

Scanning-less optical near-field characterization for structured light fields based on nonlinear effects

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Conventional methods for near-field characterization have typically relied on the nanoprobe to point-scan the field, rendering the measurements vulnerable to external environmental influences. Here, we study the direct far-field imaging of the near-field polarizations based on the four-wave mixing effect. We construct a simulation model to realize the instantaneous extraction of the near-field distributions of a wide range of structured light fields, such as cylindrical vector vortex beams, plasmonic Weber beams, and topological spin textures, including photonic skyrmions and merons. This method is valuable for the studies on manipulation of structured light fields and light-matter interaction at the micro/nano scales.

Keywords: scanning-less optical near-field detection; near-to-far-field transform; nonlinear effect; structured light field; topological quasi-particle.

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1. Introduction

The technique of characterizing the optical field in the near-field region, as exemplified by near-field scanning optical microscopy (NSOM)^[1], has been extensively employed for investigating surface plasmon polaritons (SPPs)^[2,3], surface wave conduction of inorganic materials^[4], photoionization of two-dimensional materials^[5,6], near-field photocurrents^[7,8], and semiconductor carrier concentration^[9,10]. In nanophotonics, this technique provides a means to observe and study the physical properties of light fields at micro/nano scales, which exhibit distinct characteristics compared to their macroscopic counterparts. Moreover, it offers valuable insights into unique phenomena that emerge from interactions between light and micro/nanostructures. Consequently, this technique plays a pivotal role in investigating novel physical phenomena and advancing innovative applications.

However, the conventional imaging process of NSOM requires point-by-point scanning of the nanoprobe, which prolongs the imaging time and increases the system's complexity, rendering the measurements vulnerable to external environmental influences, such as platform vibration and temperature fluctuations during the scanning process^[11]. In addition, the intrusion of the nanoprobe into the evanescent field can disturb the field to some extent, affecting the accuracy of measurements^[12]. Moreover, the nanoprobe may collide with the sample during the scanning process when the measured area is not

sufficiently flat, leading to potential damage to the sample and reduced lifespan of the nanoprobe. As a result, such methods are unsuitable for samples with time-varying light fields and complicated nanostructures.

Recently, a method for converting near-field optical information to far-field radiation based on the nonlinear effect has been proposed^[13,14]. This mechanism is different from the traditional NSOM, which needs to introduce nanoprobe in the near-field optical field to realize the scattering of the near-field information to the far-field radiation. Instead, the near-field information can be encoded into the nonlinear signal propagating in the far field, and the direct detection of the near-field optical field information in the far field can be realized. Moreover, compared to the NSOM's inability to differentiate different polarizations, this nonlinear near-field characterization method can extract different in-plane near-field polarizations to reconstruct the 3D electric field vectors and photonic spin vectors.

In the realm of light field manipulation, a wide range of complicated structured light fields has been discovered continuously. In particular, recent discoveries of photonic topological quasi-particles, such as skyrmions^[15,16], merons^[17,18], and hopfions^[19,20], have garnered significant interest. Due to the stabilities protected by topology and deep-subwavelength features, the photonic-skyrmion-related topological spin textures have demonstrated potential applications in optical information storage^[21], picometre metrology^[22], and magnetic domain detection^[23]. The investigation and manipulation of the

complicated structured light field exemplified by the photonic skyrmion necessitate the development of a near-field characterization technique capable of measuring time-varying light fields and different polarizations.

In this Letter, we investigate the direct far-field imaging of the near-field polarization utilizing the four-wave mixing effect, which can be applied in the scanning-less near-field characterization of the structured light fields. We construct a simulation model through the analysis of nonlinear coefficients, and the transformation from the near-field polarization distributions to the far-field nonlinear signals has been realized for a wide range of complicated structured light fields, such as cylindrical vector vortex beams, plasmonic Weber beams (PWBs), and topological spin textures, which includes photonic skyrmions and merons. It provides a simulation tool to guide further scanning-less near-field characterization experiments in the future, which is of great value to the dynamic observations and manipulations of the structured light fields, as well as the nonradiative optical modes such as bound states in the continuum (BIC)^[24,25]. Moreover, this constructed simulation model extends beyond the four-wave mixing nonlinear process and can be utilized for the guidance of a wide range of nonlinear interactions involving multiple sources, such as the sum frequency and difference frequency generations and the related investigations.

