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RAPID COMMUNICATION

Dynamic near-field nanofocusing by V-shaped metal groove via a femtosecond laser excitation

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Abstract The ultrafast dynamics of plasmonic near-field nanofocusing by a V-shaped groove milled on Au film via a femtosecond laser excitation is theoretically studied based on finite element method. The spatiotemporal evolution of the focused e-fields around the V-groove geometry is obtained. It is revealed that the strong nanofocusing at the V-shaped groove occurs at the moderate electron temperature of 3000 K in the electron–phonon uncoupled state via a femtosecond laser pulse excitation. The phenomenon is explained as the electron thermal dynamics manipulation of plasmon resonances due to femtosecond laser fluence modifications. This study provides basic understanding of ultrafast dynamics of near-field nanofocusing in V-shaped geometry for wide applications in the fields such as super-resolution imaging, SERS, and photothermal therapy.

1 Introduction

The nanofocusing of plasmon at tapered nanostructures is important for the near-field applications such as high-resolution imaging, SERS, optical tweezers and photothermal surgery [1–4]. Illumination of these tapered nanostructures by bulk electromagnetic radiation at a resonant frequency can cause strong resonant enhancement of the localized e-field of the supported plasmonic mode, reducing the incident light beam into a region with dimensions smaller than those allowed by the diffraction limit of light. Physically, the plasmon near field formed on nanofocusing geometry can be supported by the collective electron oscillation mode at designed nanostructures such as tapered insulator–metal (IM) and metal–insulator–metal (MIM) structures. Among them, the V-shaped groove milled on a metal film exhibits great importance for concentration of light into nanoscale spot for near-field nanofocusing applications. Most of previous works focused on investigations of the nanofocusing with respect to the structure matching of the shape and size of V-groove geometry to the designed one [5–10]. However, it is usually in the cost of the fabrication engineering's complexity for obtaining the designed geometry, which brings in a challenge to accurately fabricate the ideal geometry in order to enhance the near e-field of nanofocusing.

In recent years, the femtosecond laser excitation of nanostructures for advancing near-field applications had attracted great interest [11–14]. The use of femtosecond laser excitation of V-shaped groove can bring in potential benefits to greatly enhance near field at focused region compared with continuous laser or long pulsed laser irradiation (ns or longer). It can benefit many applications from super-resolution imaging, SERS, to photothermal therapy. Generally, the electron system of the metallic groove can be excited via a femtosecond laser (the electron temperature rises up to 1 eV). It causes variations of dielectric function, leading to the modification of optical properties. Previous study of the effect of different properties of nanostructures was performed at the room temperature. While the optical properties of both the bulk metals and the nanostructures in the low-temperature regime are now well studied, the understanding of the effects of electron temperature (typically of the order of

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10^3 K) on the plasmon modes has remained a challenge for plasmonic nanofocusing and is not so well understood as to the temperature at room temperature. It is believed that the near-field plasmon resonances on a typical metal groove geometry can be dynamically modified via femtosecond laser pulses excitation. It enables the new opportunity for promoting the nanofocusing applications for the V-shaped grooves, especially for poorly fabricated V-groove geometry with size and shape deviating from the designed one. However, the physical mechanism behind the dynamic nanofocusing is still puzzling for well exploring the near-field nanofocusing applications of the V-shaped grooves. Generally, as a femtosecond laser pulse irradiation of the metallic nanostructures, the electron system of the metal nanostructures is firstly excited on femtosecond timescale [15, 16]. Then the electrons will transfer its energy to cool lattice, which occurs on the timescale of several picoseconds. It is believed that the localized e-field around tip structures can be sharply modified due to the thermal dynamics tuning of the dielectric properties [17]. As a result, the thermal excitation plays an important role in affecting the near-field nanofocusing. However, one of the important practical aspects that have received only very limited attention so far is the possibility of non-equilibrium heating of nanofocusing structures, which may lead to significant alteration in their properties, self-influence of focused dynamics plasmon, and even rapid destruction of the structure, especially near the tip where the energy dissipation rates are the greatest. It becomes important to understand the ultrafast dynamics of near-field nanofocusing by a V-shaped groove via femtosecond laser excitation for exploring a wide range of potential applications.

