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Tuning near-field enhancements on an off-resonance nanorod dimer via temporally shaped femtosecond laser

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Abstract

We theoretically investigated ultrafast thermal dynamics tuning of near-field enhancements on an off-resonance gold nanorod dimer via temporally shaped femtosecond (fs) laser double pulses. The nonequilibrium thermal excitation is self-consistently coupled into a near-field scattering model for exploring the ultrafast near-field enhancement effects. It is revealed that the near electric-field localized within the gold nanorod dimer can be largely promoted via optimizing the temporal separation and the pulse energy ratio of temporally shaped femtosecond laser double pulses. The results are explained as thermal dynamics manipulation of plasmon resonances in the nanorod dimer via tailoring temporally shaped femtosecond laser. This study provides basic understanding for tuning near-field properties on poorly fabricated metallic nano-structures via temporally shaped femtosecond laser, which can find potential applications in the fields such as fs super-resolution near-field imaging, near-field optical tweezers, and fs photothermal therapy.

Keywords: thermal dynamics, near-field enhancement, nanorod dimer, temporally shaped laser

(Some figures may appear in colour only in the online journal)

1. Introduction

Giant electric field enhancement localized within the gap of nano-dimer antenna is important for both scientific and practical aspects [1–3]. The enhanced electric field associated with dimer plasmon has been demonstrated to depend, to a large extent, on the nano-dimer shape, and structure [4, 5]. However, the electric field enhancement for a metallic nano-dimer can be usually limited due to structure-mismatching in a poorly fabricated dimer, which severely degrades the performance of a perfect nano-dimer antenna. It becomes urgent to obtain a strong local field in mismatched nano-dimer to facilitate potential applications of the giant electric field enhancement in, e.g. super-resolution near-field imaging, near-field optical tweezers, and photo thermal therapy.

In recent years, femtosecond laser excitation of nano antenna has been attracting much interest for modifying near-field properties for a wide range of applications. It is believed that the plasmon process in a nano-dimer can be sharply modified due to ultrafast temperature-modulation of dielectric function via a femtosecond laser excitation [6]. Itina *et al* revealed that the electron thermal dynamics achieved via a train of shaped femtosecond pulses, namely multi-pulse sequence with variable seperation can be unprecedentedly adjusted with respect to the correlation of pulse-to-pulse energy coupling into target [7]. It is expected that the electron temperature of a metallic nano-dimer can be modulated via the temporally shaped femtosecond pulses. It can bring in essential advantages to the applications of fs near-field imaging, fs optical tweezers and fs photo-thermal therapy. For example, the near-field imaging with a localized nonlinear light source can be

realized via applying double fs laser irradiation of nanorod dimer. Through adjusting double pulse parameters of pulse separation and pulse energy ratio, the near-field intensity can be significantly enhanced for the given moderate laser fluence, leading to the high-nonlinear of the near-field light source for ultra-resolution imaging. The near-field optical tweezers can be usually applied for manipulation of the biology cells. It requires that the high-precision for manipulation of the target cells but keeps the smallest damage to the surrounding cells. As a temporally shaped femtosecond laser interacts with metallic nano-dimer, the electron system of the nano-dimer is initially excited by the pre-pulse, synchronously tuning the dielectric properties on the thermal excitation zone of nano-dimer. After that, the second pulse will interact with the pre-pulse thermal excitation zone, leading to the modifications of the localized electric-field on the excitation zones. It indicates that the spectral and spatial properties of localized near-field can be modulated due to thermal dynamics modulation with respect to pulse-to-pulse relevance. It enables the new opportunity for manipulating the near-field properties in an off-resonance nano-dimer antenna via tailoring temporally shaped femtosecond laser. Because of the complex dynamics with respect to temporally shaped femtosecond laser excitation [8–10], the ultrafast non-equilibrium thermal dynamics tuning of near-field enhancement via temporally shaped femtosecond laser is still an undeveloped topic. It becomes urgent to promote the plasmon resonance process in an off-resonance nano-dimer antenna to facilitate the potential applications, e.g. fs super-resolution near-field imaging, near-field optical tweezers, and fs photo thermal therapy.