2. Method

The scanning-less imaging capability of nonlinear optical near-field microscopy (NNOM) stems from a nonlinear process known as partially degenerate four-wave mixing (FWM). In the FWM process, the nonlinear signal photons are generated from the interaction between two pump light photons and the near-field photons, exemplified by the SPPs at the metal-dielectric interface. This process transforms the spatial

information stored in a near-field wave pattern into nonlinear signals propagating in free space, which can then be directly captured by a CCD. An illustration of this nonlinear interaction is shown in Fig. 1(a). The pump light (800 nm) is approximately vertically incident on the gold film, which is focused by a long working-distance objective lens to increase power density. This area is exactly the location of the SPP field that is generated via the process where the excitation light (1030 nm) is focused with an objective lens and then illuminates the gratings. When the wave vector matching condition is satisfied, the nonlinear polarization field radiates to the far field and is collected by the same long-working-distance objective and separated by the dichroic mirror from the background. At last, the nonlinear signal could be collected with a CCD.

In the nonlinear near-field characterization process involving the SPPs, the photon momentum conservation and transverse wave vector matching conditions in the FWM process can be described as^[26]

$$\omega_{\text{nl}} = 2\omega_{\text{pump}} - \omega_{\text{spp}}, \quad (1)$$

$$k_{\text{nl},\parallel} = 2k_{\text{pump},\parallel} - k_{\text{spp},\parallel}, \quad (2)$$

where ω_{nl} , ω_{pump} , and ω_{spp} refer to the frequencies of the nonlinear signal light, pump light, and SPP, respectively. $k_{\text{nl},\parallel}$, $k_{\text{pump},\parallel}$, and $k_{\text{spp},\parallel}$ refer to the components of the nonlinear signal light, pump light, and SPP vector in the direction of the wave vector parallel to the surface of the metal. For the pump light focused with a long-working-distance objective lens vertically incident onto the metal surface, the transverse wave vector $k_{\text{pump},\parallel} \approx 0$, thus $k_{\text{nl},\parallel} = -k_{\text{spp},\parallel}$. The in-plane information of the near field is encoded into the generated new frequency nonlinear signal field. In this process, the nonlinear photon is inside the light cone compared to the corresponding near-field photon, which is outside the light cone, as shown in Fig. 1(b). When the

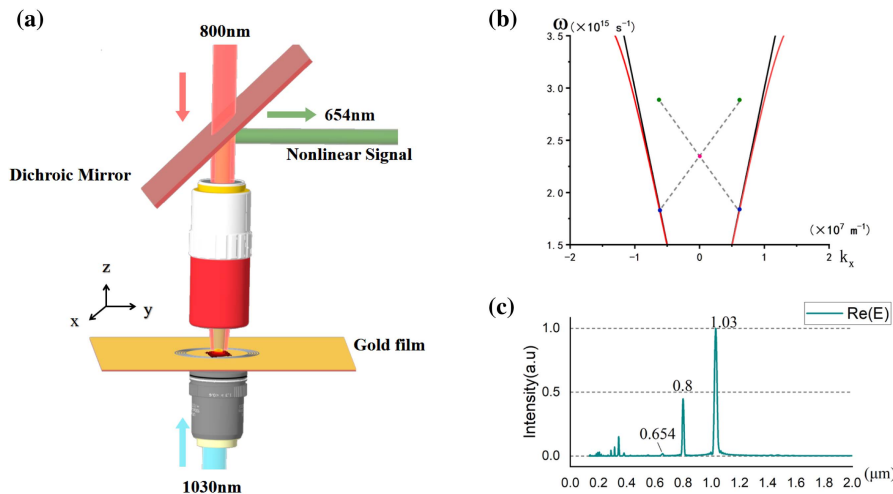


Fig. 1. (a) Schematic diagram of the nonlinear process. (b) Dispersion curve of the SPP on the gold film surface (red line) and the dispersion curve of the photons in vacuum (black line). The pump photon (pink), SPP (blue), and nonlinear photon (green) are connected by dashed lines, which represent the partially degenerate four-wave mixing interaction. (c) Spectrum detected at 5 nm above the gold film in simulation when the FWM happens.

in-plane wave vector component of the nonlinear polarized field is smaller than its wave vector component in free space at a certain frequency, it will radiate to the far field, which means the information of the SPP can be measured via the signal of the nonlinear light field, thereby enabling the retrieval of the near-field local information from the transformed light wavefield.

