

# Nonlinear Raman-Nath second harmonic generation of hybrid structured fundamental wave

HUAQING ZHOU,<sup>1</sup> HAIGANG LIU,<sup>2,3</sup> MINGHUANG SANG,<sup>1</sup> JUN LI,<sup>4</sup> AND XIANFENG CHEN<sup>2,3,\*</sup>

<sup>1</sup>Jiangxi Key Laboratory of Photoelectronics & Telecommunication, Department of Physics, Jiangxi Normal University, Nanchang 330022, China

<sup>2</sup>State Key Laboratory of Advanced Optical Communication Systems and Networks, Department of Physics and Astronomy, Shanghai Jiao Tong University, Shanghai 200240, China

<sup>3</sup>Key Laboratory for Laser plasma (Ministry of Education), Collaborative Innovation Center of IFSA (CICIFSA), Shanghai Jiao Tong University, Shanghai 200240, China

<sup>4</sup>Science and Technology College, Jiangxi Normal University, Jiangxi 330027, China

\*xfchen@sjtu.edu.cn

**Abstract:** We numerically and experimentally investigated the nonlinear Raman-Nath second harmonic generation of hybrid structured fundamental wave whose phase modulation combined periodic and random structure. The second harmonic generation of both one- and two-dimensional hybrid structured fundamental wave were investigated in this paper. The results show that more diffraction spots can be obtained in these hybrid structures than the pure periodic modulation cases. Besides, the total intensity of the second harmonic not only can be dramatically enhanced without altering the diffraction angles, but also is increasing with the degree of randomness of the structure. This study enriches the family of second harmonic generation of structured fundamental wave and has potential application in dynamically controlling second harmonic wave in arbitrary directions.

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## 1. Introduction

The structural property of the matter always play an important role in many physics processes, in other words, many physics phenomena can be realized and controlled by this kind of structure. Hence the structural property of different matter had been intensively investigated in the past, such as the diffraction phenomenon in grating structure [1], plasmons [2], photonic band gap in different structured photonic crystals [3], and so on. In the field of nonlinear optics, lots of studies have introduced these structures into nonlinear photonic crystals (NPCs) [4,5], in which the second-order susceptibility  $\chi^{(2)}$  is spatially modulated while the linear susceptibility remains constant for the second harmonic generation (SHG) processes. For example, efficient SHG could be achieved in a periodically modulated NPC under quasi-phase-matching (QPM) manner [6,7] and the broadband frequency conversion was also realized in a NPC, in which the structure are in combination of periodic and disordered or chirped modulation [8–10]. When the fundamental wave (FW) propagates along perpendicular direction of the QPM, nonlinear Bragg diffraction [11], nonlinear Čerenkov radiation [12,13] and nonlinear Raman-Nath diffraction [14,15] can be observed in different conditions. In addition, different structured NPCs can also be used for shaping the second harmonic (SH). For example, the SH vortex beams could be generated in a fork-shaped NPC [16]. Arbitrary two-dimensional nonlinear beam shaping could also be realized in special designed NPCs [17].

However, in the processes of shaping the SH, most attention paid on the structures of nonlinear medium. The disadvantages of this method are complex fabrication and unchangeable SH pattern. Hence, to solve these problems, in our previous research, the concept of structured FW, in which the wavefront phase of the FW is periodically modulated by a spatial light modulator (SLM) in real time, was introduced to the process of SHG. We have proposed and experimentally demonstrated that nonlinear Raman-Nath SH [18] and scattering-assisted SH [19] can be achieved when a structured FW incident in a homogeneous nonlinear medium. But these papers only discussed the SHG of periodically modulated FW.

In this paper, we numerically and experimentally investigated the nonlinear Raman-Nath SHG of hybrid structured FW whose phase modulation combined periodic and random structure. As shown in Fig. 1, each period was divided into  $n$  random bars. The SHG of both one- (1D) and two-dimensional (2D) hybrid structured FW have been investigated. The results show that the introduction of the random modulation to the FW can provide more reciprocal vectors than the pure periodic structure. Consequently, more diffraction spots and

higher conversion efficiency can be obtained. Besides, we also characterize the intensity dependence of nonlinear Raman-Nath diffraction on the degree of randomness of the structure, which shows that the randomness only affects the intensity of the diffraction signal, but have no effect on the emission angles.

