Nonlinear vectorial holography with quad-atom metasurfaces

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Vectorial optical holography represents a solution to control the polarization and amplitude distribution of light in the Fourier space. While vectorial optical holography has been experimentally demonstrated in the linear optical regime, its nonlinear counterpart, which can provide extra degrees of freedom of light-field manipulation through the frequency conversion processes, remains unexplored. Here, we experimentally demonstrate the nonlinear vectorial holography through the second harmonic generation process on a quad-atom plasmonic metasurface. The quad-atom metasurface consists of gold meta-atoms with threefold rotational symmetry. Based on the concept of nonlinear geometric phase, we can simultaneously manipulate the phase and amplitude of the left and right circularly polarized second harmonic waves generated from the quad-atom metasurface. By superposing the two orthogonal polarization components, the quad-atom metasurface can produce nonlinear holographic images with vectorial polarization distributions. The proposed metasurface platform may have important applications in vectorial polarization nonlinear optical source, high-capacity optical information storage, and optical encryption.

Significance

Vectorial optical holography, as a scheme to control the polarization distributions of light in the Fourier space, has been experimentally demonstrated in the linear optical regime. However, its nonlinear optical counterpart, which can generate the holographic images at different frequencies to that of the incident light, remains unexplored. Here, we demonstrate nonlinear vectorial holography through the second harmonic generation process on a quad-atom plasmonic metasurface, which consists of gold meta-atoms with threefold rotational symmetry. The proposed metasurface platform may have important applications in vectorial polarization nonlinear optical source, high-capacity optical information storage, and optical encryption.

Optical holography, which can be used to reconstruct the wave front of light, has been successfully applied to many areas, such as advanced microscopy (1), data storage (2), display (3), optical tweezer (4), optical communications (5), and quantum science (6, 7). Since the advent of optical holography, one of light’s degrees of freedom (i.e., polarization) has been attracting scientists’ attentions (8, 9). In the past decades, vectorial polarization holography was developed by using photoanisotropic materials, such as Azobenzene, silver-halide materials, liquid crystal polymers, and so on (10). However, this kind of polarization holography is usually limited by the working wavelength of light and is only able to reconstruct two wave fronts with orthogonal polarizations (9). Recently, three-dimensional vectorial holography was also developed by using spatial light modulators (SLMs) (11). However, the SLM-based systems are very bulky and limit the applications in integrated optics. This constraint could be partially circumvented by using optical metasurfaces.

The optical metasurfaces, which consist of spatially variant plasmonic or dielectric meta-atoms, have been extensively used to manipulate the phase, polarization, and amplitude of light (12). Compared with the photoanisotropic materials, which are sensitive to a specific wavelength range of light, the metasurface as an artificial structure can be easily designed at ultraviolet to infrared, terahertz and even longer wavelengths. The optical efficiency of the metasurface device could be up 80% or even higher (13–15). All these properties make optical metasurface more practical for vectorial holography applications (16, 17). By controlling the phase response of meta-atoms, various vectorial holography schemes have been experimentally demonstrated. For example, the high-efficiency polarization-sensitive holography was achieved through the geometric phase-controlled meta-atoms (13, 18). By combining geometric phase and resonant phase (19) or topological phase (20), the multiplexed holography with orthogonal circular polarization states can be achieved. Later, the multiple-polarization input and multiple-polarization output were also utilized to design the metasurface vectorial holograms (21–23). Very recently, the vectorial holography based on one polarization input was also demonstrated by engineering the meta-molecules of the metasurfaces (24–30) or by using the protocol of Jones matrix holography (31).

In the nonlinear optical regime, polarization-controlled holography through second harmonic generation (SHG) (32, 33) has been demonstrated by using the nonlinear geometric phase-controlled metasurfaces. In these kinds of devices, by switching the polarization of the fundamental wave (FW), one can easily change the polarization of the nonlinear holographic images. However, the more generalized nonlinear vectorial holography, such as the one–polarization input and multiple-polarization output scheme, has not been studied. This is because the simultaneous manipulation of...
polarization, phase, and amplitude of nonlinear optical waves from the meta-atoms is challenging. In this work, we demonstrate the nonlinear vectorial holography through the SHG process on a quad-atom metasurface. The quad-atom metasurface consists of four gold meta-atoms with threefold (C3) rotational symmetry and spatially variant orientation directions. The four meta-atoms in a meta-molecule can be used to manipulate the phase and amplitude of both left and right circularly polarized second harmonic (SH) waves. As shown in Fig. 1, under the pumping of a linearly polarized FW, the SH waves generated from the quad-atom metasurface form a holographic image with vectorial polarizations. In principle, there is no limit to the types and distribution of the polarization states in the holographic image.

