasmon resonances of nanodimer with dimer distance. This is aimed at targeting various electromagneto-thermo-optical applications [24–28]. The schematics of metallic nanodimer which is placed in water as shown in figure 1. In this schematics, gold spheres are electromagnetically coupled to form a nanodimer in two different situations whose dielectric constant is taken from the literature [36]. The coupling between spheres is measured by interaction parameter where L is the surface to the surface distance between spheres, the radius of each sphere and it is 2R when the radius of each sphere are equal. These parameters play a significant role to explore the thermoplasmonic properties like heat generation, temperature, and absorption cross-section.

2. Steady-state heat equation in presence of metal

To explore the temperature distribution of plasmonic nanogeometries surrounded by water, we solve the steady-state heat equation. Under steady-state conditions, the heat transport equation is equivalent to Poisson’s equation whose solution provides the temperature profile [17, 37, 38] corresponding to two different plasmonic nanogeometries. The heating effect of plasmonic nanogeometries and their influences in surrounding media are analyzed by solving the heat transfer equation using first principle. The heat transfer equation is expresses as

\[
\rho c \frac{\partial T}{\partial t} = \nabla \cdot (K \nabla T) + Q
\]

(1a)

where \( \rho \), \( c \) and \( K \) are mass density, specific heat and thermal conductivity of surrounding medium respectively and \( T \) is the temperature distribution produced by the heat source \( Q(r) \).

The general expression of temperature distribution throughout the particle are obtained by solving the steady-state heat transfer equation as

\[-K \nabla^2 T = Q \]

(1b)
Since we are calculating the expression for the temperature at arbitrary point ‘$r$’, we express our Laplacian operator in spherical polar coordinates. The radial part is expressed as

$$\nabla^2 T = \frac{\partial}{\partial r}\left(r^2 \frac{\partial T}{\partial r}\right) = Q$$

The solution of the above equation is obtained as

$$T(r) = \frac{Q}{4\pi Kr} = \frac{C_{abs} I}{4\pi Kr}$$

In the above equation, Q is a source term that arises due to the optical absorption which is the absorption of light by the nanoparticle. The calculation of optical absorption of light by the nanoparticle is expressed in terms of absorption cross-sections.

### 3. Absorption cross-section of gold nanostructure

When a metal nanoparticle is illuminated by light, a fraction of light gets scattered in the surroundings, while the other part gets absorbed and ultimately dissipated in form of heat. Since we are investigating the heating mechanism of metallic nanoparticles, our interest is centered around the absorption of light by the metal nanoparticle. The efficiency of light absorbed by the nanoparticle is expressed in terms of its optical cross-section [37]. The optical absorption cross-section of the metal sphere is given by

$$C_{abs}^{Sp} = k\text{Im}(|\alpha|) = k\text{Im}(4\pi a^4|\varepsilon_p - \varepsilon_m/\varepsilon_p + 2\varepsilon_m|)$$

while the absorption cross-section corresponding to metal nanodimer is expressed in the form.

$$C_{abs}^{Di} = k\text{Im}(\alpha_{Di})$$

$$= k\text{Im}\left[\left(\alpha_1 + \alpha_2 + \frac{\alpha_1\alpha_2}{2\pi l^3}\right)/(1 - 2\alpha_1\alpha_2/(2\pi l^3)^2)\right]$$