In this paper, we theoretically investigated ultrafast thermal excitation dynamics for tuning of near-field enhancements on an off-resonance gold nanorod dimer antennas via temporally shaped femtosecond laser. The near electric-field enhancement spectra with respect to electron temperature modifications are acquired based on a proposed self-consistent model, in which the non-equilibrium thermal excitation dynamics is self-consistently coupled into a near-field scattering model for exploring the ultrafast near-field enhancement effects.

2. Modeling and methods

In metallic nanostructures the laser radiation is mostly absorbed by free electrons with minimal capacity for heat via Inverse Bremsstrahlung mechanism, rapidly attaining high temperatures, and transferring thermal energy to material lattices. These processes do not occur instantaneously: time must be allowed for the cooling of the electrons and the heating of the lattices. The electron cooling and lattice heating mechanisms have time delays on the order of femtoseconds and picoseconds, respectively. Ultrashort laser pulses end before the thermal energy transfer to the lattice is complete, requiring two-temperature models in order to describe the further conversion of energy from electron excitation to heat within the lattice system. For temporally shaped double fs pulses excitation of nanorod dimer, the first pulse gives rise to the nanorod dimer electron temperature, the second pulse then coupling into the excited electron of the nanorod dimer. As a result, the cooperative works of double fs pulse with different energy can plays an key role in determining the total heating property of the nanostructure. The electron and phonon temperature changes comply with the two-temperature model, which is widely accepted for investigations of ultrashort laser interaction with metals, dielectrics and nano-particles [11–15]. In this investigation, the electron ballistic transfer length (100 nm) for gold is larger than the cross section of the nanorod in light incident direction, so the electron heating in the nanorod dimer can be reasonably considered as a homogeneous heating process. The two-temperature model (TTM) for describing the homogeneous thermalization in a gold nanorod dimer can be written as

$$C_{\mathrm{e}}\frac{\partial T_{\mathrm{e}}}{\partial t} = \nabla\left(k_{\mathrm{e}}\nabla T_{\mathrm{e}}\right) - \gamma(T_{\mathrm{e}} - T_{\mathrm{l}}) + S(t) \tag{1}$$

$$C_{\mathrm{l}}\frac{\partial T_{\mathrm{l}}}{\partial t} = \gamma(T_{\mathrm{e}} - T_{\mathrm{l}}) \tag{2}$$

where C_{e} and C_{l} are the heat capacities of the electron and lattice subsystems, respectively. The heat capacity of electron can be written as the following piecewise function:

$$C_{\mathrm{e}}(T_{\mathrm{e}}) = \begin{cases} B_{\mathrm{e}}T_{\mathrm{e}}(T_{\mathrm{e}} < T_{\mathrm{F}}/\pi^2) \\ 2B_{\mathrm{e}}T_{\mathrm{e}}/3 + C_{\mathrm{e}}^{'}/3(T_{\mathrm{F}}/\pi^2 \leqslant T_{\mathrm{e}} < 3\ T_{\mathrm{F}}/\pi^2) \\ n_{\mathrm{e}}k_{\mathrm{B}} + C_{\mathrm{e}}^{'}/3(3\ T_{\mathrm{F}}/\pi^2 \leqslant T_{\mathrm{e}} < T_{\mathrm{F}}) \\ 3n_{\mathrm{e}}k_{\mathrm{B}}/2(T_{\mathrm{e}} \geqslant T_{\mathrm{F}}) \end{cases} \tag{3}$$

here

$$C_{\mathrm{e}}^{'} = B_{\mathrm{e}}T_{\mathrm{F}}/\pi^2 + \frac{3n_{\mathrm{e}}k_{\mathrm{B}}/2 - B_{\mathrm{e}}T_{\mathrm{F}}/\pi^2}{T_{\mathrm{F}} - T_{\mathrm{F}}/\pi^2}(T_{\mathrm{e}} - T_{\mathrm{F}}/\pi^2). \tag{4}$$

In fact, the electronic heat capacity is much smaller than the lattice heat capacity, therefore, the electron temperature of nano-rod dimer can be promoted to be as high as ~1 eV, but the lattice remains undisturbed during laser pulse duration on femtosecond timescale.

