Two-color multiphoton emission for comprehensive reveal of ultrafast plasmonic field distribution

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Abstract
We experimentally demonstrate comprehensive reveal of the ultrafast plasmonic field distribution in a bowtie nanostructure by two-color photoemission electron microscopy (PEEM). We attribute the comprehensive reveal of the field distribution to an effective opening of the two-color quantum channel in multiphoton photoemission, which leads to a dramatic reduction of the nonlinear order (from 4.07 down to 2.01) of the plasmon-assisted photoelectrons and a huge increment of the photoemission yields (typically 20-fold enhancement). Furthermore, we have found that opening extent of the quantum channel strongly related with the photoemission yields generated from one-color 400 and 800 nm laser pulse illumination, and the optimized ratio between the yields for effective opening of two-color quantum channel in our experiment is also achieved. Additionally, benefiting from the high spatial resolution of PEEM, we found there exists a large difference in the nonlinear order of two-color photoemission under the plasmonic excitation within a nanostructure, which has not been reported yet. This work introduces multicolor quantum channel photoemission into the PEEM imaging and offers new way to flexibly control the nonlinear order of the plasmon-assisted photoemission, and it will enable PEEM as a versatile tool in many potential applications.

1. Introduction

Plasmons, which are electromagnetic waves bounded to interfaces related to the collective oscillations of nearly free electrons in the conduction band [1], own the characteristic of strongly near-field enhancement and have been widely used in the field of sensing, biomedical, light cathode, solar cell and so on [2–7]. Mapping of the near-field distribution down to the nanometer scale is a key to understand the basic properties and expand the application of plasmons. Nowadays, several experimental approaches have been utilized to visualize the near-field, including scanning near-field optical microscopy [8, 9], electron energy loss microscopy [10], cathodoluminescence microscopy [11], nonlinear luminescence or fluorescent microscopy [12, 13], nonlinear photopolymerization [14] and so on [15, 16]. Since the amount of electron emission at the position of the excited plasmons is significantly increased due to strong near-field enhancement, the plasmon-mediated photoelectrons from the nanostructures has been used to image the plasmonic near-field distribution [17–19].

Photoemission electron microscopy (PEEM), an instrument that use light as an excitation source to image the near-field of a nanostructure by means of the photoelectrons, has the advantage of high spatial resolution, fast, non-invasive and temporally resolved accessibility, and it has become a powerful tool in advancing near-field characterization of plasmonics [18, 20–23].

Nowadays, lots of researches have been carried out using PEEM to characterize the plasmons directly or to obtain other physical parameters via imaging the plasmonic field, including the measurement of orbital angular momentum of light [24], characterization of the dynamic evolution of plasmons [25–28] and near-field coherent control [29–32]. Strongly near-field enhancement utilized in most of these researches relies on...
lightning rod effects or plasmonic resonance. With further development, weakly excited plasmons (e.g. off-resonant plasmons, high-order plasmons, surface plasmons polaritons, plasmonic Fano resonance or the plasmons supported by non-tip structure) have received great attention [33–37], while revealing those weakly excited plasmons by PEEM is still a challenge due to a lower photoelectron signal. Moreover, in most cases, such kind of weakly excited plasmons and strongly excited one co-exist in a nanostructure, and comprehensive reveal of the plasmonic field distribution by PEEM method becomes more difficult. As the photoemission signal from the weakly excited plasmons is low compared to the value by the strongly excited plasmons or by defects excitation in the PEEM image, only the plasmonic field distribution from the strongly excited region can be presented, whereas the field distribution signal from the weakly excited plasmons is usually lost in the PEEM image [30, 37–39].

The solution to the above problem is lowering the nonlinearity of the emission electrons, thereby enhancing signal intensity from the weakly excited plasmons in PEEM image to make them visible. Deposition of alkali metal (e.g. cesium) on the surface of the plasmonic structure is generally applied for the purpose of lowering the work function of the material required to be overcome by photoelectrons [16, 29, 40]. For example, the work function of gold can be reduced from nearly 5 eV to less than 3.1 eV by vapor deposition of alkali metal, a remarkable reduction of the nonlinear order of photoemission from the nanostructure can be obtained [40]. However, drawback of this method is that the deposition of alkali metal will lead the sample difficult to be applied for potential practical application since it undoubtedly will limit the reusability of the sample once exposed to air due to the highly reactivity of alkali metal [41], and it is not suitable for imaging localized surface plasmons supported by nanostructures due to uneven distribution of the deposited alkali metal on nanoscale. Moreover, plasmonic research is expanding from visible, near-infrared to optical communication waveband and even to THz spectra, the photoelectrons created by those lower energy photons will still suffer from a high-order nonlinear photoemission process even though alkali metal is applied [42–44]. Therefore, comprehensive reveal of the plasmonic field distribution in PEEM image without the introduction of additional material on the sample surface is desired.