where $\varepsilon_m$, $\varepsilon_p$ are the dielectric constant of medium and particle $k = 2\pi/\lambda_0$ is the wave vector, $l = 2R + L = a_1 + a_2 + L$ is the distance between centers of two spheres, $a_1$, $a_2$ are the radii of each spheres in dimer when it is different, 2R when it is equal. The terms $\alpha_1$, $\alpha_2$ are the polarisability of each nanosphere which can be obtained by solving the Laplace equation $\nabla^2 \Phi = 0$ under suitable boundary condition. In order to obtain the polarisability expression, consider a sphere embedded in a uniform electric field $E_0$. The induced dipole moment per unit volume and the polarizability of the first sphere are expressed as $P = \varepsilon_0|\varepsilon_m|a_1E$ and $\alpha_1 = 4\pi a^4|\varepsilon_p - \varepsilon_m/\varepsilon_p + 2\varepsilon_m|$ respectively. The corresponding values for the second sphere are given as $p_2 = \alpha_2\varepsilon_0|\varepsilon_m|E$, while $\alpha_2$ is polarisability of sphere dimer. Mathematically, the plasmonic properties of metal nanostructures have been explored by Frohlich conditions that appear in the denominator part of polarizability expression given by equation (3). The Frohlich condition directly depends on the SPR resonances and their spectral width. This condition also establishes the relationship among the plasmon frequency, incident light frequency, and dielectric constant of metal both at the bulk and nano level. For the case of the sphere, one can obtain the Frohlich condition by taking the denominator of the equation (3) equals zero i.e. $|\varepsilon_p + 2\varepsilon_m| = 0$. The Frolich condition of the sphere is $|\varepsilon_p + 2\varepsilon_m| = 0$, and corresponding plasmon frequency is $\omega = \omega_p/\sqrt{3}$. This relation is for the simplest situation, in which the metal sphere is embedded in air. When metal is placed in a higher dielectric medium like water, the plasmon frequency is $\omega = \omega_p/\sqrt{7.48}$ which suggests that SPR resonances are red-shifted with higher index medium.

Using the above mentioned analogy, we derived the Frohlich condition for spherical dimer geometry which has not been attempted in the literatures. This explains the relationship among the plasmon frequency, the dielectric constant, and incident light frequency. The Frohlich condition for sphere dimer is obtained by taking the denominator part of equation (4) equals to zero as

$$|1 - 2\alpha_1\alpha_2/(2\pi l^3)^2| = 0$$

Now, substituting the vale of $\alpha_1$, $\alpha_2$ we have

$$|1 - 2(4\pi a^3)^2|\varepsilon_p - \varepsilon_m/\varepsilon_p + 2\varepsilon_m|/ (2\pi l^3)^2| = 0$$

$$|1 - 2(4\pi a^3)^2|\varepsilon_p - \varepsilon_m/\varepsilon_p + 2\varepsilon_m|/ (2\pi l^3)^2| = 0$$

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Equation (6) demonstrates how the value of the plasmon frequency is dependent on the dielectric constant of medium and incident light frequency. The dielectric constant of metal plays a major role in revealing the plasmonic signature and has different characteristics at bulk and nanolevel. At nanolevel, we modify the Drude model to include the contribution of the size effect [27, 37]. The size-dependent dielectric constant has been derived from the Drude-Lorentz Sommerfeld model and is expressed as.

\[
\varepsilon_p = \varepsilon_{\text{bulk}} + \frac{\omega_p^2}{(\omega^2 + j\Gamma_{\text{bulk}}\omega)} - \frac{\omega_p^2}{(\omega^2 - j\Gamma\omega)}
\]

(7)

where \(\omega_p = \sqrt{n^2 e^2/m}\) is plasmon frequency and the bulk damping constant \(\Gamma_{\text{bulk}} = v_F/r\). Equation (7) explains the dependence of the nanolevel dielectric constant of metal on the damping factor \(\Gamma = \Gamma_{\text{bulk}} + A - v_F/\lambda\). Where \(v_F\) is the Fermi velocity of electron and \(\lambda\) is the effective radius of sphere nanodimer \(A\) is the geometrical factor and its value is unity for isotropic scattering. Since the optical response of metals is highly dependant on their dielectric constant therefore we have plotted it at bulk and nanolevel to see their optical nature. Figure 2 shows the clear difference between the wavelength-dependent dielectric constant of gold metal at bulk and nanolevel which also explains that how nano effect comes into the picture after reducing the dimension of the chosen geometry.