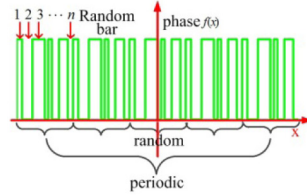


Fig. 1. Schematic of the hybrid structured FW whose phase modulation combine periodic and random structure.

## 2. The theoretical analysis

Suppose that the FW propagates along the  $y$ -axis of the crystal and the phase of the fundamental Gaussian wave modulates along  $x$ -direction. Under the assumption of non-diffraction, the incident FW can be generally expressed as the following Fourier series

$$E_1 = A_1(x) \exp[i(k_1 y - \omega t)] \sum_{m=-\infty}^{+\infty} C_m \exp(imG_0 x), \quad (1)$$

with  $A_1(x) = A_1 \exp(-x^2/a^2)$ , in which  $A_1$  and  $a$  represent the amplitude and beam width of the fundamental Gaussian wave, respectively. Where  $\omega$ ,  $C_m$ ,  $k_1$  and  $G_0 = 2\pi/\lambda$  ( $\lambda$  is the modulated period of the FW) are the frequency, Fourier coefficients, wavevector and basic reciprocal vectors of the FW, respectively. Under the assumption of paraxial and undepleted pump approximation, the spatial evolution of the amplitude of the SH field  $A_2(x, y)$  in the nonlinear medium can be obtained [20,21]:

$$\left( \frac{\partial}{\partial y} + \frac{i}{2k_2} \cdot \frac{\partial^2}{\partial x^2} \right) A_2(x, y) = i\beta_2 A_1^2(x) \exp(i\Delta k y) \left[ \sum_{m=-\infty}^{+\infty} C_m \exp(imG_0 x) \right]^2, \quad (2)$$

where  $\Delta k = k_2 - 2k_1$  ( $k_2$  is the wavevector of SH wave),  $\beta_2 = k_2 \chi^{(2)} / (2n_2^2)$  is the nonlinear coupling coefficient with  $n_2$  being the refractive index of the SH. Then the amplitude of the SH field can be expressed as

$$A_2(k_x, y) = a \sqrt{\frac{\pi}{2}} A_1^2 \beta_2 y \exp\left(\frac{ik_x^2}{2k_2} y\right) \text{sinc}\left[y\left(\Delta k - \frac{k_x^2}{2k_2}\right)/2\right] \sum_{q=-\infty}^{+\infty} b_q \exp[-a^2(qG_0 + k_x)^2/8], \quad (3)$$

with  $\text{sinc}(x) = \sin(x)/x$ ,  $\text{sinc}(0) = 1$ ,  $q = m + n$  and  $b_q = \sum_{m,n}^{m+n=q} |C_m C_n|$ ,  $k_x$  is the reciprocal vectors of the FW along the  $x$  direction. Considering the spectral density of the SH field  $S_2(k_x, y) = |A_2(k_x, y)|^2$ , we obtain

$$S_2(k_x, y) = \frac{1}{2} \pi a^2 \beta_2^2 y^2 A_1^4 \left\{ \text{sinc}\left[y\left(\Delta k - \frac{k_x^2}{2k_2}\right)/2\right] \right\}^2 \left\{ \sum_{q=-\infty}^{+\infty} b_q \exp[-a^2(qG_0 + k_x)^2/8] \right\}^2. \quad (4)$$

Nonlinear Raman-Nath SH can be achieved when the condition  $qG_0 + k_x = 0$  is satisfied. The condition can also be transformed into the form  $\kappa_x = -qG_0 = k_2 \sin\alpha_m$  ( $q$  refer to the diffraction order). If the FW is hybrid structure whose phase is sharply modulated from 0 to  $\phi$ , in one period  $\Lambda$ , the phase distribution can be expressed as  $\exp[i(1+(-1)^j)\phi]$  and the

length of the corresponding structure of the FW is  $\Lambda B(j)/\sum B(j)$ , where  $B(j) = rand(n)$  is a series of random number with the length  $n$ ,  $j$  is the element index of  $B$ . In this case, as the existing of the random structure, there is no analytical solution of the Fourier coefficients. After the generation of the FW structure, the Fourier coefficients can be calculated according to  $C_m = \frac{1}{\Lambda} \int_0^\Lambda f(x) \exp(-imG_0x) dx$ ,  $m = 0, \pm 1, \pm 2, \dots$ .