According to the symmetry-governed nonlinear selection rule, the third-order nonlinear effect exists in materials exhibiting C_2 symmetry, a characteristic that is frequently observed in various materials. Therefore, this near-field mapping process based on the third-order nonlinear effects can be applied to a wide range of materials. In this work, gold is utilized for excitation of nonlinear signals, characterized by a face-centered cubic crystal structure that conforms to C_2 symmetry.

Moreover, the polarization component of the near-field mapped by the nonlinear signal can be selected by the polarization of the pump beam, which satisfies the relation^[27]

$$E_{nl,x} \propto E_{pump,x} E_{spp,x}, \quad (3)$$

$$E_{nl,\sigma_{\pm}} \propto E_{pump,\sigma_{\pm}} E_{spp,\sigma_{\pm}}, \quad (4)$$

where $\sigma_{\pm} = \frac{\sqrt{2}(E_x \pm iE_y)}{2}$ refer to the left and right circular polarized lights. This indicates that when the linear or left/right circular polarized light is used as the pump beam, only the component of the excitation field that aligns with the polarization state of the pump light is effectively detected. The polarization-aligned nonlinear field component exhibits a much higher intensity than the orthogonal component, which can be regarded as background noise.

For $E_{spp,z}$, it can be expressed by $E_{spp,x}$ and $E_{spp,y}$ via the gradient of the components of the vector field derived from the

Maxwell equation^[28], which can be obtained from our method. Based on the obtained electric field distributions, the spin angular momentum S can be further calculated owing to the inherent relationship between E and H of the SPPs^[29].

In this work, the simulation is performed by the commercial software FDTD Solutions from Lumerical Solutions Inc. In the simulation, the source power needs to be large enough to generate the third-order nonlinear signal, and therefore, the electric field strength is set as 1000 V/m. In addition, to mitigate the impact of the spectral expansion of the light source on the relatively weak nonlinear signals, it is essential to consider the spectral domain of the excitation light during the configuration of the pulse, for which the pulse width is set as 120 fs.

As an example, we simulated a concentric ring structure fabricated on the gold film, as shown in Fig. 2(a). In the model, the wavelength of the pump light is set as 800 nm, and the excitation light is set as 1030 nm. The thickness of the gold film is set as 150 nm, the slot width of the groove is set as $w = 508.5$ nm ($\lambda_{spp}/2$), the grating period is $d = 1017$ nm (λ_{spp}), and the radius of the minimum ring is 8 μm . The depth of the grooves is 150 nm, which means the gold layer is entirely etched. In terms of material model setup, we chose to use the built-in Chi2/Chi3 model and Au(Johnson and Christy) as the base material.

The SPP field is excited by a radially polarized light with the third-order vortex ($l = 3$) and $\lambda = 1030$ nm. The x -polarized pump light with $\lambda = 800$ nm is incident vertically into the unstructured area in the center, and the nonlinear signal with $\lambda = 654$ nm is generated. The calculated results of the extracted nonlinear signal light field are influenced by the nonlinear coefficients. Improper setting of the nonlinear coefficients can lead to errors in the simulation process. When the coefficient is excessively high, the generated nonlinear field acts as a secondary source in the simulation, leading to simulation collapse,

