Subwavelength imaging and control of ultrafast optical near-field under resonant- and off-resonant excitation of bowtie nanostructures

Boyu Ji, Jiang Qin, Haiyan Tao, Zuoqiang Hao and Jingquan Lin

1 School of Science, Changchun University of Science and Technology, Changchun 130022, People’s Republic of China
2 International Research Center for Nano Handling and Manufacturing of China, Changchun 130022, People’s Republic of China
3 Author to whom any correspondence should be addressed.

E-mail: zqhao@cust.edu.cn and linjingquan@cust.edu.cn

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Abstract
We demonstrate subwavelength imaging and control of localized near-field distribution under resonant and off-resonant excitation of identical gold bowtie nanostructures through photoemission electron microscopy. Control of the near-field distribution was realized by polarization rotation of single femtosecond laser pulse and variation of the phase delay of two orthogonally polarized femtosecond laser pulses. We show that the localized optical near-field distribution can be well controlled either among the corners of the nano-prisms in the bowtie for resonant excitation or the edges for off-resonant excitation. A better visualization of the PEEM image is achieved for resonant excitation than in the case of off-resonant excitation. The experimental results of the optical near-field distribution control are well reproduced by finite-difference time-domain simulations and understood by linear combination of electric charge distribution of the bowtie by s- and p- polarized light illumination. In addition, a shift of the near-field excitation position with inverted or unchanged phase, alternatively an un-shift of the excitation position but only with inverted phase of the near-field, can be realized by rotating the polarization angle of a single pulse and coherent control of two orthogonally polarized fs laser pulses.

1. Introduction
Localized surface plasmons (LSPs) are coherent collective oscillations of charge carriers inside a metal nanostructure that result in the confinement of the electromagnetic field below the diffraction limit of traditional optics and have attracted considerable research interest for a broad range of applications in sensing [1, 2], imaging [3–5], solar cell [6] and so on. Special attention has been given to the active control of LSPs in the vicinity of noble metal nanostructures which offers a way to manipulate light on the nanoscale [7]. Such control has a great potential in many fields, such as information technology and biomedicine, and it has great promise to improve the development of nano-optics.

Near-field control plays an important role in the research field of plasmonics [8–10]. Active near-field control in the vicinity of nanostructures can be realized in many ways. For example, phase modulation of one ultrashort pulse (i.e. modulating the chip of pulse) [11], adaptive near-field control related to multi-parameter pulse shaping with a learning algorithm [12, 13], polarization rotation of a single linearly polarized pulse [14–17] and relative phase delay modulation of two orthogonally polarized laser pulses (hereby named as ‘coherent control’) [14, 18]. Among these techniques, polarization rotation and coherent control are two widely used control schemes because they avoid the use of complex closed-loop learning algorithms and are easily achievable by a convenient experimental setup [14].

Effort has been made to pursue maximal field enhancement in the research of excitation and control of LSPs, and this has been realized by tuning illumination wavelength to the resonant peak of a nanostructure [19]. This is
vital to some applications such as single molecule sensing, detecting, high harmonic generation and surface enhanced Raman scattering (SERS) in which a maximal LSP enhancement is desired \[7, 20–22\]. Meanwhile, with a broadening application of LSP effect, researchers have found that excitation of LSP with illumination wavelength tuning off the resonant peak often becomes important, i.e. employing LSP effects under off-resonant conditions. For example, off-resonant excitation of LSPs in nanostructure is found to have great potential applications in the field of biomedicine, in which the generation of nanobubbles and simultaneously avoiding the melting of nanoparticles under resonant excitation that could bring toxicity issues is required in this application \[23\]. Hence, in addition to the resonant excitation, study of near-field control under off-resonant excitation in nanostructures is of importance. Up to now, a full and direct study to the optically induced near-field control within single nanoparticles both under resonant and off-resonant excitation is still absent. Besides this point, the dependence of the relative phase status of near-field upon the excitation scheme is an interesting and unexplored topic.

