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Conversion of the optical orbital angular momentum in a plasmon-assisted second-harmonic generation

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We experimentally demonstrate the plasmon-assisted second-harmonic generation of an optical orbital angular momentum (OAM) beam. Because of the shape resonance, the plasmons in a periodic array of rectangular metal holes greatly enhance the nonlinear optical conversion of an OAM state. The OAM conservation (i.e., 2l₁ = l₂ with l₁ and l₂ being the OAM numbers of the fundamental and second-harmonic waves, respectively) holds well under our experimental configuration. Our results provide a potential way to realize nonlinear optical manipulation of an OAM mode in a nano-photonic device.

In the past decades, plasmons have attracted considerable attentions due to their potential applications in miniature photonic devices. The classical experiment is the extraordinary optical transmission (EOT) phenomenon through a periodic metal-hole array, which is first reported by Ebbesen et al.¹ The EOT spectrum can be modulated by adjusting the parameters of samples, such as the shape, the period, and the size of metal hole.²–⁴ Now, the plasmonic devices have been widely applied in numerous fields including wavefront manipulation,⁵,⁶ polarization control,⁷ and phase modulation.⁸ Interestingly, because the local field is greatly enhanced by plasmons, metal nano-structures have been utilized to realize nonlinear optical conversion in an integrated chip.¹¹–¹³ Usually, second-harmonic generation (SHG) in a metal film is very weak because metal is centrosymmetric. However, SHG can be significantly improved by fabricating bowtie nano-apertures, G-shaped arrays, and double-hole structures in the metal film.¹³–¹⁷

Recently, the light carrying an orbital angular momentum (OAM), which has a helical wavefront perpendicular to its propagation direction, attracts increasing interests.¹⁸ The related studies have been extended from the fundamental understanding of photons to practical applications in optical manipulation,¹⁹,²⁰ optical trapping,²¹,²² and quantum information processing.²³,²⁴ One important development is to manipulate the optical OAM beam in an integrated plasmonic device.²⁵,²⁶ For example, in a previous work,²⁷ it has been demonstrated that the OAM information survives during an EOT process in a metal-hole array. However, nonlinear optical conversion of an OAM state in a plasmonic structure has not been clearly studied. In a plasmon-free SHG process, the OAM conservation (i.e., 2l₁ = l₂ with l₁ and l₂ being the OAM numbers of the fundamental and second-harmonic waves, respectively) has been proven in most of the experiments.²⁸–³¹ It is interesting to re-examine this process with involving a photon-plasmon-photon process. In this letter, we experimentally investigate the OAM conversion through a plasmon-assisted SHG process in a periodic array of rectangular metal holes. Such periodic array does not introduce an extra spatial phase into the optical beam,²⁷ which makes it a good system to study the role of the plasmon in such nonlinear OAM conversion process.

We first use the finite-difference-time-domain (FDTD) method to simulate the metallic nanohole array. Because of the shape resonance effect, the aspect ratio of the rectangular hole (i.e., a/b in Fig. 1(a)) in a metal film critically determines the resonant enhancement of the SHG. The optimized value of the aspect ratio for SHG is ~2 as reported by Wang et al.³² Because of the rectangular hole in the metal film, the polarization of the fundamental wave (FW) has a strong influence on the excitation of the plasmon, and therefore, the SHG.⁵,³³ In our simulation, the sample is optimized to utilize the FW with its polarization along the y-axis. To maximally excite the plasmonic resonance, the input fundamental wavelength is set at the EOT peak of the sample. To search the optimized parameter for the SHG experiment, the aspect ratio of the rectangular nano-hole is scanned near a value of 2. After numerical calculations, the period of the array is selected to be 410 nm and the size of the rectangular hole is optimized to be 240 nm × 120 nm (Fig. 1(a)). The nanostructure is fabricated in a 150 nm thick gold film on a glass substrate by using a focused-ion beam (Strata FIB 201, FEI

FIG. 1. (a) The unit cell of the nanohole array with a = 240 nm, b = 120 nm, and p = 410 nm. (b) The SEM image of the sample from the top view.

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Company). The scanning electron microscope (SEM) image of the nanohole array is shown in Fig. 1(b). The whole array has an area of about $45 \text{ \mu m}^2$.