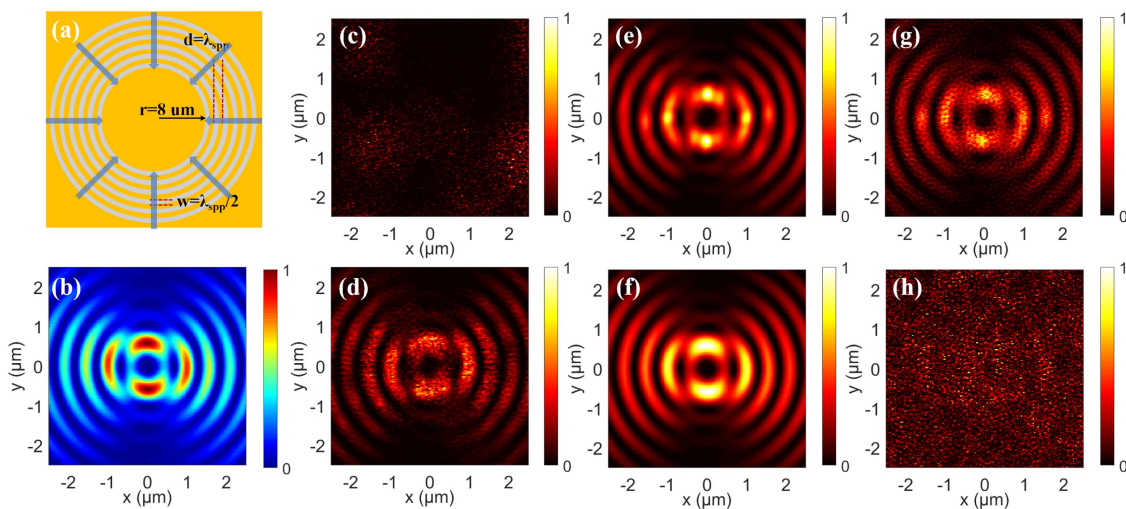


Fig. 2. (a) Schematic of the grating structure used to excite the SPP. The arrow (blue) represents the polarization direction of the excitation light, which is radially polarized. (b) The E_x component of the SPP field generated by the third-order vortex radially polarized light incident on the grating. (c)–(h) The nonlinear signal with the x -polarization pump light when the coefficient is set as (c) 1.2×10^{-4} (m/v)², (d) 10^{-4} (m/v)², (e) 10^{-6} (m/v)², (f) 10^{-7} (m/v)², (g) 10^{-8} (m/v)², and (h) 10^{-9} (m/v)², respectively.

as shown in Fig. 2(c). Conversely, if the nonlinear coefficient is too low, the nonlinear signal cannot be detected, as shown in Fig. 2(h). As shown in Figs. 2(d)–2(g), the simulation results of the nonlinear signals obtained from this procedure align well with the near-field distribution in Fig. 2(b) over a significant range of the nonlinear coefficients. The appropriate setting of the nonlinear coefficients depends on the system settings in different simulation environments. In the following parts of this work, the second-order nonlinear coefficients are set as 0, and the third-order nonlinear coefficients are set as $10^{-7}(\text{m/V})^2$. This allows the simulation process to accurately respond to the intensity mapping of the nonlinear field while ensuring the overall convergence and stability of the simulation.

3. Results

First, we utilize the constructed simulation model to calculate the nonlinear signals from the evanescent fields of the cylindrical vector vortex beams. The sample is a concentric ring structure fabricated on the gold film as shown in Fig. 2(a). The radially polarized light carrying optical vortex with different topological orders l is used to illuminate the structure, generating SPP vortex with various light field structures. The excitation light source is sufficiently large to adequately cover the grating structure, enhancing the intensity of the evanescent wave. Since the polarization state of the light varies angularly and a polarization singularity exists at the geometric center of the light source, it is necessary to align the geometric center of the light source with the center of the structure to create an ideal symmetrical vector light field. When the geometric positions of the light source and the grating structure are correctly set, the generated evanescent wave propagates in the direction of the concentric circle radius. The near-field right circular polarized components of the electric field of the SPP vortex with $l = 1, 2, 3, 4$ at the wavelength of $\lambda = 1017 \text{ nm}$ are shown in Figs. 3(a)–3(d), respectively. It is

found that the singularity in the central area increases in size as the topological order l increases. The presence of singularities indicates that the light field carries orbital angular momentum, which can exert rotational torque on particles^[30]. Furthermore, the intensity gradient formed in space generates light pressure, allowing tiny particles to be trapped and moved^[31]. The size of the pump light source is confined to the flat area of the structure within the concentric circles to prevent direct interaction with the excitation light passing through the groove. When the pump light is set as right circular polarized, the right circular polarized component of the near-field electric field is extracted. The corresponding nonlinear signals at the wavelength of $\lambda = 654 \text{ nm}$ are shown in Figs. 3(e)–3(h), which are consistent with the near-field results in Figs. 3(a)–3(d). In future experiments, the topological order can be controlled dynamically by the spatial light modulator, and the direct imaging of the near-field SPP vortex through NNOM can facilitate the real-time precise manipulation of nanoparticles in the light field gradient at the subwavelength scale.