The proposed nonlinear vectorial holography may have important applications in ultracompact and multifunctional nonlinear optical sources, high-capacity optical information storage, and so on.

Results

Design of the Nonlinear Quad-Atom Metasurface. Fig. 2 shows the design principle of the nonlinear quad-atom metasurface. In order to generate an arbitrary state of polarization (SoP), it is necessary to simultaneously manipulate the phase, amplitude, and polarization of the SH waves. From the previous work on nonlinear diatomic metasurface, it is shown that the phase and amplitude of the left circularly polarized (LCP) and right circularly polarized (RCP) SH waves can be well controlled by varying the relative orientation angle of the two meta-atoms in a diatomic unit cell (33). However, one disadvantage of the diatomic design is that the intensity of the SH waves with LCP and RCP states cannot be independently controlled. To solve this problem, we design a nonlinear quad-atom metasurface, which consists of two groups of diatomic meta-molecules. As shown in Fig. 2A, two gold plasmonic meta-atoms with C3 rotational symmetry are arranged in the top (Fig. 2A, Upper) and bottom groups (Fig. 2A, Lower). Based on the concept of symmetry selection rules (34, 35) and nonlinear geometric phase (36, 37), we know that under the pumping of FW with the LCP or RCP state, the SH wave generated from the gold meta-atom has opposite circular polarization to that of the FW. The SH wave also experiences a nonlinear geometric phase factor of \( \exp(3i\sigma\theta) \), where \( \sigma = \pm 1 \) corresponds to the circular polarization state of the FW and \( \theta \) is the orientation angle of the C3 meta-atom. The top group (purple color) (Fig. 2A, Upper) in the quad-atom meta-molecule is used to control the amplitude \( A_R \) and phase \( \phi_R \) of SH waves with the RCP state. Similarly, the bottom group (blue color) (Fig. 2A, Lower) is used to control the degrees of freedom (amplitude \( A_L \) and phase \( \phi_L \)) of the SH wave with the LCP state. By independently controlling the intensities of the SH waves with LCP and RCP states, we can realize arbitrary SoP, \( P(\psi, \chi) \), where the polarization parameters are described by the main axis angle \( \psi \) and the ellipticity angle \( \chi \). As shown in Fig. 2B, the relationship between the polarization parameters and the amplitude and phase components of the SH waves can be described as \( \psi = (\phi_R - \phi_L)/2 \) and \( \chi = \sin^{-1}[(A_R^2 - A_L^2)/(A_R^2 + A_L^2)]/2 \). As the two polarization parameters \( \psi \) and \( \chi \) can be exactly mapped to the Poincaré sphere (Fig. 2C) (38), we can easily identify the design of the nonlinear quad-atom meta-molecule to obtain the arbitrary SoP of the SH waves.