For a wide range of electron temperature ranging from 300 K to Femi temperature, the temperature-dependent electron heat conductivity during the two temperature relaxation processes is expressed as follows [16]:

$$k_{\mathrm{e}} = \chi\frac{(\theta_{\mathrm{e}}^2 + 0.16)^{5/4}(\theta_{\mathrm{e}}^2 + 0.44)\theta_{\mathrm{e}}}{(\theta_{\mathrm{e}}^2 + 0.092)^{1/2}(\theta_{\mathrm{e}}^2 + \eta\theta_{\mathrm{p}})}. \tag{5}$$

An analytical expression of the electron–phonon coupling strength was proposed by Zhang and Chen, which can be represented as follows [16]:

$$\gamma = \gamma_0\left[A_{\mathrm{e}}/B_{\mathrm{p}}(T_{\mathrm{e}} + T_{\mathrm{p}}) + 1\right] \tag{6}$$

where, γ_0 is the electron–phonon coupling strength in room temperature, and the coefficients A_{e} and B_{p} are constants.

The laser fluence absorption by the nano-dimer plasma via resistive heating is given by

$$S(t) = \frac{C_{\mathrm{abs}}(t)\cdot P_{\mathrm{SPT}}(t)}{V_{\mathrm{rod}}} \tag{7}$$

where $C_{abs}(t)$ is the absorbing cross section P_{SPT} is the laser power of the temporally shaped double pulses and V_{rod} is the volume of the nanorod. The input double pulse waveform is in the form of Gaussian profiles in both of space and time. For simulations of the ultrafast near-field scattering process, the dielectric permittivity of gold nano-rod is modeled as a set of explicit equations that are functions of the wavelength, temperature and time with respect to a Drude-like intraband transition term in [17], written as

$$\varepsilon_{intra}[\omega,\ T_e(t),\ T_l(t)] = 1 + \varepsilon_b - \frac{\omega_p^2}{\omega^2 + i\gamma\omega} \tag{8}$$

with parameter,

$$\gamma[T_e(t), T_l(t)] = \gamma_0 + \gamma_1 \times T_l(t) + \gamma_2 \times T_e^2(t) + \gamma_3 \times \omega^2. \tag{9}$$

Here, ω is the optical frequency, t is the time, and $T_l(t)$ and $T_e(t)$ are time-dependent lattice and electron temperatures, respectively. where, ε_b is the background dielectric constant, γ is the damping constant, ω_p is the bulk plasma frequency, and γ_0, γ_1, γ_2, γ_3 are constant coefficients. The transient modification of optical near-field properties can be described as follows: The electron temperature of the nanorod dimer is calculated from the Ohm resistive heating. Then the developed temperature gives rise to modifications to the permittivity applied in Helmholtz electromagnetic scattering equation. The refreshed e-field will cause the rise of a new developed electron temperature. In this way, the near electric fields and the nanorod temperature is developed synchronously on femtosecond timescale until the end of femtosecond pulse duration. Because of dynamics modulation of the electron temperature for gold nano-rod dimer via temporally shaped femtosecond laser, the dielectric function of gold nano-rod dimer can be consequently modified, leading to manipulative plasmon resonance processes in an off-resonance nanorod dimer. To measure the electric field enhancement effects during pulse duration of temporally shaped femtosecond laser, a normalized time average enhancement factor modulated by the shape of the laser is defined as follows:

$$\eta_{ave} = \frac{\int |E(t)| dt}{E_0 \cdot t_p} \tag{10}$$

where, $E(t)$ is the localized electric-field, E_0 is the incident laser field, t_p indicates the pulse envelope duration of temporally shaped femtosecond laser. The temporally shaped double fs pulse consists of double Gaussian pulses with temporal interval from 0 fs to 200 fs. Here, we proposed a dynamics model, in which the two-temperature processes are self-consistently coupled into the near-field scattering model for predictions of fs laser excitation of nanorods dimer. Numerically, model equations including the two-temperature equations and the Helmholtz electromagnetic equation is solved by a iterative algorithm based on Finite Element Method (FEM). The current FEM theme for solving the temperature coupled Helmholtz equation is convergent