In this paper, we propose to effectively open multicolor quantum channel in photoelectron emission to reduce the difference in electron yield between the weakly- and strongly excited plasmons regions for a comprehensive reveal of the ultrafast plasmonic field distribution in two-color excitation PEEM. The results demonstrate that the weakly excited plasmonic field region, whose information is lost under one-color 800 nm laser excitation, has been nicely revealed in the two-color PEEM image without the introduction of additional material on the sample. The improvement in the reveal of plasmonic field distribution is attributed to a dramatic reduction of the nonlinear order of the plasmon-assisted photoelectrons emitted from the nanostructures as the results of the effective opening of the two-color quantum channel in multiphoton photoemission under two-color ultrafast laser excitation [45]. Moreover, the optimized ratio between the photoemission electron yields induced by one-color 400 and 800 nm laser pulse excitation for the effective opening of multicolor quantum channel in the case of our experiment is also discussed. Additionally, we have found there exist a large difference in the nonlinear order of the two-color photoemission under the plasmonic excitation within a nanostructure, which has not been reported in the previous investigations on the two-color photoemission electron generation [46, 47].

The photo-emitted electrons from noble-metal nanostructures under visible or near-infrared light illumination typically undergo multiphoton photoemission with incident laser field intensity in the perturbation regime [46, 48, 49]. Figure 1 shows the schematic diagram of the photoemission process from gold under the illumination of one-color 800 nm pulse and two-color 400 nm + 800 nm pulses. For one-color 800 nm pulse excitation, the electrons located near the Fermi level of gold will be stimulated to photoelectron via intermediate states with simultaneously absorbing of 4 photons (work function $E_w = 3\hbar\nu < E_w < 4\hbar\nu$). Under this condition, the photoemission yield follows $P \propto E_z^8$, in which $E_z$ corresponds to the $z$ component of the electric field generated by 800 nm pulses illumination. Two-color excitation induced photoemission, which implies a multicolor quantum process, can emit through three quantum channels: the electron interacts with the photons of 800 nm, with the photon of 400 nm, or particularly with both. Two-color quantum channel as the latter case is multiphoton transitions process in which photons of different colors are simultaneously absorbed or emitted [46]. In this case, the dependence of electron yield upon $E_z @800$ nm is dramatically reduced to $P \propto E_z^4$. Furthermore, as the plasmonic resonance of gold nanostructure can be exclusively adapted to the wavelength of 800 nm, the nonlinear order variation of the 800 nm-induced photoelectron will strongly affect the contrast ratio of the plasmonic PEEM image and, therefore, a PEEM image which reveals the comprehensive near-field information of the plasmons is expected to be obtained through two-color excitation.
2. Experimental setup

The experimental setup is shown in figure 2. PEEM (FOCUS, IS-PEEM-DLD) with better than 40 nm spatial resolution was used to image the near-field distribution of a bowtie nanostructure under ultrafast laser illumination. A femtosecond Ti-Sapphire laser oscillator (Coherent, Mira 900), delivering pulses with a duration of 200 fs, tunable output wavelength (from 680 to 900 nm), single pulse energy of 13 nJ and a repetition rate of 76 MHz was used in the experiment. We generate fundamental and second harmonic pulses by passing the output of the Ti-Sapphire laser through a 500 μm thick β-barium borate (BBO) crystal. Central wavelengths for the fundamental and the second harmonics are 800 nm and 400 nm, respectively. Intensities of these two laser pulses can be adjusted independently by neutral density filter wheels. The sample was illuminated at a 65° to the normal of the substrate. The polarization direction of the femtosecond laser pulse was varied using half-wave plate. Throughout the experiment, polarization direction of 400 nm laser pulse was always maintained p-polarized direction to obtain relatively high PE yield. The two-color laser pulses were separated with the steps of several femtoseconds by a Mach–Zehnder interferometer and was focused onto the sample by a quartz lens with the focus length of 300 mm. The phase between the two colors pulses is not measured in our experiment. Fabrication of the gold bowtie nanoantenna was realized via electron beam lithography technology. The antenna consists of two triangular nano-prisms with a thickness of 40 nm, equilateral sides with a length of 350 nm and a gap distance of 100 nm. The substrate of the sample is 1 mm thick silica and there is an ITO conducting layer between silica and gold nanostructure.

The scanning electron microscopy image of the bowtie is shown in the left part of figure 2. The investigated bowtie nanostructure exhibits quadrupole mode resonance peak at 825 nm for simulated absorbing spectrum. It is noticed in the figure 2(I) that the plasmonic mode excitation at 800 nm can be still very strong even though the excitation wavelength is off-resonant peak of the structure. PEEM image of the bowtie nanostructure with 400 nm excitation only is shown in the inset (III) of figure 2. The electron emission across the gold structure surface shows a high yield over the ITO substrate due to higher density of states of gold. Moreover, higher photoemission yields from the corners of bowtie nanostructure can also be observed due to the near-field enhancement of lightning rod effects. In the experiment, delay line detector (DLD) was used to accurately obtain the number of photoemission electrons.