Second, another type of SPP beam with self-focusing characteristics and particle manipulation capability has also been designed and generated. The PWB is a special type of diffraction-free beam that can maintain its shape, intensity distribution, and focusing effect during propagation. By altering the input conditions, characteristics, such as shape, direction, and degree of focusing, can be modulated and local field strength can be enhanced at a specific location, which is suitable for optical trapping and imaging. Here, we employ the angular spectrum synthesis method to generate the PWB, as depicted in Fig. 4(a). According to the approach described in Ref. [32], the groove structure is set using the angular spectrum of the PWB. The focusing position of the designed beam is at the coordinate origin, which is denoted as O in Fig. 4(a). The groove corresponding to the first-order component of the beam's angular spectrum (phase π) is located on the x -axis at a distance of $15 \mu\text{m}$ from the origin, and the distance from the origin varies with the

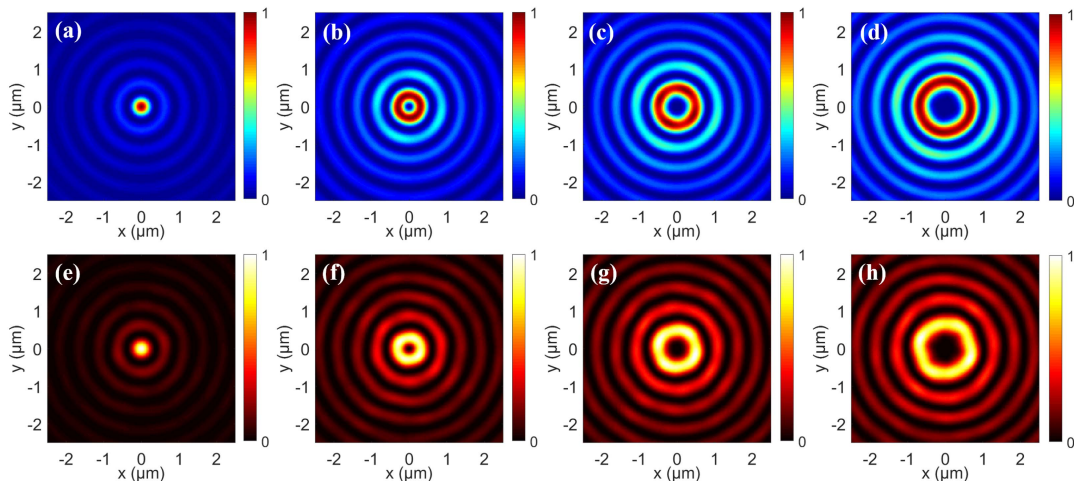


Fig. 3. (a)–(d) Right circular polarized components of the SPP field generated by the radially polarized light of l ($l = 1\text{--}4$) order vortex incident on the grating. (e)–(h) The nonlinear signals when the pump light is set to right circular polarized.

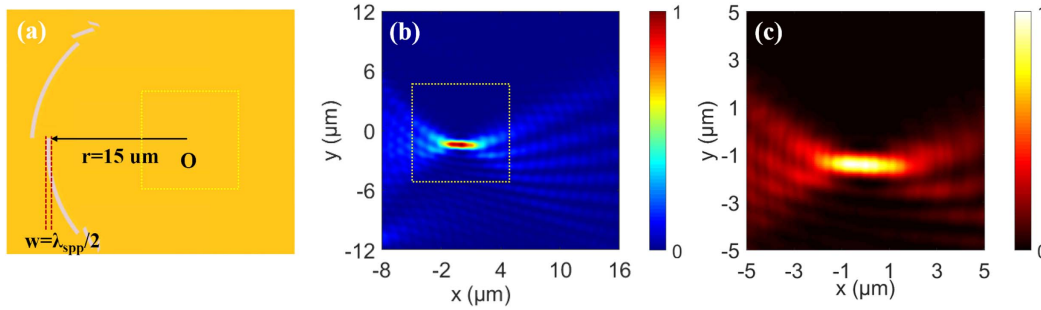


Fig. 4. (a) The structure to generate the PWB is designed based on the principle of the Fourier angle spectrum synthesis. O is the origin of the coordinates. (b) PWB excited under x linearly polarized light. (c) Nonlinear signal [yellow dashed areas in (a) and (b)] obtained under x linearly polarized pump light.

angular spectrum angle. The width of the engraved groove is selected to match the excitation wavelength as $w = 508.5 \text{ nm}$ ($\lambda_{\text{spp}}/2$). The excitation light source for generating the evanescent field is set as x -polarized light, covering all the grooves to generate the SPP waves. The generated SPP wave is focused inward within the arc, and the phase from each part of the arc is precisely controlled by the distance from O . As the phase of the evanescent wave at the focal point varies with the angle, it interferes with the designed angular spectrum component to produce the PWB shown in Fig. 4(b). The x -polarized pump beam is placed at the focusing position of the PWB and away from the grooves [yellow dashed area in Fig. 4(a)] to extract the x -polarized component in the PWB. The nonlinear signal obtained from the constructed simulation model is consistent with the marked area in Fig. 4(b), as shown in Fig. 4(c).