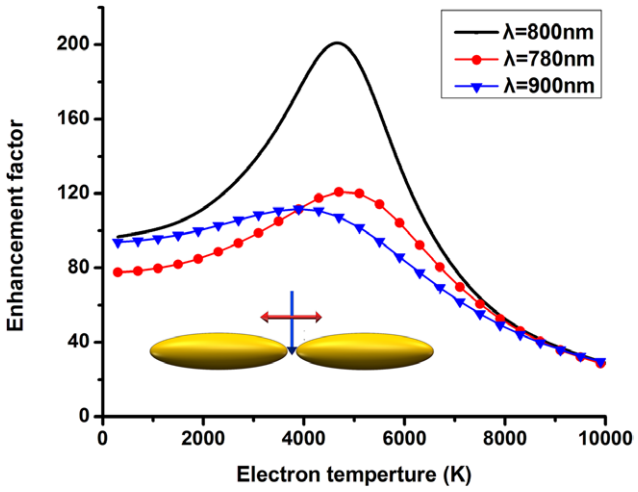

Figure 1. The near electric-field enhancement spectra for a gold nanorod dimer with respect to electron temperature modifications. The length of an individual rod is 220 nm, the gap of the nanorod is 15 nm. The test point is taken at the center of the gap of the nanorod dimer.

by using a FEM software package, consuming about 0.4 h at a given wavelength for the current geometry. The time and space steps are set as 0.5 fs and 1 nm, respectively. Firstly, we build a 2D geometry of nanorod dimer surrounded by ambient medium. A perfect matching layer (PML) is set outside of geometry of the rods dimer. The scattered light from the rods dimer is totally absorbed through the PML in the far-field. The boundary condition at the interface between the dimer and ambient medium is treated as continuous one. Then the dimer geometry, ambient medium and the PML zone are divided into many small meshes. A Helmholtz equation coupled with the two-temperature model is built on the meshed geometry for describing the near-field scattering processes. The Helmholtz equation is then discretized at every mesh points to form a large sparse matrix. Finally, we obtained the numerical solutions of the equation via alternately solving the built matrix using FEM.

3. Results and discussions

The electric-field enhancement spectra for an off-resonance gold nanorod dimer with respect to non-equilibrium thermal excitation dynamics is shown in figure 1. The current dimer geometry is chosen based on the published literature for a individual rod [18]. It shows that the rod length corresponding to the resonance is at around 220 nm. For the nanorod dimer, the plasmon resonances slightly deviate from the individual rod. As a result, we have chosen the length of rod as 220 nm as a base for exploring the tunable resonances of nanorod dimer via further tuning temporally shaped femtosecond laser double pulses.

We can see from figure 1 that the electric-field enhancement factor for the gold nanorod dimer can reach only 90 at room temperature. However, as the electron temperature is tuned to the range from 4000 K to 6000 K, the electric-field enhancement factor can be significantly promoted to be