In this paper, by tuning the output laser wavelength, we control the distribution of near-field in bowtie structure under resonant (850 nm) and off-resonant (700 nm) conditions with the schemes of polarization rotation of single laser pulses and variation of relative phase delays of orthogonally polarized two femtosecond laser pulses. In the mean time, we investigated the dependence of the relative phase status upon the two excitation schemes. The control of LSP processes in bowtie nanostructure is imaged with a photoemission electron microscope (PEEM), which provides a non-invasive probe of optical near-field by imaging field-induced photoemission electrons with high spatial resolution. The technology has been widely used to study the dynamics of surface plasmon evolution \[24–27\]. Experimental results show that, under resonant wavelength excitation, enhancement of the near-field mainly locates at the corners of the nano-prisms and can be controlled by rotating polarization of a single laser pulse or by varying the relative phase delay of orthogonally polarized two femtosecond laser pulses. Under the off-resonant wavelength excitation, the enhanced near-field mainly locates at the edge regions of the nano-prism, and the near-field shows a relatively less prominent visualization compared with the resonant case. Furthermore, a linear combination of the simulated charge distribution in s- and p- polarized cases could be used to better understand near-field control behavior, and the simulation results are in good agreement with the results of the PEEM measurement. To the best of our knowledge, this is the first engaged discussion on the dependence of the relative phase status upon the excitation scheme and also near-field distribution difference within a nanostructure between resonant and off-resonant excitations.

2. Experimental and simulation setup

Figure 1 shows the experimental set up. PEEM (FOCUS, IS-PEEM) with 30 nm spatial resolution was used to image the optical near-field of a bowtie nanostructure. Two excitation sources were used, one was an unpolarized Hg lamp (one photon energy less than 5.2 eV, to contour the bowtie antenna) and the other was a femtosecond Ti-Sapphire laser oscillator (Coherent, Mira 900), delivering pulses with a duration of 200 fs, tunable output wavelength (from 680 nm to 900 nm), single pulse energy of 13 nJ and a repetition rate of...
3. Results and discussion

We first clarify the resonant wavelength for the fabricated bowtie structure. The wavelength-dependent near-field property of \( \Gamma_1 \) of the bowtie (marked in figure 1(d)) by PEEM measurement as well as by FDTD simulation, and also absorption spectrum of the bowtie structure by FDTD simulation are given in figure 2. To present curves of similar physical significances, the cube root of the photoemission electron yield was displayed [32]. From the wavelength-dependent PEEM signal as well as the FDTD simulated near-field intensity curve in figure 2, we found that resonance of the structure was obtained at a wavelength near 850 nm. The simulated absorption curve in figure 2 shows a resonant peak appearing at wavelengths of 836 nm. Difference in resonant wavelengths between the absorption and maximum electric intensity at the surface had already been reported by several other groups [33–35]. Based on the laser spectral tunable range (from 680 to 900 nm, shadow area in figure 2), 850 nm is used as resonant excitation wavelength and 700 nm is selected as off-resonant excitation for the nanostructure in the current research.

Near-field distribution control under femtosecond resonant wavelength excitation was investigated. Figure 3(a) shows the PEEM images by rotating polarization direction of the femtosecond laser pulse for 850 nm resonant modes. Comparing with the one photon PEEM image with Hg lamp illumination as shown in figure 1(c), PEEM images obtained with a femtosecond laser pulse show that near-field distribution is not uniform on the nano-prisms surface anymore but mainly is located at corners of the structure as hotspots in the PEEM images. It is seen that hotspot distributions depend on the angle \( \phi \) as polarization direction deviates (in the anti-clockwise direction) from \( p \)-polarization of the incident light (0°). Figure 3(a) shows that near-field is mainly enhanced at \( \Gamma_1 \) position for \( p \)-polarization cases. As the polarization angle \( \phi \) of a laser pulse tunes to
\[ \theta = 45^\circ, \text{ the local field at G}_1 \text{ position becomes weak and strong near-field starts to emerge at C}_2 \text{ position. As the polarization } \theta \text{ increase to } 90^\circ (s\text{-polarization}), \text{ the photoemission electron signal induced by local field at the G}_1 \text{ position becomes invisible, excitation of C}_1 \text{ and C}_2 \text{ are clearly visible as well. With the further increase of polarization angle } \theta \text{ to } 135^\circ, \text{ the hotspot at G}_1 \text{ position reappears and the strong near-field enhancement shifts to C}_1 \text{ from the C}_2 \text{ position that is symmetrical to the case of } 45^\circ. \text{ The results clearly show that strong near-field distribution can be controlled among the corners of the bowtie nanostructure by tuning the polarization angle of the excitation laser pulse. In the experiment, we have also performed control of local field distributions by using a pair of orthogonally polarized laser pulses; the corresponding PEEM images are displayed in figure 3(c). Similar to the results obtained by rotating the polarization direction of single laser pulses, strong local field enhancement at C}_1 \text{ and C}_2 \text{ positions can also be selectively achieved by varying relative phase delay } \Delta \Phi \text{ of the two pulses. As shown in figure 3(c), strong near-field can be shifted from the C}_2 \text{ to C}_1 \text{ positions by changing the phase delay from } \Delta \Phi = 0 \text{ to } \Delta \Phi = \pi. \text{ It should be noted that the near-field distribution control under the latter scheme has been realized at nanometer spatial resolution and attosecond temporal precision (} \Delta \Phi = \pi/2, \text{ corresponding to the time delay of } \Delta t = 708 \text{ attoseconds between the two pulses). We should mention that the coherent control method we used here is actually also a polarization control, however, it is switched from...} \]
diagonally linear polarization under 45° (equivalent to the case of $\Delta \Phi = 0$), to right handed circular polarization ($\Delta \Phi = \pi/2$), and then to diagonally linear polarization under $-45^\circ$ ($\Delta \Phi = \pi$), and finally to left handed circular polarized ($\Delta \Phi = -3\pi/2$) illumination.