Nonlinear Holography with a Single-Polarization State. To verify the ability to realize nonlinear vectorial holography, we designed and fabricated four quad-atom metasurface devices named Meta-LCP, Meta-RCP, Meta-LP (H) (linear polarization along horizontal direction), and Meta-LP (V) (linear polarization along vertical direction). Using nonlinear metasurfaces, we can reconstruct vectorial holographic images with LCP, RCP, H, and V polarization states, respectively. Here, we choose four Greek letters as the target images for demonstration (Fig. 3 A–D). From our previous experience, the gold/ITO (indium tin oxide) hybrid metasurface, which consists of C3 meta-atoms, can radiate strong SHG signals at the FW length of \( \sim 1,210 \) nm (33, 39). In this work, the Gerchberg-Saxton (GS) algorithm was used to calculate the corresponding phase distributions (Fig. 3 E–H) of the SH waves.
wave directly generated from the metasurface hologram (40). The polarization states of the holographic images are determined by both the phase and the amplitude of the SH waves with RCP and LCP states. Based on the principle of nonlinear geometric phase, only one C3 meta-atom in a unit cell is needed to realize the nonlinear holographic image with left or right circular polarization. For linear and elliptical polarization states of the holographic images, the spatially variant orientation angles of the C3 gold meta-atoms in the quad-atom meta-molecule will be utilized. A 30-nm-thick gold metasurface was then fabricated on the 15-nm-thick ITO-coated glass substrate by using the electron beam lithography (EBL) process. The size of the gold meta-atom has been optimized to obtain efficient SHG. Fig. 3 I–L shows the scanning electron microscope images of the metasurface holograms. The periods of the meta-atoms are 500 nm in both x and y directions. The arm width and arm length of the gold meta-atoms are ~80 and 185 nm, respectively. To characterize the nonlinear optical property of the C3 meta-atoms, we also fabricated a control metasurface in which the C3 gold meta-atoms are uniformly distributed in a square lattice with a period of 500 nm along both x and y directions (SI Appendix, section 3). It can be found that the SHG from the control metasurface has the peak efficiency at the FW length of 1,210 nm (SI Appendix, Fig. S1). To demonstrate the nonlinear holographic images with a single-polarization state, a horizontally polarized (SI Appendix, Fig. S2) femtosecond laser at the wavelength of 1,210 nm (Materials and Methods), which consists of both LCP and RCP components, was used to pump the four holograms from substrate to metasurface direction. Then, the holographic images at SH frequency were collected by a 10× objective lens and recorded with an sCMOS (scientific complementary metal-oxide-semiconductor) camera. The polarization states of the nonlinear holographic image can be analyzed by using a linear polarizer and a quarter-wave plate. As shown in Fig. 3 M–T, the Greek letters with designed polarization states were successfully reconstructed at the top right of the recording area. The twin images at the bottom left come from the contribution of the opposite geometric phases of the SHG waves with RCP and LCP states, respectively. This phenomenon was also observed in most of the geometric phase–based metasurface holography (17). It can be found that the qualities of Fig. 3 M and P are better than those of Fig. 3 Q and T. This is mainly because one C3 meta-atom in a unit cell is enough to realize the circularly polarized nonlinear holographic image. Thus, the effective pixel numbers of metasurfaces in Fig. 3 I and J are four times those of the devices in Fig. 3 K and L.
Nonlinear Holography with Multiple-Polarization States. In the previous section, we have verified our ability to realize the nonlinear holographic images with a single-polarization state. Now, we aim to demonstrate the multiple-polarization state nonlinear holography with the quad-atom metasurface. To do this, we adopt the modified GS algorithm, which was proposed in ref. 26. First, we designed a vectorial hologram device Vector-A, which can be used to generate an image with spatially variant linear polarizations. The target intensity and polarization distributions are shown in Fig. 4A. In the “cross” image, the linear polarization distributions in the quadrants I to IV are along 0°, 45°, 90°, and 135° directions, respectively. For the linearly polarized holographic images, the required amplitude ratio of SH waves from the top and bottom groups of a quad-atom meta-molecule is \( A_T/A_L = 1 \). Then, the phase distributions (Fig. 4B and C) of SH waves with RCP and LCP states can be calculated by using the modified GS algorithm (26) and a self-developed MATLAB code. According to the working principle of the nonlinear quad-atom metasurface, the designed amplitude ratio and the phase distributions of the generated SH waves are directly related to the orientation angles of the four meta-atoms in a unit cell (SI Appendix, sections 1 and 2). Then, the corresponding metasurfaces, which consist of gold meta-atoms, were fabricated on the ITO-coated glass by using the EBL process. The scanning electron microscope image of the gold metasurface is shown in Fig. 5D. The dashed lines in the scanning electron microscope image show the quad-atom meta-molecule. As expected from our design, under the pumping of a horizontally polarized FW at the wavelength of 1,210 nm (SI Appendix, Fig. S2), the Vector-A hologram can be used to reconstruct the intensity profile of the cross image at the wavelength of 605 nm (SI Appendix, Fig. S3). In addition, we characterized the polarization parameters (\( \psi, \chi \)) to verify if the nonlinear vectorial holography has been realized. It can be found that the calculated distribution of the main axis angle \( \psi \) in Fig. 4E agrees well with the measured values shown in Fig. 4F. This means that the SH images are linearly polarized along 0°, 45°, 90°, and 135° directions in the quadrants I to IV, respectively. Fig. 4E, Lower and F, Lower correspond to the twin images due to the opposite geometric phase (17). As we know, for the SH waves with linear polarizations, the calculated values of \( \psi \) are equal to zero (Fig. 4G). To experimentally verify this, we also measured the spatial distribution of \( \chi \) (Fig. 4H). It is found that although the measured values are very random, the averaged values of \( \chi \) are close to the theoretical ones. In a more generalized case, we designed another nonlinear vectorial hologram Vector-B, with which we can reconstruct the SH image with circular and elliptical polarizations. In the target image of a “flag” (Fig. 5A), quadrants I and III have LCP and RCP states, respectively, while quadrants II and IV correspond to the two elliptical polarization states with \( (\psi, \chi) = (45°, 18.43°) \) and \( (135°, -18.43°) \), respectively. Fig. 5B and C represents the phase experienced by the SH waves with RCP and LCP states. To generate the elliptically polarized states, the amplitude ratio of SH waves from the top and bottom groups of the meta-molecule is \( A_T/A_L = 1.0463 \). From the phase and amplitude distributions of the SH waves with RCP and LCP states, we can calculate the in-plane rotation angles of the meta-atoms. The scanning electron microscope image of the gold metasurface is shown in Fig. 5D. Then, under the pumping of an H-polarized FW, the polarization parameters of the holographic image were also analyzed (SI Appendix, Fig. S3). Fig. 5E and G shows the calculated main axis and ellipticity angles of the flag image and its twin counterpart. The value of \( \psi = 0 \) is used to represent circular polarization states in quadrants I and III. In quadrants II and IV, the main axis angles of the two elliptically polarized regions are 45° and 135°, respectively. From Fig. 5F and H, we can find that the measured results of \( \psi \) and \( \chi \) agree with the calculated ones.