as high as 200 for typical wavelength of 800 nm of a femtosecond laser. Considering the fact that phonon system of the nanorod dimer can keep undisturbed during femtosecond pulse duration due to the large phonons mass compared to that of the electrons system. As a result, the phonon temperature contribution to electric-field enhancement spectra can be reasonably ignored in the current model. It shows that the maximal enhancement can be achieved when the electron temperature is tuned to a moderate regime. It can be explained based on the fact that the off-resonance nanorod dimer can be returned to resonant one due to electron temperature modulation of dielectrics function. As the electron temperature changes from 4000 K to 6000 K, the simulations show that the dielectric permittivity can be tuned to satisfy the resonance condition. We can also observe that as increasing the incident laser wavelength, the peak of electric-field enhancement spectra shifts toward the low temperature regime. However, when the electron temperature approaches 10^4 K, the electric-field enhancement factor for different wavelengths present consistent declines with increasing the electron temperature. The phenomenon can be attributed to the strong resistive heating process of gold nanorod in high temperature regime, which can be presented by the electron thermal conductivity with respect to the electron collision mechanism. The calculated electron thermal conductivity shows that as the electron–electron collisions term is taken into account, which is the current situation for simulation, the electron thermal conductivity curve appears a peak approximately at the temperature around 5000 K, not shown. When the electron temperature is less than 5000 K, the electron–electron collision becomes tiny. The plasmon resonance gets weak due to the weak role of the resonant electron collisions. On another hand, as the electron temperature exceeds 5000 K, the thermal conductivity gets smaller, indicating an obvious resistive heating localized on the skin layer of the Au nanorod. It potentially causes the near-field energy loss and the reduced e-fields as the electron temperature is larger than 5000 K.

The e-field enhancement as a function of the electron temperature with respect to different incident angle (0°, 10°, 20°) as shown in figure 2. The laser wavelength is at 800 nm. We can see that the increase of incident angle causes the drop of the e-field localized at the gap center of the nanorod dimer. Although the absolute value of e-field is decreased with increasing incident angle, the general law of the dependence of e-field enhancement on the electron temperature is less affected by changing the incident angle. It indicates that the e-field enhancement dependences on the laser parameters of temporally shaped fs double pulses can also be less affected by increasing the incident angle. But the absolute value of e-field enhancement with respect to the temporally shaped fs pulses excitation can be extracted from figure 2 for the oblique incidence situation.

As the incident angle is very small, the decrease of e-field can be very tiny due to the increase of the incident angle. The results will be helpful for well understanding of ultrafast thermal dynamics for tuning of near-field effects via a oblique incidence temporally shaped femtosecond laser.

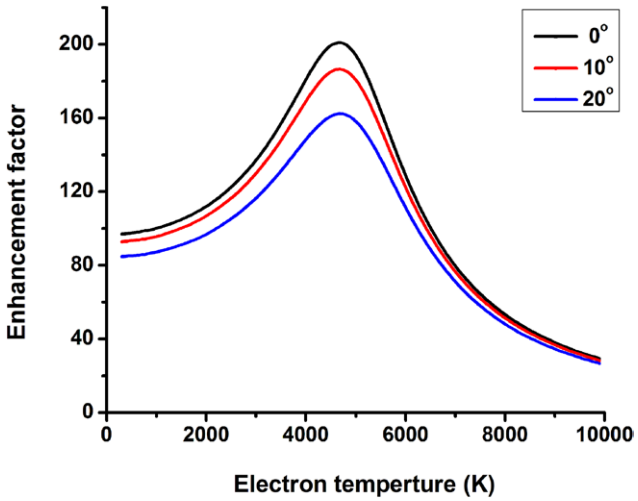

Figure 2. The e-field enhancement as a function of electron temperature for different incident angle. The angle increases from top to bottom lines.