3. Results and discussion

Time delay measurement of the photoemission electron yield from three interested positions as marked in figure 2 under two-color illumination was firstly investigated. The temporal–resolved PE yield exhibits an obvious peak in the time delay range as shown in figure 3, which is attributed to the opening of the multicolor

2 See supplemental material available online at stacks.iop.org/NJP/20/073031/mmedia.
quantum channel under two-color multiphoton emission as discussed in the above section. The opening of a multicolor quantum channel lead to a nearly twofold increase in photoemission yield for all the investigated positions, while the PE yields among the selected positions in the structure are largely different (the number of electrons emitted from G1 is about 20 times larger than the case from C1 or M1 under two-color light illumination). We attribute the PE yield difference of those three hot spots to stronger near-field enhancement in G1 than the case in C1 or M1 for p-polarized 800 nm pulse illumination [30, 50]. The significant difference in photoemission yield fluctuation among the selected positions can also be noticeable from the figure. It clearly shows that the fluctuation amplitude of the curve decreases in turns from G1, C1 to M1. We choose the time delay corresponding to maximum photoelectron yield as the zero point of two-color pulses in the following experiment. At this time delay, maximum photoelectrons yields can be predicted with the opening of two-color quantum channel [46].

The near-field distribution of bowtie structure presented by PEEM image and FDTD simulation are shown in figure 4. It can be seen from figure 4(a) that the hot spots are concentrated at the corners of the structure and the image contrast between the G1 corners and the surface of bowtie is much larger for the case under the excitation of 800 nm than that under 400 nm excitation (see the inset III of figure 2), which is due to the excitation of plasmons as well as higher nonlinear order of photoemission under 800 nm illumination. Hot spots at G1 and at C1, C2 are visible when laser polarization angle \( \theta \) is set at 0°. Furthermore, hot spots shifting among the corners of the bowtie with the variation of laser polarization direction are also observed. Plasmonic field in the bowtie that represented by those hot spots in the PEEM images seems to be well revealed by PEEM images. Meanwhile, figure 4(c) displays the near-field distributions with single 800 nm excitation by FDTD simulations, and the strong near-field distribution in corners is in excellent agreement with the PEEM images in figure 4(a). However, near-field enhancement in the edges of bowtie structure as marked by green ellipses can clearly be seen in the simulation while the corresponding local electron emission cannot be observed in the PEEM images under one-color 800 nm laser pulse illumination as shown in figure 4(a).

To fully reveal the plasmonic field distribution in the bowtie PEEM image, two-color laser excitation is performed, and the results are shown in figure 4(b). The PEEM images clearly show that, accompanying with the hot spot in the corners, excitation in the edge of nanostructure that does not show up in the PEEM images under
Figure 3. PE yield of two-color multiphoton emission. The temporal overlap of the two-color pulses opens the two-color quantum channels of G1, C1 and M1. We define $\tau = 0$ at maximum counts in the temporal overlap. The negative value of x-axis corresponds to 400 nm laser pulse irradiating the sample surface earlier than 800 nm laser pulse. The intensities of 800 and 400 nm laser pulses are 23 MW cm$^{-2}$ and 0.5 MW cm$^{-2}$, respectively. The PEEM image of bowtie in this figure corresponds to zero delay between the two-color pulses.

Figure 4. Near-field distribution of the bowtie nanostructure for different polarization of the incoming 800 nm pulse. Multiphoton PEEM images of the bowtie nanostructure under one-color 800 nm (a) and two-color 400 nm + 800 nm (b) illumination. FDTD simulated electric field magnitude of the plasmonic near-field distributions on the top surface of the bowtie nanostructure excited by 800 nm light (c). The polarization direction is indicated by double blue arrow as shown on the top and changed from 0$^\circ$ to 180$^\circ$ by a step of 45$^\circ$. The outline of the bowtie is shown by the gold dashed lines to guide the eye. Green ellipses, magenta and green cycles are used to label the position of the weakly excited plasmons on the edge of the bowtie nanostructure. The laser intensity of 800 nm pulse used in (a) and (b) is 19.7 MW cm$^{-2}$ and the laser intensity of 400 nm pulse used in (a) is 0.6 MW cm$^{-2}$. 
one-color 800 laser pulse illumination are clearly revealed. With variation of the polarization angles of 800 nm pulse, a good agreement at the weak excitation edges between the PEEM images under two-color light excitation and the FDTD simulation is reached. Note that the polarization dependence of photoemission in the edge of nanostructure under two-color excitation indicates the local excitation of the plasmons controlled by 800 nm laser. Compared to the PEEM images under one-color 800 nm excitation, an improved consistency between the near-field PEEM image under two-color excitation and the FDTD simulated plasmonic near-field distribution for the whole nanostructure demonstrates that we have succeeded in the comprehensive reveal of plasmonic field distribution in a nanostructure by means of two-color scheme.