In this paper, we theoretically investigated the ultrafast dynamics of plasmonic near-field nanofocusing at a V-shaped groove milled on Au film via femtosecond laser excitation. The non-equilibrium thermal excitation dynamics is self-continuously coupled into the near-field scattering model for well exploring the spatiotemporal dynamics at a V-groove milled on Au film. The potential thermal dynamics mechanism for regulating the ultrafast near-field plasmonic nanofocusing is examined in details. The ultrafast dynamics of the near-field nanofocusing properties with respect to a femtosecond laser irradiation parameters are explored based on the proposed model. We proposed a self-consistently dynamics model, in which the two-temperature thermal excitation process is coupled into the near-field scattering model for well predicting the femtosecond laser excitation of metal V-groove. First, the laser pulse is absorbed by the electronic system of V-groove, creating nascent non-thermal electrons system. After 100 fs, a thermal equilibrium is reached corresponding to the Fermi distribution as a result of the electron–electron scattering with cold conduction band electrons. Subsequently, the electron energy is transferred to the Au groove lattice via electron–phonon coupling (<a few ps). In the current study, we adopt the well-known two-temperature model, in which the electron-to-electron and electron–phonon coupling processes are taken into account based on the temperature-dependent electron collisions mechanisms. Herein, the dielectric permittivity can be calculated as a set of explicit equations as functions of frequency and time with respect to a Drude-like intraband transition term as follows: [18]

$$\varepsilon_{\mathrm{intra}}[\omega, T_{\mathrm{e}}(t), T_{\mathrm{l}}(t)] = 1 + \varepsilon_{\mathrm{b}} - \frac{\omega_{\mathrm{p}}^2}{\omega^2 + i\gamma\omega} \tag{1}$$

with parameter,

$$\gamma[T_{\mathrm{e}}(t), T_{\mathrm{l}}(t)] = \gamma_0 + \gamma_1 \times T_{\mathrm{l}}(t) + \gamma_2 \times T_{\mathrm{e}}^2(t) + \gamma_3 \times \omega^2 \tag{2}$$

where ω is the optical frequency, t is the time, and $T_{\mathrm{l}}(t)$ and $T_{\mathrm{e}}(t)$ are time-dependent lattice and electron temperatures, respectively. Based on current model predictions, the temperature-dependent plasmon collision frequency can significantly affect the dielectrics function for the near-field of nanofocusing. As a result, the resonance position can depend on the electron temperature.

The model equation including two-temperature equations and Helmholtz electromagnetic equation is solved by an iterative algorithm based on finite element method. Here, we have carefully chosen the FEM meshes as triangles with side length of 1.5 nm to assure the accuracy of the simulation results. The electron temperature of the groove geometry is firstly calculated due to laser excitation based on the Ohm resistive heating. Then the developed temperature gives modification to near e-field due to the variation of the permittivity of gold groove. The refreshed e-field will lead to a new developed electron temperature of the V-groove. In this way, the near e-fields and the V-groove temperature are developed synchronously on femtosecond timescale until the end of femtosecond pulse duration.

The calculated 2-D images of normalized e-field magnitude for nanofocusing by a V-shaped groove milled on Au film with different electron temperatures in the uncoupled electron–phonon state are shown in Fig. 1. Herein, the phonon system of Au film is considered as being at room temperature because the Au film phonon less can be disturbed during the plasmon excitation period on femtosecond timescale. It can be seen that the e-field magnitude at the nanofocusing spot is slightly promoted at 300 K (Fig. 1a), indicating the weak nanofocusing at room-temperature condition. With increasing the electron temperature at 1500 K, the e-field magnitude at the

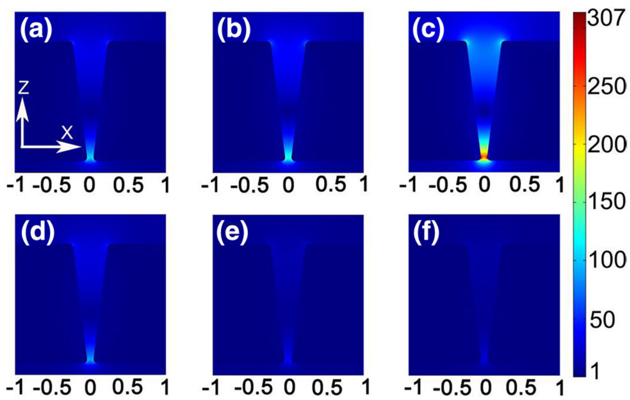

Fig. 1 Calculated 2-D images of e-field magnitude within the nanofocused region at different electron temperature in the uncoupled electron–phonon non-equilibrium state of the V-shaped groove milled on Au film. The depth of the groove is 160 nm, and the gap distance at the front and rear surfaces is 54 and 19 nm, respectively. The incident laser wavelength is 800 nm. **a** 300 K, **b** 1500 K, **c** 3000 K, **d** 4500 K, **e** 6000 K, **f** 7500 K

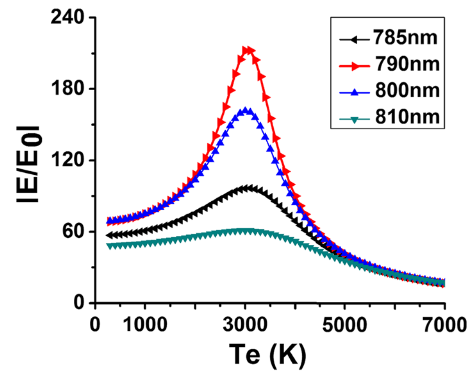

Fig. 2 The e-field enhancement spectra calculated at the gap center of the *bottom* for the V-groove milled on Au film