The generated PWB exhibits a marked local enhancement and non-diffractive characteristic, with the beam shape being maintained in the y direction. Similarly, the scanning-less detection of the near-field light field of the PWB provides a vital tool for potential applications of PWB, including dynamic manipulation of optical trapping at the subwavelength scale.

Moreover, we demonstrate the calculation of the spin textures of the photonic spin meron and skyrmion lattices. As depicted in Figs. 5(a) and 5(b), the quadrilateral and the hexagonal structures consisting of parallel gratings are constructed on the gold film. In each unit of parallel grating, the grating period is set as $d = 1.017 \text{ } \mu\text{m}$ (λ_{spp}), the lengths are 15 and 10 μm for the quadrilateral and hexagonal structures, respectively. The fill factor is 0.5, the number of gratings is 5, and the distance from the nearest grating to the center is 10 μm . The excitation light source is set to left circular polarized, and the SPP excited by the gratings propagates along the direction perpendicular to the gratings and interferes in the flat area at the center. The left and right circular polarized lights are utilized as the pump lights with the illuminated area of the light source confined within the central flat area, and the corresponding nonlinear signals are demonstrated in Figs. 5(e), 5(f) and 5(i), 5(j), respectively, which are consistent with the near-field left and right circular polarized components of the SPP fields, as shown in Figs. 5(c), 5(d) and 5(g), 5(h). The photonic spin topologies are determined by the field symmetries, and therefore through the quadrilateral and

hexagonal gratings, as shown in Figs. 5(a) and 5(b), the photonic spin meron lattice with four-fold symmetry and the photonic spin skyrmion lattice with six-fold symmetry can be generated, respectively^[33]. For the photonic spin textures, after the left and right circular polarized components of the SPP fields are extracted, the longitudinal component of the spin angular momentum can be calculated, and the 3D spin vector distribution can be reconstructed^[29]. Recently, the transformation between different topological states of the photonic skyrmion has been realized on the same sample^[34], but the experiment still relied on the NSOM system, and the field polarizations cannot be differentiated. In the future, based on the guidance provided by the simulation tool proposed in this work, the dynamic observation of the spin structures of different topological states on the same sample can be realized, which not only sheds light on the physical insight of the topological transformation but also contributes to the development of photonic-skyrmion-related applications.

4. Conclusion

In summary, the direct far-field imaging of the near-field distributions of the structured light fields through the nonlinear FWM effect has been investigated in this work. A FDTD simulation model has been constructed through the analysis of the setting of the incident source and the third-order nonlinear coefficients of the FWM process. By modulating the polarization state of the pump light, different near-field in-plane polarization components can be selectively retrieved for a wide range of structured light fields, including cylindrical vector vortex beams, PWBs, and topological spin textures. By constructing concentric gratings on the gold film, we employed the generated nonlinear signal to illustrate the intensity variations of the vortex light fields with different topological charges ($l = 1-4$), exhibiting the possibility of dynamic manipulation of nanoparticles in the light field gradient at the subwavelength scale. In addition, we detected PWBs characterized by non-diffractive and self-focusing properties, produced using the angle spectrum synthesis method, demonstrating the potential for real-time manipulation of the local enhancement point. Moreover, we