To elucidate the underlying mechanism that responsible for the consistence between two-color excited PEEM images and simulated near-field distribution of plasmons, we record the dependence of photoemission yields from the interested points of G1, C1 and M1 upon the intensity of p-polarized 800 nm pulse for one-color 800 nm and two-color (800 nm + 400 nm) illumination conditions, respectively, and the results are shown in figure 5. It can be seen that the slopes for the positions of G1, C1 and M1 under one-color 800 nm laser pulse illumination are 4.00, 4.08 and 4.07, respectively, meaning a 4-photon photoemission process. With the introduction of p-polarized 400 nm light, it is found that the slopes of the investigated three positions decrease: from 4.00 down to 3.12 for G1, from 4.08 down to 2.50 for C1 and, in particular, the slope of M1 decreases significantly from 4.07 to 2.01 (with a reduction of 2.06) as marked by the color arrows in figure 5. The results obtained from figures 4 and 5 indicate that a large nonlinear order difference exists in two-color photoemission under the plasmonic excitation within a tiny area of the nanostructure and it has been well disclosed assisted by the high spatial resolution of PEEM.

Furthermore, it can also be seen from figure 5 that reduction of the slope is closely related to the increment of the photoelectron yields from one-color to two-color excitation. Typically, it is observed that the electron yield of position M1 under two-color excitation is about 20-fold higher than that under one-color 800 nm pulse excitation. Meanwhile, for G1 and C1 positions, the electron yield increases by only about 5.6 and 2.3 times. These results show that the larger drop in slope, the higher ration of electron yield increment for the position. We attribute this photoelectron yield increment to higher photoemission probability of 2PPE process for the position M1 under two-color illumination than a 4PPE process under one-color irradiation [46, 51].

The 2PPE process observed for the position M1 indicates an occurrence of the effective opening of multicolor quantum channel in the photoemission, which leads to a significant reduction of the nonlinear order and a huge enhancement of the photoemission from M1 [46, 47]. As a result, the discrepancy of photoelectron yield among M1 and C1, G1 under two-color light illumination is greatly reduced accompanying with a clear visibility of photoemission electron signal at M1 position in PEEM images as shown in figure 4(b). The slope variation range of the three interested positions indicates that the opening of two-color quantum channel in photoemission electron relates with the local PE yield induced by one-color 800 nm excitation. In the meantime, the effect of the intensity of 400 nm pulse upon the slope variation of the position (here take G1 as an example) is given in the inset of figure 5. The slope of G1 decreases from 3.84 to 3.44 when the intensity of 400 nm pulse changes from 0.83 to 2.17 MW cm$^{-2}$ in the two-color scheme (varying the 800 nm laser intensity for obtaining

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**Figure 5.** Intensity dependence of one- and two-color multiphoton photoemission. The signal is plotted against the intensity of p-polarized 800 nm pulse and the photoemission yield exclusively contributed by one-color 400 nm illumination was deducted from the data. Dashed lines representing the slope magnitude are guides to the eye. The color arrows represent the change of the nonlinear order of photoelectrons from 800 nm excitation only to two-color excitation at the same location. The intensity of 400 nm is 5.26 MW cm$^{-2}$. 

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the slope), meaning a good trend towards the effective opening of the two-color quantum channel in photoemission at G1 position with the intensity increase of 400 nm pulse. The reduction of the slope accompanies with an increment of the PE yield under two-color excitations. Furthermore, we fixed 800 nm laser intensity at 79 MW cm$^{-2}$ and varied the 400 nm laser intensity from 2.6 to 8.3 MW cm$^{-2}$ and found that photoelectron yield enhanced with the increasing of 400 nm laser intensity, corresponding to the measured nonlinear orders (slope of the PE yield versus laser intensity curve) of 1.3, 1.5 and 1.6 at G1, C1 and M1 positions, respectively. This result demonstrates that opening quantum channel can also be reached in the case of varying 400 nm laser intensity. Therefore, we affirm that the variation of slope (the extent of opening of the two-color quantum channel) strongly relates with the PE yields generated from one-color 800 and 400 nm excitation.