Although we have a large number of studies that express the relation between plasmon frequency and bulk dielectric constant. There is no theoretical explanation available for dimer in the nanoplasmonic research community that elucidates the relationship among the plasmon frequency, bulk damping constant, bulk dielectric constant, and dielectric constant of metal at nanolevel and the surrounding medium in terms of the Frohlich explanation. Using Frohlich condition from equations (5), (7), we derive the new mathematical expression of plasmon frequency for sphere dimer geometry to explain its resonant physics. The new form of Frohlich expression in terms of the dielectric constant of metal at nanolevel is calculated as.

\[
\omega_p = \varepsilon_{\text{bulk}} \left(1 + 2 \frac{\eta_2}{\eta_1}\right) - \varepsilon_{\text{bulk}} \left(1 - \frac{\eta_2}{\eta_1}\right) \left(1 - \frac{\eta_2}{\eta_1}\right) \left(\frac{1}{\omega^2 + j\Gamma_{\text{bulk}}\omega} - \frac{1}{\omega^2 - j\Gamma\omega}\right)
\]

(8)

It is conspicuous that equation (8) establishes the relation between plasmon frequency of sphere dimer, the bulk dielectric constant of metal, and the dielectric constant of the surrounding medium. This equation can be used to explain the importance of SPR resonances and their dependency and tunability on the interaction length of the dimer.
The study of the role of plasmonic nanodimer and nanosphere spectra is crucial to gain insight into its tunability and optical heating applications. For this, we took gold metallic nanoparticles in two different shapes which are surrounded by a homogeneous medium of dielectric constant \(\varepsilon_m = 1.8\) (water) and illuminated by a plane wave. This arrangement is envisioned to model situations where the metal nanoparticles are immersed in an aqueous medium (water), for example in biological applications. For instance, gold nanodimer are foreseen to be ideal candidates for photothermal cancer therapy because their resonance of the heat generation can easily be turned into the broader range of spectral window of human tissue ranging from 500 to 900 nm. There are two basic parameters on which the optical heating mechanism in metallic nanoparticles is dependant; one being the absorption cross-section and the other is the surface plasmon resonance. The mechanism of light absorption is explained in terms of absorption cross-section which is the cross-section under influence of incident light and always greater than the geometrical cross-section. On the other hand, the surface plasmon resonance plays an important role in determining the spectral width in which optical heating is dominant. The heating mechanism of gold metal in two different shapes, namely sphere and sphere dimer is mainly dependant on the absorption of light by nanoparticles under the SPR condition as indicated in equation (2).

Figure 3 shows the wavelength-dependent normalized absorption cross-section of the sphere and sphere-dimer. This leads to a two-fold discussion where we discuss the absorption cross-section of the sphere which is quite present in literature. Secondly, we discuss the characteristics of absorption spectra exhibited by the sphere nanodimer. To analyze the absorption cross-section & resonance spectra which is inline with the temperature expression. We have plotted the absorption cross-section profile of the 10 nm radius of the gold sphere in the water environment as shown in figure 3(a) where it was observed that the SPR wavelength corresponding to such wavelength is around 520 nm. Next, the plot of the absorption cross-section of 10 nm radius of sphere nanodimer whose separation distance \(L\) is different as shown in figure 3(b). It was seen that the SPR wavelength of the gold nanodimer can be tuned by managing the distance \(L\) between two spheres in dimer geometry. As the gap between the two spheres increases, correspondingly blue-shifted SPR wavelengths are observed which can be understood with the help of equation (8). Moreover, the simulated results corresponding to nanodimer covered the broader range of electromagnetic spectrum that can be helpful to target the various applications.

In figure 4, we have plotted the normalized surface electric field of the gold nanodimer under their SPR wavelength using the boundary element method based MNPBEM toolbox. The planer profile of the electric field is plotted with four different SPR wavelengths having the value of \(L = 0, 2, 4, 6\) nm as shown in the figure. From the field plot, it is inferred that the magnitude of the normalized electric field is directly proportional to the surface to surface distance between two spheres \(L\). The results demonstrate that large electric field enhancements were observed in the region between the nanospheres. The field enhancement between the gap of both nanospheres is caused by constructive interference wherein the amplitude of the electric field corresponding to both spheres superimpose in phase under SPR condition. These enhancements and tunability of the field corresponding to different values of \(L\) make the dimer a suitable candidate for heating applications at the nanoscale.