It is interesting to note that a common feature of the near-field pattern of the PEEM images in figure 3 is that only the farther side of individual nano-prisms in the bowtie (referring to C1 and C2 corners of the left side nano-prism, and G1 corner of the right side nano-prism, respectively) get stronger enhancement rather than the nearer side (referring to G2 corner on the left side nano-prism and C3, C4 corners on the right side nano-prism). This characteristic regarding to local field distribution in the bowtie may be attributed to a retardation effect that occurs when the structural size under study is in the order of the wavelength of the oblique incident of the excitation light pulse [18], or to interference between bright- and dark-modes that are supported by a bowtie structure [36]. It is noted that the design of the antenna is actually just two rather independent nano-prisms. The difference in response between both nano-prisms arises because of the different orientation with respect to the incident beam. There is no strong coupling between the nano-prisms, which is different from the case of a real bowtie antenna structure.

In this work, off-resonant mode near-field control is also investigated with the two control schemes as introduced in the resonant wavelength excitation case. Figure 4 shows the PEEM images obtained by rotating the polarization direction and varying the phase delay of two laser pulses for the 700 nm off-resonant mode. Different from the near-field distribution under resonant wavelength excitation, a prominent feature of the figure 4 is that the enhanced near-field regions distribute along edges of the nano-prisms. As shown in figures 4(a) and (c), the local field distributions show noticeably controllable modulation when the polarization angle and relative phase delay are changed. It is seen that near-field in figure 4(a) mainly locates in two edges M3 and M4 (labeled by red dot) of the right prism at 0°, and then the field focus on the M4 as the polarization angle tunes to 45°. The field shifts to M5 as the angle further tunes to 135°. For the case of the two laser pulses scheme, the fields mainly locate in the lower edge M3 of the right nano-prism for the relative phase delay $\Delta \Phi = 0$, and the near fields shift to the upper edge M4 of the right nano-prism as the relative phase delay is changed to $\pi$.

Finally, the field returns to M4 again as the relative phase tines to $\Delta \Phi = 2\pi$. These results demonstrate that active control of near-field at edge of the nano-prism can be achieved by rotating polarization of single pulse and changing relative phase delay of the two laser pulses. In addition, it is seen that near-field on the vertical edge of the left nano-prism shows high intensity in the case of the off-resonant excitation.

Obviously, visualization of the near-field distribution control is less prominent for the off-resonant excitation compared to the resonant case. And also, for the off-resonant excitation case it shows that the near-field enhancement distributes along edges of the two nano-prisms, which is totally different from the near-field enhancement pattern observed for the case of resonant wavelength excitation in that strong near-field enhancement focuses on corners of the nano-prism and meanwhile locates at the farther side away from the excitation source. It has to be noted that the investigated bowtie nanostructure exhibits dipole mode at resonant peak of 1300 nm, and quadrupole mode resonance at 850 nm. The different signature that the enhanced near-field distribution on the corners for resonant illumination at 850 nm while along the edges for off-resonant at...
700 nm is ascribed to that a higher order excitation of the bowtie structure occurs as the excitation light is blue-shifted to the off-resonant wavelength at 700 nm.