### 3. Experimental results and discussion

In our experiment, a Nd:YAG nanosecond laser with wavelength  $\lambda_1 = 1064nm$  was used as the FW pump source. The phase of the FW was modulated by a XY Phase Series SLM (reflection mode) which produced by the BNS Company of America. The SLM has a resolution of  $512 \times 512$  pixels, each with a rectangular area of  $19.5 \times 19.5 \mu m^2$ . Without loss of generality, a 5mol% MgO:LiNbO<sub>3</sub> bulk crystal ( $10 \times 0.5 \times 10 mm^3$  in  $x \times y \times z$  dimensions) was used. The FW was kept as o-polarized and the type of SH interaction was oo-e ( $n_1 = 2.2322$  and  $n_2 = 2.3231$  according to the Sellmeier equation in [22]). In our experiment, the FW was imaged by a 4-f system after it modulated by the SLM. The beam waist of the FW  $a = 2mm$ . After the crystal, the FW was filtered out by a shortpass filter. Then, the generated SH was projected on a screen in the far-field and recorded by a camera.

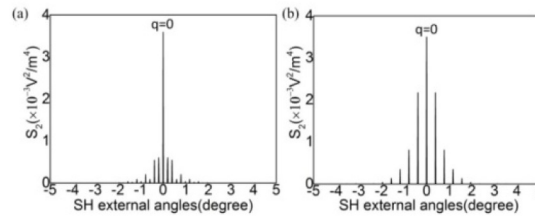


Fig. 2. (a)(b) The calculated spectral density of the SH field  $S_2(k_x, y)$  as the function of the external emission angle corresponding to the pure periodic modulation cases of  $\phi = \pi/2$  and  $\phi = \pi$ , respectively.

To the pure periodic modulation cases analyzed in [18], the spectral density of the SH field  $S_2(k_x, y)$  was calculated as a function of the external emission angle  $\beta_m$  shown in Fig. 2, where  $\sin \beta_m = \lambda_2 k_x / 2\pi$  and  $\lambda_2$  was the wavelength of the SH. Figure 2(a) and 2(b) are corresponding to the 1D cases of  $\phi = \pi/2$  and  $\phi = \pi$ , respectively, with the duty cycle  $D = 0.5$  and the period  $\Lambda = 156 \mu m$ . As shown in Fig. 2, the SH waves are emitted symmetrically along multiple directions. In the case of  $\phi = \pi/2$ , both even and odd orders are present. However, in the case of  $\phi = \pi$ , only even orders are present. The simulation results are in good agreement with the experimental analysis in [18].

To illustrate the difference of nonlinear Raman-Nath SHG in pure periodic and hybrid structure of the FW, we did a comparison between them both in numerical simulation and experiment in 1D case, as shown in Fig. 3. Figure 3(a) represents the hybrid structure of the FW, which combines periodic and random modulation in 1D case with  $\phi = \pi/2$ ,  $n = 8$  and  $\Lambda = 156 \mu m$ . Figure 3(b) is the calculated angular distribution of the multi-order Raman-Nath SH signal corresponding to Fig. 3(a). Figures 3(c) and 3(d) are the experimental results of the nonlinear Raman-Nath SH signals of pure periodic modulation and hybrid modulation, respectively. In Figs. 3(c) and 3(d), we can see that the SH are both a set of symmetrically distributed discrete spots. And these two structures have the same SH external radiation

angles. Nevertheless, the number of SH spots remarkably increase in hybrid structure. Even higher order SH spots are observed and this phenomenon doesn't occur in the pure periodic structure of the FW. It's clear that the experimental results shown in Fig. 3(d) are in well agreement with the numerical prediction shown in Fig. 3(b). What's more, the numerical prediction show that the density of SH field is enhanced notably in hybrid modulated structure. Hence, it indicates that the introduction of the random modulated structure has a big influence to the total SH intensity and has no effect on the radiation angles.