V-grooved nanofocusing geometry is less affected (Fig. 1b). More interestingly, it shows that the strongest nanofocusing occurs at the moderate electron temperature of 3000 K for the current V-shaped groove geometry (Fig. 1c). Once the electron temperature exceeds 3000 K, the maximal e-field amplitude at the near-field nanofocusing geometry of the V-shaped groove presents obvious drop with increase in the electron temperature (Fig. 1d–f). It is explained as the thermal dynamics enhanced surface plasmon resonance at the moderate electron temperature of 3000 K corresponding to the resonant temperature modification of permittivity of Au film as demonstrated by the e-field enhancement spectra (Fig.2). As the electron temperature is at room temperature, the V-groove is off resonance at 800 nm due to the structure mismatching of current geometry. As the electron temperature is at 3000 K, the dielectric function modifications for the exact Au plasmon resonance of a V-groove geometry can be assured due to the contribution of the temperature-modified collision frequency, leading to the enhanced near fields during the nanofocusing. However, as the electron temperature departures from 3000 K, the detuned electron collisions can cause the off-resonance process in the V-groove. The near field for nanofocusing is lowered as the electron temperature departures the resonance temperature, and simultaneously the resistive heating in high excited electron system begins to play an important role in causing reduction in near-field energy. Therefore, the drop in the e-field amplitude at the nanofocusing point of the V-groove is observed in high-electron-temperature regime until Femi level. The results can be important for basically understanding of the near-field nanofocusing properties with respect to the electron thermal excitation in high non-equilibrium state of a metallic focusing geometry.

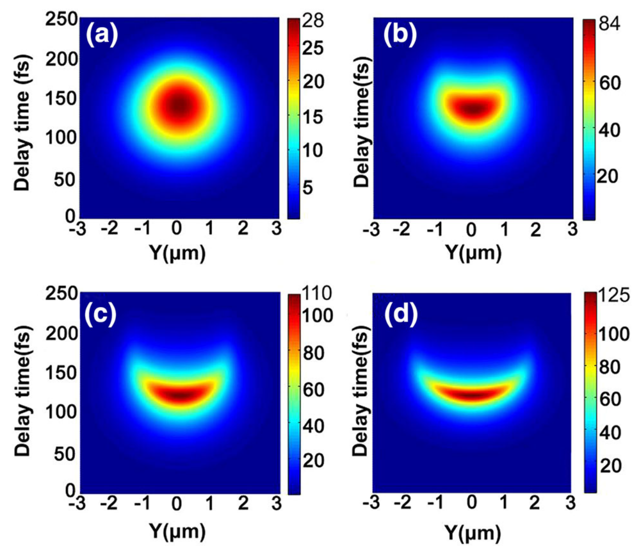

Fig. 3 The spatiotemporal evolution of near-field enhancements in the V-shaped groove. The depth of the groove is 160 nm, and the gap distance at the front and rear surfaces is 54 and 19 nm, respectively. The pulse fluence is **a** 0.3 Fth, **b** 0.5 Fth, **c** 0.7 Fth, **d** 0.9 Fth. The pulse duration is 65 fs, the laser wavelength is 800 nm

The spatiotemporal evolution of normalized e-field magnitude along the focusing line of the V-shaped groove with a femtosecond laser pulse irradiation is shown in Fig. 3. Here, the e-field polarization direction is treated to be parallel to surface of the V-shaped groove. The incident femtosecond laser pulse is treated as standard Gaussian type in both temporal and spacial dimensions. The spectral profile is also Gaussian-shaped with wavelength centered at 800 nm. We can see that the e-field images at the focusing line of V-groove exhibits Gaussian-like profiles in both the temporal and spatial regimes for lower laser fluence of 0.3 Fth. The laser fluence can also be called energy density in the unit of J/cm^2. Fth is defined as the threshold fluence for damage of the Au film. However, with increasing laser fluence, the Gaussian-like profile is reshaped at the top of