Less prominently visualized images under off-resonant excitation than that for resonant excitation could be attributed to the following reasons. Weak plasmon excitation at 700 nm laser wavelength illumination determines that a much strong local field enhancement can not be expected under this circumstance. Also, since the local field position of off-resonant mode is mainly on the edge of nano-prisms, lightning rod effect, which reinforces local field enhancement for the case of sharp corner of the nano-prism under resonant wavelength excitation, can not be expected either. In addition, the excitation of possible imperfection that introduced from the electron beam lithography process could occur [37]. The appearance of imperfection can enlarge the near-field intensity from several to more than a dozen times [38, 39] and makes it comparable to the field enhancement of the off-resonant excitation. Thus the imperfection induced near-field could show up and affect the visualization effect under off-resonant illumination. As a result, degraded visualization of PEEM images are observed in the case of the off-resonant illumination. It has to be noted that even though visualization of the images for off resonant illumination is lower than the resonant case, the degree of control is still high for off resonant case. A rather high degree of the field control achieved in the off-resonant case is attributed to the fact that the relative strength of the local field induced by both incident polarizations is similar.

The simulated \( |E|^2 \) images of polarization rotation control and coherent control from FDTD simulations supports the measured PEEM images. These images were eventually convolved by a Gaussian filter to account for the finite resolution of the PEEM [17]. As one can see that figures 3(b) and (d) are in excellent agreement with experimental multiphoton PEEM images in figures 3(a) and (c). And also, as shown in figure 4, the simulated results for off-resonant excitation well correspond to the PEEM measurement, the small discrepancy between the simulated and experimental results in figure 4 could be attributed to weak excitation under off-resonant illumination and the excitation of possible imperfection in the nanostructure induced during the lithography process as those discussed in the above.

The experimentally demonstrated control of the near-field localization can be further understood as linear combinations of near-field excited by \( s \) - and \( p \) -polarized light, respectively. Under the excitation of single laser pulse the local electric field can be expressed as [17]:

\[
E^{\alpha}_{\text{loc}}(r, \theta) = E^{\alpha}_{\text{loc},p}(r) \cos \theta + E^{\alpha}_{\text{loc},s}(r) \sin \theta.
\]

where \( E^{\alpha}_{\text{loc},p} \) and \( E^{\alpha}_{\text{loc},s} \) with \( \alpha = x, y, z \) are the local electric fields on bowtie structure excited by \( p \)- and \( s \)-polarized light, respectively. \( E^{\alpha}_{\text{loc}}(r, \theta) \) alternately shows the characters of \( E^{\alpha}_{\text{loc},p} \) (for \( \theta = 0^\circ, 180^\circ \)) or \( E^{\alpha}_{\text{loc},s} \) (for \( \theta = 90^\circ, 270^\circ \)), apart from this, of the superposition of \( E^{\alpha}_{\text{loc},p} \) and \( E^{\alpha}_{\text{loc},s} \) with a certain ratio in the intermediate polarization angle. For the case of polarization rotation, if the polarization angle is turned to \( \theta = 45^\circ \) as an example, the total near-field is:

\[
E^{\alpha}_{\text{loc}}(45^\circ) = \sqrt{2}/2 E^{\alpha}_{\text{loc},s}(r) + \sqrt{2}/2 E^{\alpha}_{\text{loc},p}(r).
\]

For the case of coherent control by two orthogonally polarized laser pulses, the local field response can be expressed as [13]:

\[
E^{\alpha}_{\text{loc}}(r) = \{ G^s(r)A_p + G^p_r(r)A_s \exp [i\Delta \Phi] \} \times \exp [i\varphi_p].
\]

the amplitudes \( A \) describe the extent to which the two far-field polarization propagation, \( \Delta \Phi = \varphi_s - \varphi_p \), corresponds to the relative phase delay of two orthogonal pulses, \( G_i(r) \) determines the local near-field response of the structure with the illumination of two far-field polarization components, \( i = s \) and \( p \). The term in brace reflects a period modulation to the near-field response. If we assume optical path of \( E_p \) is constant and of \( E_s \) is changed, term \( \varphi_p \) is constant, while, the second term in the brace will change relative to \( \Delta \Phi \). Ultimately, the near-field of superposition region between \( E_p \) and \( E_s \) can add constructively or destructively corresponding to \( \Delta \Phi \). For fixed illumination layout, the term of \( \exp [i\varphi_p] \) will not affect the local near-field distribution anymore.