The two-color quantum channel induced slope variation in photoemission electron yield versus 800 nm laser intensity under two-color laser pulse excitation (variation in nonlinear order of photoemission process) can be used to explain the fluctuation difference in the electron emission yield among G1, C1 and M1 positions as observed in figure 3. It is noted that the fluctuation amplitudes for the curves of G1, C1 and M1 positions are roughly 23%, 19% and 15%, respectively. For nonlinear photoemission process, a large fluctuation in the photoemission yield can be attributed to a slight variation of incident laser power. For example, a 20% change in the photoemission yield in 3 photon process (e.g. the nonlinear order of point G1 reduces from 4 to 3.12 when changing the excitation from only 800 nm to two-color scheme) corresponds to only a 0.8% variation of the incident light intensity (the variation magnitude of the incident laser power is reasonable to our experimental setup) with a 10-fold near-field enhancement induced by plasmonic resonance effect. Furthermore, the figure shows that fluctuation amplitudes of G1(23%), C1 (19%) and M1 (15%) reduce in turn. This can be attributed to different nonlinear orders of photoemission of G1 (3.12), C1 (2.5) and M1 (2.01) positions, resulting from different extent of two-color quantum channels opening in photoemission under two-color laser pulse irradiation.

We have shown that the control of the nonlinear order of the excited electrons induced by plasmons can be achieved by changing the excited laser pulses intensity. On the other hand, as the energy gap between the intermediate state and the vacuum level (that can be controlled by the photon energy of non-plasmon-excited light) as shown in figure 1(b) determines the nonlinear order of the plasmon-assisted photoemission, change of the wavelength of the non-plasmon excited light in the multicolor quantum channel can be employed to control the nonlinear order of the plasmon-assisted photoemission. In principle, the multicolor quantum channel technology can provide a flexible way to control the nonlinear order of plasmon-assisted photoemission electron via varying the wavelength of the non-plasmon excited light.

In the following, we will get deep insight into the relation between the opening extent of two-color quantum channel and the photoelectron yields. Photoemission coupling degree $E$, which reflects the coupling efficiency between the electrons and two-color laser light, is hereby introduced

$$E = (P_{400+800} - P_{800} - P_{400}) / (P_{400} + P_{800}),$$

where $P_{400+800}$ represents the sum number of photoelectrons at a specific location under the illumination of two-color laser pulse, corresponding to the total electrons emitted through three quantum channels that cover two-color quantum channel, 800 and 400 nm single channel; $P_{800}$ and $P_{400}$ represents the number of photoelectrons emitted at the same location through only 800 nm or 400 nm single channel. $E = 0$ means the two-color quantum channel is closed and no extra electron yield is brought by the overlapping of two-color pulses. When $E$ is close to 0, it means a lower ratio of the emitted electrons induced by synergism effect of the two-color light in the total photoemission yield, i.e., the increase of PE yield or the reduction of the slope under two-color excitation is small as result of low extent of opening of two-color quantum channel. On the contrary, if the ratio is large, the increment of photoelectron yields will be high, which corresponds to an effective opening of two-color quantum channel with a significant coupling between electrons and two-color light in photoemission. The $E$ values of M1, C1 and G1 positions is 2.8, 1.8 and 1.2, respectively, under two-color excitation with the intensity of 23 MW cm$^{-2}$ for 800 nm and 0.5 MW cm$^{-2}$ for 400 nm. The lower $E$ value of G1 and C1 positions, compared to that of M1, results from the fact that the electron yield of C1 and G1 under one-color 800 nm pulse illumination is much higher than the case under one-color 400 nm pulse illumination (the measured resonant peak of the bowtie is at 850 nm, and 800 nm is close to the peak); on the contrary, the difference in photoemission yield under one-color 400 and 800 nm illumination for M1 is much smaller. Therefore, a much effective opening of two-color quantum channel represented by a drastic reduction of slope and increment of photoemission yield for M1 over the positions of G1 and C1 have been obtained as validated in figure 5. Moreover, it can be concluded from figure 5 that the extent of quantum channel opening (slope discrepancy) among the positions under two-color excitation relies on the different ratios of the photoemission yield under one-color 400 and 800 nm illumination, i.e. $P_{800}$ versus $P_{400}$. This conclusion is further supported by the inset of figure 5: a linear reduction of the slope of G1 can be observed with the increment of $P_{400}$ as the rise of 400 nm laser intensity under two-color excitation.
The optimal ratio of $P_{400}$ and $P_{800}$ for the effective opening of multicolor quantum channel in photoemission from the identical position (taking M1 as example) can be explored based on the equation (1). Figure 6 shows the $E$ factor of M1 under different intensities of 400 nm illumination. The factor $E = 2$, as shown by dashed green line for guiding the eye, is taken as an example for analyzing the conditions for effective opening of multicolor quantum channel. It can be seen that the curve shifts to higher intensity of 800 nm laser pulse with the increase of 400 nm laser intensity for keeping the same $E$ factor (see asterisk points). To further clarify the condition which gives rise to the highest photoemission yield increment, we record the ratio of $P_{400}$ to $P_{800}$ at the maximum $E$ factor (marked by arrows in figure 6) and display in the inset of figure 6. The inset clearly shows that the value range of $P_{400}/P_{800}$ rough from 1.9 to 2.7 corresponds to the maximal $E$ factors under the various intensities of 400 nm light illumination. i.e., in our experiment, effective opening of two-color quantum channel is achieved when the photoemission yield under 400 nm pulse illumination is roughly two times higher than that of 800 nm case. The obtained larger photoelectron yield required by 400 nm excitation than that by 800 nm light for effective opening of the two-color quantum channel reflects the fact that enough electrons should be populated in the intermediate state by the 400 nm light as prerequisite for the rest two 800 nm photons to free the electron in the channel.