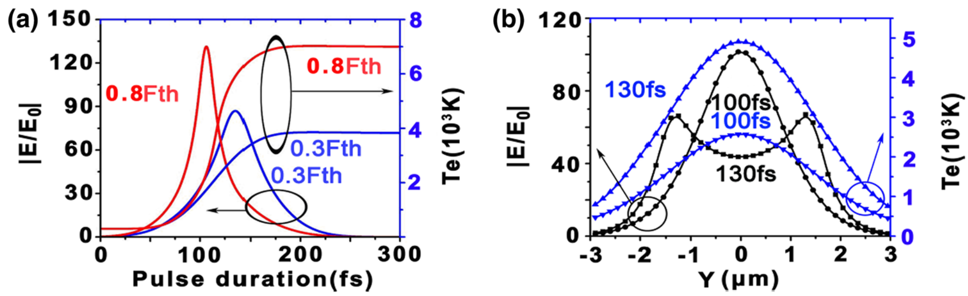

Fig. 4 The normalized e-field magnitude distributions for the V-groove with respect to the electron temperature modifications with a femtosecond laser pulse irradiation. The depth of the groove is 160 nm, and the gap distance at the front and rear surfaces is 54 and 19 nm, respectively. (**a**) Temporal shape of e-field and temperature, (**b**) Spatial distribution of e-field and temperature

the image. At laser fluence of 0.5 Fth a shallow hole is formed at the top of the Gaussian-like image.

It shows that the distorted spacial distribution of the nanofocusing e-field profile begins to appear at laser fluence of 0.5 Fth. The near e-field localized at the tips of V-groove is observably promoted at the laser fluence of 0.5 Fth. With increasing the laser fluence, a sickle moon shape appears at the fluence of 0.7 Fth, and the near e-field of the focusing image at 130 fs is further promoted (Fig. 3c), as indicated in the center of the sickle moon shape. The maximal e-field enhancement at the nanofocusing zone reaches 110 for laser fluence of 0.7 Fth. As the laser fluence further increases to 0.9 Fth, the sickle moon-like shape is bended; meanwhile, the e-field enhancement at the focusing image is promoted to the value of 125. The spatiotemporal evolution with respect to the laser fluence can be attributed to the electron thermal dynamics manipulation of localized surface plasmon resonances for the V-groove, which will be explained in detail in the following section. The results are important for understanding of the spatiotemporal nanofocusing properties at a V-shaped groove, which provides the base for tuning nanofocusing properties via modifying laser fluence.

The normalized e-field magnitude distributions for the V-groove with respect to the electron temperature modifications with a femtosecond laser pulse irradiation are shown in Fig. 4. We can see from Fig. 4a that the temporal shape of the e-field magnitude presents Gaussian pulse-like profile for laser fluence of 0.3 and 0.8 Fth, respectively. More interestingly, the near-field focusing magnitude can be obviously modified by laser fluence, indicating that the plasmon resonances of V-groove nanofocusing geometry can be well controlled via tuning laser fluence, which is extremely important for manipulation of the near-field of nanofocusing for a wide range of applications. It can be explained as follows: For the lower laser fluence of 0.3 Fth, the electron temperature presents slow increase from 300 to 3800 K as in the current simulations. In such situation, the strong resonance peak of localized surface plasmon appear at 130 fs, at which the plasmon resonance is satisfied due to the appearance of moderate temperature modification at 3000 K (Fig. 1), leading to large e-field amplitude of plasmonic nanofocusing spot.

However, for laser fluence of 0.8 Fth, the resonance peak appears at an early time of 100 fs. It can be attributed to the fact that the 3000 K temperature will be achieved earlier for higher fluence, along with a stronger near-field enhancement, meanwhile, which will be achieved at later time for a lower laser fluence. Also, we can see that the peak magnitude of the nanofocusing is obviously larger at the laser excitation with fluence 0.8 Fth compared with the lower laser fluence of 0.3 Fth. Figure 4b shows the e-field magnitude spatial distributions along the focusing line at the bottom of the V-shaped groove at different delay time. It can be seen that the near e-field distributions of the nanofocusing exhibits large difference at different delay time. At 100 fs, the spatial e-field amplitude distributions for the V-groove nanofocusing present Gaussian-like along the focusing line. However, it is obviously hollowed at the central line at 130 fs. It originates from the high-temperature (>3000 K)-driven plasmon off-resonance mechanism in the V-shaped groove. The results are important for understanding of the dynamics for tuning the near-field nanofocusing via modifying the laser fluence.

2 Conclusion

In conclusion, we have theoretically investigated the ultrafast dynamics of near-field plasmonic nanofocusing by a V-shaped groove milled on Au film via a femtosecond laser pulse irradiation. The spatiotemporal evolution of the near e-fields of plasmonic nanofocusing at the V-groove geometry is obtained via solving the proposed self-continuously dynamics model. It is found that the strong nanofocusing at the V-shaped groove occurs at the moderate electron temperature of 3000 K in the electron–phonon uncoupled state. Moreover, the near-field focusing magnitude can be obviously modified by laser fluence, indicating that the plasmon resonances of V-groove nanofocusing geometry can be well controlled via tuning laser fluence, which is extremely important for manipulation of the near-field of nanofocusing for a wide range of applications. The study provides the guideline for optimizing the near-field properties in V-shaped groove for a wide range of potential applications.

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