The experimental results in figure 3 show that near-field enhancement as well as its control mainly locate on corners of the nano-prisms for resonant wavelength excitation, while in figure 4 at the edges of the prism for off-resonant case. To qualitatively demonstrate characteristics of the near-field enhancement and control for both the resonant and off-resonant cases, we firstly plotted the simulated \( |E|^2 \) image of the bowtie illuminated by 850 nm resonant and 700 nm off-resonant of \( p \)- and \( s \)-polarized light, respectively, in figure 5. Figures 5(a) and (c) show that the strongly excited near-field mainly locates at the corners of prisms under the illumination of 850 nm, i.e. the near-field distribution is dominated by \( G_1 \) position, meanwhile with much strong field in \( C_1 \) and \( C_2 \) points for \( p \)-polarization laser illumination. The corresponding field focuses at \( C_1 \) and \( C_2 \) positions for \( s \)-polarization excitation. The near-field distribution in \( s \) - and \( p \) -polarization excitation under the 850 nm resonant wavelength implies that near-field distribution and its control on corners of \( C_1 \), \( C_2 \) and \( G_1 \) of the nano-prism are expected in the experiment. On the other side, the near-fields in edge region of the prism dominate for 700 nm excitation for both \( p \) - and \( s \) -polarization cases, therefore, the near-field enhancement and its control
Figure 5. The near-field distribution of $|E_z|$ under illumination of 850 nm resonant (a) and (c) and 700 nm off-resonant (b) and (d) of $p$- (a) and (b) and $s$- (c) and (d) polarized light, respectively. The blue double arrows on the top represent the direction of electric field of illumination light.
same but just with 180° phase shift between the case of \( \varphi_p = 0, \Delta \Phi = 0 \) and \( \varphi_p = \pi, \Delta \Phi = 0 \) (or between the case of \( \varphi_p = 0, \Delta \Phi = \pi \) and \( \varphi_p = \pi, \Delta \Phi = \pi \)) for coherent control. Similar to the case of the control by rotating the polarization angle of single pulse, we can realize the near-field excitation position shift with the inverted phase or unchanged phase. Alternatively, we can invert the phase of the near-field excitation points while keeping its position unchanged. Furthermore, it is seen from figure 6(II) that the phase and position of the excitation points are controlled by \( \varphi_p \) and \( \Delta \Phi \), respectively.

On the other hand, the phase relation of the excitation points can be revealed from the temporal evolution of the electric field on corners of the bowtie structure. Figure 7 shows phase relation among the near-field excitation points corresponding to the results in figure 6. Figure 7(a) shows that the near-field at points of C1 and C2 under the polarization rotation scheme is with inverted phase between \( \theta = 45° \) (\( \theta = 315° \)) and \( \theta = 135° \) or \( \theta = 225° \), while keeps in phase between \( \theta = 45° \) (\( \theta = 135° \)) and \( \theta = 315° \) (\( \theta = 225° \)). Similar results can be obtained from the dynamic evolution of the near-field with coherent control scheme as shown in figure 7(b). Figure 7 intuitively gives phase relations among the near-field points shown in the figure 6.

### 4. Conclusions

Subwavelength imaging and control of localized near-field distribution under resonant and off-resonant excitation within identical gold bowtie structures were demonstrated for the first time. The near-field control was established by two ways, i.e. polarization rotation of single fs laser pulse and coherent control of two orthogonally polarized fs laser pulses. We found that the modulation result to the hotspot under resonant wavelength illumination shows a better visualization in the PEEM image than the case under off-resonant wavelength illumination. The observed photoemission electron pattern in the PEEM images with both control
schemes are well reproduced using finite-difference time-domain simulation. The results of electric near-field distributions for rotation of polarization direction and relative phase delay are explained by using a linear combination of the simulated charge distribution by p- and s-polarized excitations. The demonstrated near-field control has a spatial resolution of nanometer and temporal precision of attosecond. In addition, we can realize the near-field excitation position shift with the inverted phase or unchanged phase. Alternatively we can invert the phase of the near-field excitation points while keeping its position unchanged by rotating the polarization angle of single pulse and coherent control of two orthogonally polarized fs laser pulses, respectively. Our finding for the near-field control of the identical nanostructure under resonant and off-resonant excitation and the phase modulation of the excitation points will pave the way for applications such as sensing, SERS, biomedicine and plasmonic devices.

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