Note that the obtained photoelectron ratio of rough 2 between 400 and 800 nm light excitation for the effectively opening of quantum channel may only be applicable to the above specific conditions, and a different ratio number in other experiments can be reached when following the proposed $E$ factor method. This is because we believe that the opening degree relates with material work function, excitation wavelength of laser (corresponding to nonlinear order of photoelectron emission), lifetime of the energy band of material and so on. However, it is predictable that the introduced $E$ factor method here will certainly give rise to an optimized, sample and excitation wavelength-dependent photoelectron ratio between different excitation wavelengths that corresponds to effective opening of the two-color quantum channel.

4. Conclusions

In summary, we have demonstrated comprehensive reveal of ultrafast plasmonic field distribution in bowtie structures through two-color photoemission in PEEM. The comprehensive reveal of the plasmonic field distribution is attributed to effective opening of the multicolor quantum channel in photoemission, which leads to a reduction of the nonlinear order of the plasmon-assisted photoelectrons from the nanostructure. The combining of multicolor quantum channel with plasmons is expected to provide a flexible way to control the nonlinear order of photoemission electron and the PE yield, which is benefit for expanding plasmonic research from visible, near-infrared to optical communication waveband and even to THz spectra. Furthermore, we have found a large difference in the nonlinear order of two-color photoemission under the plasmonic excitation within a nanostructure and this will be extremely important for the design of plasmon-assisted nanoscale electron pulse source for free electron laser and photocathode used in compact coherent x-ray production.
Additionally, two-color excited approach, which eliminates the necessary of introducing alkali metal, keeps the sample in its original status throughout the PEEM experiment and is superior in terms of the reusability of the sample for a further characterization or potential applications. The two-color femtosecond laser excitation photoemission together with ultra high spatial-temporal resolved PEEM can be applied, not limited to the area of plasmonic field PEEM imaging, to the investigation of above-threshold photoemission \[47, 54\] and hot-carrier dynamics \[2, 3, 35, 36\], to the control of interference fringes in the momentum distribution of electron emission \[57\], and further to many other potential applications \[51, 58–60\].