Further, we analyze the heating mechanism of the sphere dimer in terms of absorption cross-section by changing the radii of one sphere in the dimer geometry while keeping the radius of the other sphere as constant. Figure 5 shows the wavelength-dependent absorption cross-section of sphere dimer separated by length \(L = 4\) nm with one sphere radius \(a_1 = 10\) nm while the radius of the second sphere is discreetly varied as \(a_2 = 4,\)
6, 8, 10 nm. It from the result, it was observed that the absorption cross-section increases with the reduction in the radius of the second sphere. As enumerated in equation (2), this increase in the absorption cross-section of the sphere dimer spurs up the temperature and accelerates the heating mechanism of the sphere dimer. It is therefore apparent that the heating mechanism of the dimer is a function of several parameters including the material and its geometry, interaction distance between the spheres, and nature of the surrounding environment. As we change the radii of one sphere while keeping the radius of the second sphere is constant in dimer geometry, the magnitude of absorption cross-section increases. This enhancement in absorption cross-section will lead to increases in temperature near the metal surface that can be employed for several practical purposes. In all these cases, the maximum enhancement in temperature is observed at their SPR wavelength 540 nm but the off-resonant physics are also important which can be understood in terms of their spectral width. The width of the resonance spectra lies in the range of 500 to 600 nm which suggests that the temperature enhancement in this range is significant for optical heating.

4. Temperature relative factor

The temperature relative factor can be obtained by taking the ratio of temperature corresponding to sphere nanodimer and sphere which depends on their absorption cross-section ratio. The dependency of temperature on absorption cross-sections is explained in equation (2). We calculate the temperature relative factor for the nanosphere and sphere nanodimer using the following equation which states that its value depends solely on the
absorption cross-section of chosen nanogeometry.

\[ \tau(\omega, \omega) = \frac{T'}{T} = \frac{C_{\text{abs}}^{\text{D}}}{C_{\text{abs}}^{\text{Sp}}} \]  

The effective radius of nanosphere is constant but varies for sphere nanodimer whose radius depends on the center to center distance \( l = a_1 + a_2 + L = 2R + L \) between two spheres. Table 1 shows the relative temperature of nanodimer under study, compared to that of nanospheres with the same effective radii at their SPR wavelengths. It is observed that gold nanodimer with different interaction lengths yields temperature higher than the corresponding gold nanosphere.

These results corresponding to four different interaction lengths \( L \) allow us to infer that gold nanodimers can act as heat sources and find applications in several fields. These nanodimers produced heat more efficiently than the sphere based on its value of interaction length. Further, in all the cases of sphere nanodimer, the temperature relative factor \( \tau > 1 \). It was also observed that the temperature relative factor \( \tau \) increases with the decrease in distances between two spheres in dimer geometry. Therefore one can tune the heating efficiency of metal nanoparticles in the desired range of electromagnetic spectrum either by altering the coupling distance between spheres or the interaction length. The sweet spot in the parameter space in this work is the dimer geometry and its interaction length which is very important parameter to tune the surface plasmon resonance for various applications like cancer therapy, optical heating and Nanofluid preparation. This work suggest another sweet spot which can be analyses by choosing several nanogeometries (like dimer nanoellipsoid, dimer nanorod etc) to study their thermoplasmonics signature.

5. Conclusions

In conclusion, the efficacy of the gold nanodimer as a thermal heater based on their thermoplasmonics properties is established in the present study. The tunability of the heating properties based on values of the nanodimer interaction length and geometrical dimensions makes it an ideal candidate for a variety of optical heating applications. Several calculations show that a 52-fold increase in the steady-state temperature for nanodimer over the nanosphere can be achieved. Additionally, the red-shift induced on the SPR for decreasing value of \( L \) in nanodimer shows that a wide range of wavelength tunability in the visible and near-IR can be covered with various complex asymmetrical nano shapes like nanorods, nanocylinder, and nanostars. The simulated results confirm that a typical gold nanodimer can be easily fabricated with available current nanofabrication techniques and is suitable for electromagnetic thermo-optical applications. This theoretical work motivates the thermoplasmonic research community to think and study various sizes and shapes of nanodimers and corresponding thermo-optical parameters to target the different parts of the electromagnetic spectrum.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).
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