Discussion

In summary, we proposed a quad-atom metasurface for nonlinear vectorial holography. By varying the orientation angles of the four plasmonic meta-atoms with C3 rotational symmetry, we are able to independently control the phase, amplitude, and polarization of the generated SH waves. By adopting a modified GS algorithm, we designed and fabricated the quad-atom metasurfaces for producing nonlinear holographic images with single- and multiple-polarization states. Based on the theory of nonlinear geometric phase, we know that the proposed concept in this work can be applied to other nonlinear optical processes, such as third harmonic generation, four-wave mixing, and terahertz wave generation. It should be noted that the nonlinear optical efficiency on the plasmonic metasurface is usually low, which may limit the practical applications of nonlinear vectorial holography. To circumvent this constraint, the dielectric materials with high

Fig. 4. The nonlinear vectorial holography with linear polarization states. (A) The target image cross and the distribution of linear polarization states. (B and C) The calculated phase distributions experienced by the SH waves with RCP and LCP states. (D) The scanning electron microscope image of the Vector-A metasurface. (Inset) The dashed box indicates a unit cell of the quad-atom metasurface. (Scale bar, 500 nm.) (E-H) The calculated and measured main axis angle \( \psi \) and ellipticity angle \( \chi \) of the nonlinear holographic image cross and its twin counterpart. The FW at the wavelength of 1,210 nm is horizontally polarized. SEM, scanning electron microscope. Cal, calculated results. Exp, experimental results.
nonlinear susceptibilities could be introduced into the design of the nonlinear optical metasurfaces (41). Considering the growing demands of multiple functionalities in nanophotonic devices, the proposed nonlinear quad-atom metasurface platform may have important applications in high-capacity optical information storage, optical encryption, bio-imaging, and so on.

Materials and Methods

Measurement of the Nonlinear Holographic Images. The FW at the wavelength of 1,210 nm, which is generated from an optical parametric oscillator (Coherent, Inc), has a repetition frequency of 80 MHz and a pulse duration of ~250 fs. The power of the FW, which can be controlled by the fast axis angle of a half-wave plate, was set to be 45 mW. After being polarized by a linear polarizer with the transmission axis along horizontal direction (SI Appendix, Fig. S2), the incident FW is focused onto the holograms from the substrate to metasurface direction by a lens with focal length of 250 mm. The generated SH waves are collected by a 10x objective lens. Then, a quarter-wave plate and the second linear polarizer (LP2) were used to analyze the polarization states of the SH waves. A color filter was placed behind LP2 to filter out the residue of the FW. Finally, the nonlinear holographic images were projected onto an sCMOS camera through a tube lens with the focal length of 100 mm.

Data Availability. All study data are included in the article and/or SI Appendix.

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