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### References

1. Maier S A 2007 Plasmonics: Fundamentals and Applications (New York: Springer Science & Business Media)
2. Clavero C 2014 Plasmon-induced hot-electron generation at nanoparticle/metal-oxide interfaces for photovoltaic and photocatalytic devices Nat. Photon. 8 95–103
3. Brongersma M L, Halas N J and Nordlander P 2015 Plasmon-induced hot carrier science and technology Nat. Nanotechnol. 10 25–34
4. Lal S, Link S and Halas N J 2007 Nano-optics from sensing to waveguiding Nat. Photon. 1 641–8
5. Atwater H A and Polman A 2010 Plasmonics for improved photovoltaic devices Nat. Mater. 9 865
6. Zeng S, Ballargeat D, Ho H-P and Yong K-T 2014 Nanomaterials enhanced surface plasmon resonance for biological and chemical sensing applications Chem. Soc. Rev. 43 3426–52
7. Atwater H A 2007 The promise of plasmonics Sci. Am. 296 56–62
8. Rang M, Jones A C, Fei Z, Li Z Y, Wiley BJ, Younan X and Raschke M B 2008 Optical near-field mapping of plasmonic nanoprisms Nano Lett. 8 3357–63
9. Imura K, Ueno K, Misawa H and Okamoto H 2011 Anomalous light transmission from plasmonic-capped nanopores Nano Lett. 11 960–5
10. Koh A L, Fernandez-Dominguez A, Maier S, Yang J and McComb D 2011 High resolution mapping of electron-beam-excited plasmon modes in lithographically-defined gold nanostructures Nano Lett. 11 764–5
11. Das P and Chinti T K 2014 Substrate induced symmetry breaking in penta-twinned gold nanorod probed by free electron impact J. Phys. Chem. C 118 26284–91
12. Imura K, Okamoto H, Hossain M K and Kitajima M 2006 Visualization of localized intense optical fields in single gold-nanoparticle assemblies and ultrasensitive raman active sites Nano Lett. 6 2173–6
13. Ueno K, Juodkazis S, Mizeikis V, Sasaki K and Misawa H 2008 Clusters of closely spaced gold nanoparticles as a source of two-photon photoluminescence at visible wavelengths Adv. Mater. 20 26–30
14. Ueno K, Juodkazis S, Shibuya T, Yokota Y, Mizeikis V, Sasaki K and Misawa H 2008 Nanoparticle plasmon-assisted two-photon polymerization induced by incoherent excitation source J. Am. Chem. Soc. 130 6928–9
15. Garcia De Abajo F J 2010 Optical excitations in electron microscopy Rev. Mod. Phys. 82 209–75
16. Douillard L and Charra F 2011 High-resolution mapping of plasmonic modes: photoemission and scanning tunneling luminescence microscopies J. Phys. D: Appl. Phys. 44 464002
17. Hobbs R G, Putnam W P, Fallahi A, Yang Y, Kärtner F X and Berggren K K 2017 Mapping photoemission and hot-electron emission from plasmonic nanoantennas Nano Lett. 17 6069–76
18. Cinchetti M, Gloskovskii A, Nepitolo S A, Schönhhense G, Rochholz H and Kreiter M 2005 Photoemission electron microscopy as a tool for the investigation of optical near fields Phys. Rev. Lett. 95 046101
19. Losquin A and Lummen T A 2017 Electron microscopy methods for space-, energy-, and time-resolved plasmonics Front. Phys. 12 1–27
20. Weber N R, Escher M, Merkel M, Oelsner A and Schönhhense G 2008 Energy- and time-resolved microscopy using PEEM: recent developments and state-of-the-art J. Phys.: Conf. Ser. 100 072031
21. Word R, Fitzgerald J and Könenkamp R 2013 Direct coupling of photonic modes and surface plasmon polaritons observed in 2-photon PEEM Opt. Express 21 1175–81
22. Word R C and Könenkamp R 2017 Photonic and plasmonic field distributions characterized with normal- and oblique-incidence multi-photon PEEM Ultramicroscopy 183 1339–51
23. Petek H 2017 Photoemission electron microscopy: photovoltaics in action Nat. Nanotechnol. 12 3–4
24. Spektor G et al. 2017 Revealing the subfemtosecond dynamics of orbital angular momentum in nanoplasmonic vortices Science 355 1187–91
25. Sun Q, Yu H, Ueno K, Kubo A, Matsuo Y and Misawa H 2016 Dissecting the few-femtosecond dephasing time of dipole and quadrupole modes in gold nanoparticles using polarized photoemission electron microscopy file ACS Nano 10 3835–42
[26] Kubo A, Onda K, Petek H, Sun Z, Jung Y S and Kim H K 2005 Femtosecond imaging of surface plasmon femtosecond imaging of surface plasmon dynamics in a nanostructured silver film Nano Lett. 5 1124–7
[27] Märsell E et al 2015 Nanoscale imaging of local few-femtosecond near-field dynamics within a single plasmonic nanoantenna Nano Lett. 15 6601–8
[28] Qin J, Ji B, Hao Z and Lin J 2015 Probing of ultrafast plasmon dynamics on gold bowtie nanostructure using photoemission electron microscopy Chin. Phys. Lett. 32 1–5
[29] Aeschlimann M, Bauer M, Bayer D, Brixner T, De Abajo F J G, Pfeiffer W, Rohmer M, Spindler C and Steeb F 2007 Adaptive subwavelength control of nano-optical fields Nature 446 301–4
[30] Ji B, Qin J, Tao H, Hao Z and Lin J 2016 Subwavelength imaging and control of ultrafast optical near-field under resonant- and off-resonant excitation of bowtie nanostructures New J. Phys. 18 093046
[31] Hrelescu C, Saur T K, Rogach A L, Frank J, Laurent G and Douillard I 2011 Selective excitation of individual plasmonic hotspots at the tips of single gold nanostars Nano Lett. 11 402–7