The spatial-temporal dynamics of near electric-field enhancement for an off-resonance gold nanorod dimer excited by temporally shaped double femtosecond pulses is shown in figure 3. It shows that the spatial-temporal distributions of near electric-field along the gap of the nanorod dimer exhibits significant dependence on double pulse separation. At pulse separation of 50 fs, the near electric-field profiles overlap in time domain (figure 3(a)), and the spatial distribution of the near electric-field is mostly concentrated at the inner tips of the nano-rod dimer. With increasing pulse separation, the near electric-field profile is obviously broadened at 100 fs (figure 3(b)), indicating a prolonged plasmon relaxation period. However, as the double pulse separation is continuously increased to150 fs (figure 3(c)), the near electric-field profiles begin to be separated partially in time domain. Finally, the electric-field profiles is separated at 200 fs (figure 3(d)), indicating the disappear of successive electric field profile in time domain. The specific separation of 150 fs indicates the localized plasmon relaxation period for a femtosecond pulse excitation. The results show that the successive near electric-field enhancement can be closely relevant to the pulse separation of temporally shaped double femtosecond pulses. The near electric-field can be significantly modulated due to the temperature-dependent electric-field enhancement mechanism (figure 1). Considering the fact that the electron thermal dynamics for the gold nanorod dimer can be regulated via tailoring pulse separation of temporally shaped femtosecond double pulses. As a result, the near electric-field enhancement for the nanorod dimer can be flexibly manipulated via optimizing parameters of temporally shaped femtosecond laser. It becomes urgent to understand the ultrafast thermal dynamics manipulation of electric-field enhancement on an off-resonance nanorod dimer via varying temporal separation of shaped double femtosecond pulses. In fact, the near electron temperature in a gold target can be regulated not only by the simple parameter of pulse separation, but also the complex parameters such as pulse energy ratio, pulse exchange and laser fluence of temporally shaped femtosecond laser. It is highly expected to get a well insight

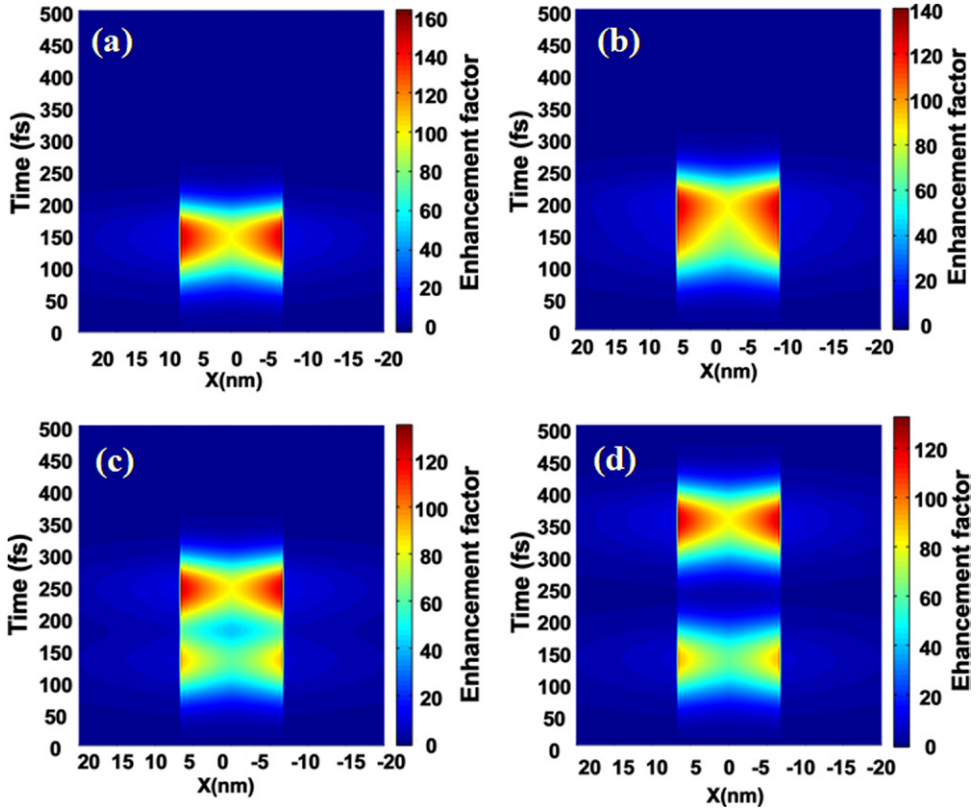

Figure 3. The spatial-temporal dynamics of electric near-field of an off-resonance gold nanorod dimer excited by temporally shaped double femtosecond pulses. Electric-field enhancement along the longitudinal direction with double pulse excitations at pulse separation of (a) 50 fs, (b)100 fs, (c)150 fs and (d) 200 fs.

into the thermal dynamics for modulation of the electric-field enhancement in a gold nanorod dimer via manipulating the complex parameters of temporally shaped femtosecond laser.