The schematic of experimental setup to perform the SHG measurement is shown in Fig. 2. The light source is a tunable Ti:Sapphire femtosecond laser with a repetition rate of 80 MHz and a pulse width of 140 fs. It has a wavelength ranging from 690 nm to 1050 nm. In the experiment, after being reshaped by a 4f system, the first quarter-wave plate (QWP) is used to change the linear polarization of the input laser to a circular polarization. Then, a Q-plate is used to imprint different OAM values of $l_1 = 1, 2,$ and $3$ into the FW. In order to guarantee the $y$-polarization of the FW, another QWP transforms the polarization of the generated vortex beam back to a linear polarization. The FW with OAM is focused on the sample by a $50 \times$ objective lens. The transmitted signal is collected by another objective lens ($10 \times$). By using different filters, the FW and SH waves are analyzed by a CCD camera. The spectrometer is used to detect the wavelength of the transmitted signal. The OAM mode is investigated by using a cylindrical lens. The topological charge can be measured by counting the dark stripes of the pattern converted by the cylindrical lens.

To efficiently realize the plasmon-assisted SHG of an OAM mode, the fundamental wavelength (i.e., the EOT wavelength) is obtained by measuring the transmission spectrum of the sample. In the previous work, we have demonstrated that the OAM mode does not change the transmission spectrum of a metal-hole array. In the experiment, we use a Gaussian beam to characterize the performance of the sample for simplification. The polarization of the input beam is perpendicular to the long edge of the rectangular hole. The transmission of the sample is measured by sweeping the input wavelength from 750 nm to 1000 nm with a step of 5 nm. As shown in Fig. 3, the experimental transmission peak at 886 nm is well consistent with the simulation. Because of the non-perfect fabrication of the sample, the transmitted intensity is lower than its theoretical value. The transmission efficiency (normalized to the area of the holes) at 886 nm is measured to be 1.84, which indicates the excitation of the plasmons at this wavelength.

As shown in Fig. 2, the FW at 886 nm first passes through the Q-plate to produce different OAM modes with topological charges of $l_1 = 1, 2,$ and $3$. The intensity of the input FW is kept to be 80 mW. In the experiment, the recorded images after the transmission are shown in Fig. 4. As shown in Figs. 4(a)–4(c), the intensity distributions of the transmitted FWs present donut-shaped patterns. After being

![Diagram](image-url)

**FIG. 2.** The schematic diagram of the experimental setup. L1: lens with a focus length of 100 mm; L2: lens with a focus length of 50 mm; $\lambda/4$: quarter-wave plate; SF: short-pass filter; CL: cylindrical lens; CCD: CCD camera.

![Graph](image-url)

**FIG. 3.** The measured (black) and calculated (red) transmission spectra of the sample. The polarization of the input FW is perpendicular to the long axis of the rectangular hole (y-polarization).

![Images](image-url)

**FIG. 4.** (a)–(c) The transmitted fundamental beams carrying different OAM modes. By using a cylindrical lens, the corresponding topological charges can be counted from the converted patterns ((d)–(f)) to be $l_1 = 1, 2,$ and $3$, respectively. The generated SH OAM modes are shown in (g)–(i). The corresponding topological charges of the SH mode are $l_2 = 2, 4,$ and $6$ as shown in (j)–(l), respectively.
converted by a cylindrical lens, the OAM numbers of the FWs are 1, 2, and 3, respectively, by counting the dark strips (Figs. 4(d)–4(f)). As expected, the OAM information of the FW survives after the EOT process in the periodic array of rectangular holes in the metal film, which is consistent with our previous experimental observations.\textsuperscript{22} After filtering out the FWs by a short-pass filter, the SH signals are selected. The plasmon-assisted SH patterns with different input OAM modes are clearly recorded by the CCD camera as shown in Figs. 4(g)–4(i). The OAM numbers of the generated SH waves are $l_1 = 2$, 4, and 6 (Figs. 4(j)–4(l)), which correspond to the input OAM modes of $l_1 = 1, 2$, and 3, respectively. Our experimental results show that the OAM conservation law (i.e., $2l_1 = l_2$) holds well during the plasmon-assisted SHG process in the periodic metal-hole array. It should be mentioned that the periodic structure in our experiment does not introduce an extra phase. If using a non-uniform structure (for example, spiral structure), the spatial phase information carried in the nanostructure should be considered in the OAM conservation. Figure 5 shows the measured intensities of different SH OAM modes. The input fundamental power is kept to be 80 mW. Obviously, the SH intensity decreases as increasing the input OAM number. This can be well explained by the fact that a higher-order OAM mode has a bigger radius (Figs. 4(a)–4(c)), and therefore, a lower power density.

In conclusion, we have experimentally demonstrated the OAM conversion during the SHG process in a periodic metal-hole array. The metal hole is designed to be rectangular with an optimized size of 240 nm $\times$ 120 nm. Because of the plasmonic resonance at the fundamental wavelength, the SHG of an optical OAM beam is greatly enhanced. The OAM is conserved during such plasmon-assisted SHG process in a periodic metal-hole structure. Our results open a door to utilize the metallic nanostructure to realize nonlinear optical conversion of an OAM state, which has potential applications in integrated optical devices.

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