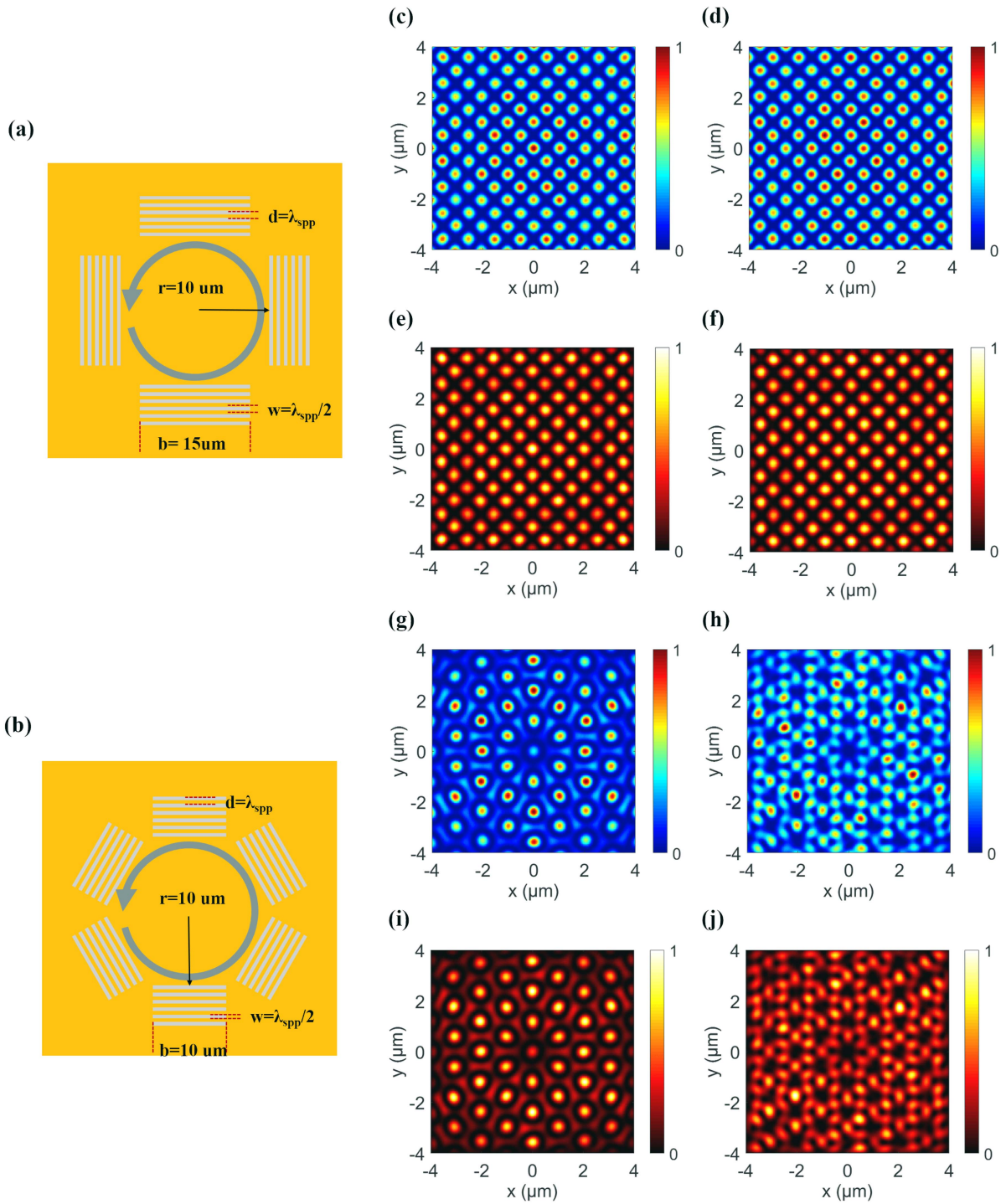


Fig. 5. (a),(b) Quadrilateral and hexagonal gratings, respectively. The blue circular arrow represents the polarization direction of the excitation light, which is left circular polarized. (c),(d) Left circular polarized and right circular polarized components of the SPP field generated by the quadrilateral grating under the left circular polarized excitation light. (e),(f) The corresponding nonlinear signals generated under the right and left circular polarized pump lights. (g)-(j) The corresponding results for the hexagonal grating.

generated photonic spin meron and skyrmion lattices using four-fold and six-fold symmetric grating structures, respectively. Two orthogonal circular polarization components were extracted from their evanescent fields, which are essential for the mapping of the 3D spin vector distribution and investigation

of the spin-orbit interaction phenomena in topological spin textures. For the structured light fields simulated above, the nonlinear signals are all consistent with the corresponding near-field polarization components. This work provides guidance for the scanning-less NNOM experiment of various kinds of the

latest emergent structured light fields, which opens new avenues for deeper insights into the light–matter interaction at the nano-scale, as well as advanced applications such as optical tweezers, non-diffractive beams, and optical information processing.

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