The time-averaged electric-field enhancement at gap center for an off-resonance nanorod dimer as a function of pulse separation of temporally shaped double femtosecond pulses are shown in figure 4. It can be seen that as double pulse separation is less than 150 fs, the increase of pulse separation gives rise to obvious promotion of the electric-field enhancement factor. However, as the pulse separation exceeds 150 fs, the electric-field enhancement factor tends to get saturation.

Physically, for double pulses excitation of metallic nanostructures, the first pulse preheats the target, leading to the generation of overheated electrons, whose temperature plays an important role in affecting the absorption of the coming second pulses. As the temporally double fs pulse separation is less than the electron-lattice coupling period, typically on the timescale of a few picoseconds, the absorption of the second pulse can be modified due to the existence of high-temperature electrons in the metallic targets according to the temperature-dependent Drude model prediction. For the current double fs pulses excitation of nanorod dimer, the first pulse gives rise to the nanorod dimer electron temperature, the second pulse then coupling into the excited electron of the nanorod dimer. As a result, the cooperative works of double fs pulse can play a key role in determining the total heating property of the nanostructure. The results can be explained as follows: as the pulse separation is less than 150 fs, the main parts of double pulses

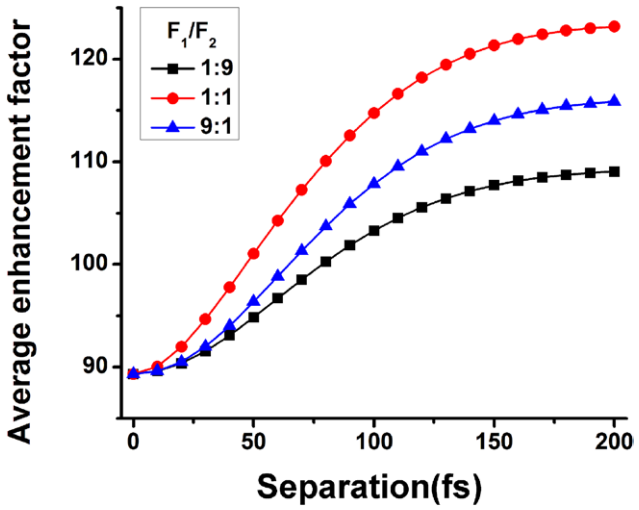

Figure 4. The average electric-field enhancement at the gap center of a gold nanorod dimer excited by temporally shaped double femtosecond pulses. The length of an individual rod is 220 nm, the gap of the nanorod is 15 nm. The incident laser wavelength is 800 nm.

overlap in time domain, the increase of pulse separation gives rise to the average electron temperature up to 4500 K as in our simulations, which falls into the strong near-field enhancement regime for the current 800 nm wavelength femtosecond laser. As a result, the electric-field enhancement factor is promoted with increasing pulse separation. In addition, our calculations show that as the transient electron temperature