[32] Yu H, Sun Q, Ueno K, Oshikiri T, Kubo A, Matsuo Y and Misawa H 2016 Exploring coupled plasmonic nanostructures in the near field by photoemission electron microscopy ACS Nano 10 10373–81
[33] Beaucourt S et al 2017 Attosecond-resolved photoionization of chiral molecules Science 358 1288–94
[34] Bouabé E, Lachaine R and Meunier M 2012 Plasma mediated off-resonance plasmonic enhanced ultrafast laser-induced nanocavitation Nano Lett. 12 2763–9
[35] Razinskas G, Kilbane D, Melchior P, Geiser P, Krauss E, Mathias S, Hecht B and Aeschlimann M 2016 Normal-incidence PEEM imaging of propagating modes in a plasmonic nanocircuit Nano Lett. 16 6832–7
[36] Luk’yanchuk B, Zheludnev I., Maier S A, Halas N J, Nordlander P, Giessen H and Chong C T 2010 The Fano resonance in plasmonic nanostructures and metamaterials Nat. Mater. 9 707–15
[37] Ji B, Wang Q, Song X, Tao H, Dou Y, Gao X, Hao Z and Lin J 2017 Dissecting dark mode of femtosecond plasmon with photoemission electron microscopy J. Phys. D: Appl. Phys. 50 415309
[38] Yu H, Sun Q, Yang J, Ueno K, Oshikiri T, Kubo A, Matsuo Y, Gong Q and Misawa H 2017 Near-field spectral properties of coupled plasmonic nanoparticle arrays Opt. Express 25 707–15
[39] Sun Q, Ueno K, Yu H, Kubo A, Matsuo Y and Misawa H 2013 Direct imaging of the near field and dynamics of surface plasmon resonance on gold nanostructures using photoemission electron microscopy Light Sci. Appl. 2 e118
[40] Lemke C et al 2013 Spatiotemporal characterization of SPP pulse propagation in two-dimensional plasmonic focusing devices Nano Lett. 13 1053–8
[41] Shackelford J F and Alexander W 2001 CRC Materials Science and Engineering Handbook vol 49 (Boca Raton, FL: CRC Press)
[42] Märsell E, Larsen E W, Arnold C L, Xu H, Mauritsson J and Mikkelsen A 2015 Photoemission electron microscopy of localized surface plasmons in silver nanostructures at telecommunication wavelengths J. Appl. Phys. 117 083104
[43] Zou S, Bao Y and Fang Z 2016 Planar plasmonic chiral nanostructures Nanoscale 8 3900–5
[44] Liu Z, Li J, Liu Z, Li W, Li J, Gu C and Li Z Y 2017 Fano resonance Rabi splitting of surface plasmons Sci. Rep. 7 80101
[45] Yamagawa K, Shibuta M and Nakajima A 2017 Two-photon photoelectron emission microscopy for surface plasmon polaritons at the Au(111) surface decorated with alkane-thiolate self-assembled monolayers Phys. Chem. Chem. Phys. 19 13455–61
[46] Cheng-Wei Huang W, Becker M, Beck J and Batelaan H 2017 Two-color multiphoton emission from nanotips New J. Phys. 19 023011
[47] Förster M, Paschen T, Krüger M, Lemell C, Wachter G, Libisch F, Madlener T, Burgdorfer T and Hommelhoff P 2016 Two-color coherent control of femtosecond above-threshold photoionization from a tungsten nanotip Phys. Rev. Lett. 117 217601
[48] Schertz F, Schmelzeisen M, Kreiter M, Elmers H J, Schönhense G 2012 Field emission of electrons generated by the near field of strongly coupled plasmons Phys. Rev. Lett. 108 236602
[49] Dombi P, Horl A, Rácz P, Mártón I, Trügler A, Krenn J R and Hohenester U 2013 Ultrafast strongly field photoemission from plasmonic nanoparticles Nano Lett. 13 674–8
[50] Melchior P, Bayer D, Schneider C, Fischer A, Rohmer M, Pfeiffer W and Aeschlimann M 2011 Optical near-field interference in the excitation of a bowtie nanoantenna Phys. Rev. B 83 235407
[51] Polyakov A, Senti C, Thompson K F, Feng J, Cabrini S, Schuck P J, Padmore H A, Peppernick S J and Hess W P 2013 Plasmon-enhanced photocathode for high brightness and high repetition rate x-ray sources Phys. Rev. Lett. 110 076802
[52] Dowell D H, Bazarov I, Dunham B, Harkay K, Henderson – García C, Legg R, Padmore H, Rao T, Smedley J and Wan W 2010 Cathode R&D for future light sources Nucl. Instrum. Methods Phys. Res. A 622 685–97
[53] Graves W S, Kärtner F X, Moncton D E and Piot P 2012 Intense superradiant x rays from a compact source using a nanocathode array and emittance exchange Phys. Rev. Lett. 108 263604
[54] Schenk M, Krüger M and Hommelhoff P 2010 Strong-field above-threshold photoionisation from sharp metal tips Phys. Rev. Lett. 105 257601
[55] Cushing S K 2017 Plasmonic hot carriers skip out in femtoseconds Nat. Photon. 11 10–1
[56] Gruson V et al 2016 Attosecond dynamics through a Fano resonance: monitoring the birth of a photoelectron Science 354 734–8
[57] Xie X et al 2012 Attosecond probe of valence-electron wave packets by subcycle sculpted laser fields Phys. Rev. Lett. 108 193904
[58] Li R K, To H, Andonian G, Feng I, Polyakov A, Scoby C M, Thompson K, Wan W, Padmore H A and Musumeci P 2013 Surface-plasmon resonance-enhanced multiphoton emission of high-brightness electron beams from a nanostructured copper cathode Phys. Rev. Lett. 110 074801
[59] Bhat R D R and Sipe J E 2000 Optically injected spin currents in semiconductors Phys. Rev. Lett. 85 5432
[60] Hübner J, Rühle W W, Klade M, Hommel D, Bhat R D R, Sipe J E and van Driel H M 2003 Direct observation of optically injected spin-polarized currents in semiconductors Phys. Rev. Lett. 90 216601