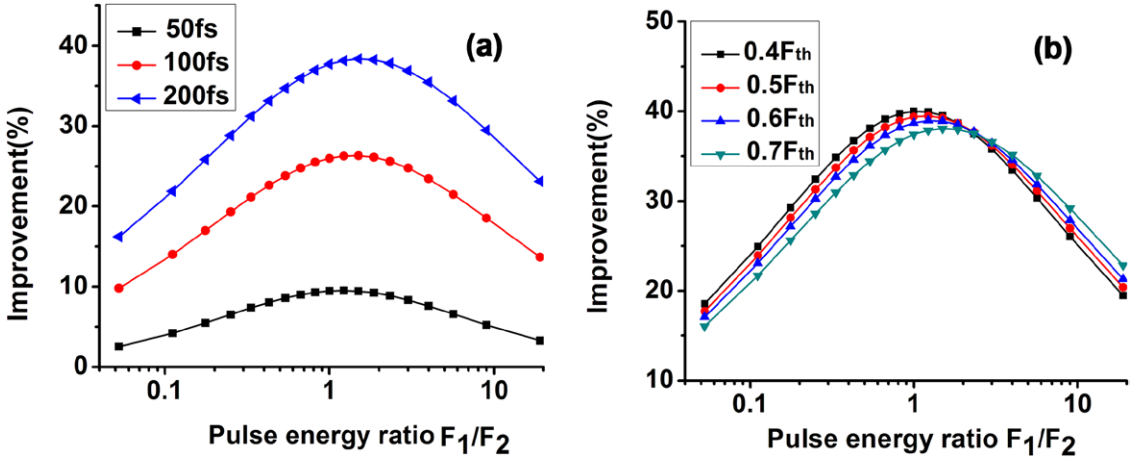

Figure 5. The time-averaged electric-field enhancement at the gap center for an off-resonance nanorod dimer as a function of pulse energy ratio of temporally shaped double femtosecond pulses. The length of an individual rod is 220 nm, the gap of the nanorod is 15 nm. The incident laser wavelength is 800 nm. (a) for different pulse seperation, (b) for different laser fluence.

of gold nanorod dimer is reduced to value as small as 3500 K at a lower fluence of 0.7 F_{th}, the near electric-field can be crippled for laser excitations with double femtosecond pulses, which is not shown. It leads to the plasmon off-resonance process according to the near-field enhancement spectra (figure 1), which is extremely unfavorable for promoting near-field enhancement for the nano-dimer.

The time-averaged electric-field enhancement at gap center for an off-resonance nanorod dimer as a function of pulse energy ratio of temporally shaped double femtosecond pulses are shown in figure 5. As the pulse energy ratio is smaller than 0.1, the electric-field enhancement factor drops to the value less than 5. Meanwhile, the electric-field enhancement factor is obviously reduced as the pulse energy ratio exceeds 10 (figure 5(a)). The results can be explained as follows: the double pulses trends to perform like a big single pulse for pulse energy ratios of 0.1 and 10. However, the electron temperature can be promoted to be as high as 1 eV for a single femtosecond pulse excitation as found in our previous publication [12]. Therefore, the near electric-field enhancement in the gold nanorod dimer is reduced due to the electron temperature driven electric-field enhancement mechanism (see figure 1). On the contrary, as the pulse energy ratio is very close to 1 : 1, the maximal electron temperature of the gold nanorod dimer can be tuned to value around 5000 K as predicted in our simulations. The naorod dimer excitation will fall into the strong plasmon resonance regime. As a result, the off-resonance gold nanorod dimer can be tuned to the resonance state due to moderate excitation. It can be seen from figure 5(b) that the maximal electric-field enhancement occurs at the laser fluence of 0.4 F_{th} for the pulse energy ratio of 1 : 1. However, the near electric-field enhancement can be reduced for a higher laser fluence, which is explained as that the high electron temperature driven detuned resonance at large laser fluence. The results will be greatly helpful for advancing the applications of giant electric-field enhancement in, e.g. fs super-resolution fs near-field imaging, fs near-field optical tweezers, and fs photothermal therapy.

4. Conclusion

In summary, we have theoretically investigated ultrafast dynamics for tuning of near-field enhancement on an off-resonance gold nanorod dimer via temporally shaped femtosecond laser. It is revealed that the maximal electric-field enhancement of more than 200 times can be acquired at an off-resonance nanorod dimer via modulating the electron thermal excitation process. The average electric-field enhancement for gold nanorod dimer can be greatly promoted at optimal pulse separation of 150 fs and the pulse energy ratio around 1 : 1. The results are explained as tunable plasmon resonance in nanorod dimer via temporally shaped femtosecond laser. This study provides basic understanding for tuning the near-field enhancement on off-resonance metallic nano antenna via tailoring the temporal features of femtosecond laser pulses for a wide range of application in the fields such as femtosecond (fs) superresolution near-field imaging, fs near-field optical tweezers, and fs laser photothermal therapy.

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