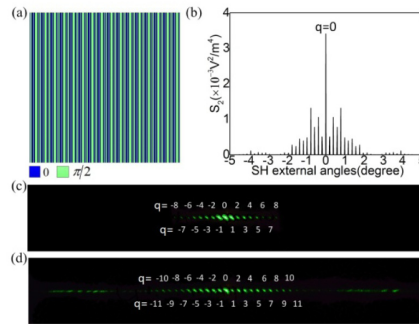


Fig. 3. (a) The hybrid structured FW with  $\phi = \pi/2$ . (b) The calculated spectral density of the SH field  $S_2(k_x, y)$  as the function of the external emission angle corresponding to (a). (c)(d) The experimental results of the Raman-Nath SH signals of the pure periodic and hybrid structure of the FW, respectively.

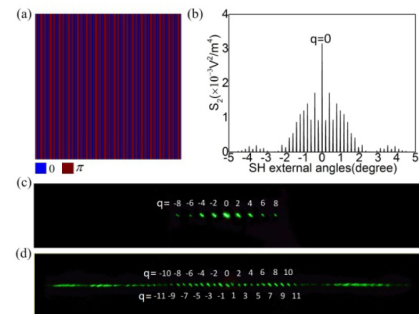


Fig. 4. (a) The hybrid structured FW with  $\phi = \pi$ . (b) The calculated spectral density of the SH field  $S_2(k_x, y)$  as the function of the external emission angle corresponding to (a). (c)(d) The experimental results of the Raman-Nath SH signals of the pure periodic and hybrid structure of FW, respectively.

We also investigated the nonlinear Raman-Nath SH generation of hybrid structure of the FW in 1D case with  $\phi = \pi$ ,  $n = 8$  and  $\Lambda = 156 \mu\text{m}$ . Figure 4(b) is the calculated angular distribution of the multi-order nonlinear Raman-Nath SH signal corresponding to Fig. 4(a). To the pure periodic modulation, only even orders are present shown in Fig. 4(c) and this is in agreement with the analysis in [18]. However, in hybrid modulated structure, different with the pure periodic structure, both odd and even orders of the SH spots were observed in the experiment, as shown in Fig. 4(d).

Not limited in 1D case, such hybrid structured FW in 2D situations were also investigated. Figure 5(a) is the ring shaped hybrid structure of the FW, in which the phase is sharply modulated from 0 to  $\pi/2$  in 2D structure. Figure 5(b) is the calculated spectral density of the multi-order nonlinear Raman-Nath SH signal corresponding to Fig. 5(a). Figure 5(c) is the experimental result of this structure. As shown in Fig. 5(b) and 5(c), from 1 to 7-order SH rings are observed in experiment, which is much more than the pure periodic modulation in

the same period. The SH of different order rings are circularly symmetric with uniform intensity distribution and the center spot is the brightest.

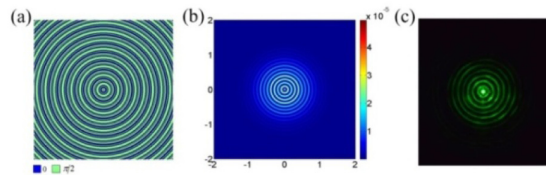


Fig. 5. (a) The ring shaped hybrid structured FW with  $\phi = \pi/2$ . (b) The calculated spectral density of the SH field  $S_2(k_x, k_y, y)$  as the function of the external emission angle corresponding to (a). (c) The experimental results of the Raman-Nath SH signals of the hybrid modulation.

Finally, the relationship between the total intensity of the SH and the degree of randomness of the structure is investigated. The randomness of the FW structure is marked by the number of the random bar, as shown in Fig. 1. The experimental results of seven different randomness of the structure are shown in Fig. 6, in which more random bar in each period means higher degree randomness of the aperiodic part. As shown in Fig. 6(a), the density of the SH spots is increasing as the bar number increased. Therefore, the intensity of the SH increased with the increase of the degrees of randomness, as shown in Fig. 6(b). With the increasing of  $n$ , more reciprocal vectors can be provided by the FW. Thus, more SH spots can be generated and this lead to the higher total intensity of the SH.

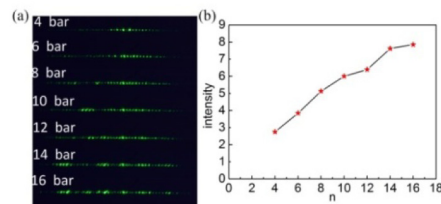


Fig. 6. (a) The experimental results of different randomness of the structure. (b) The intensity of the SH as a function of the number of random bar.

#### 4. Conclusion

In summary, we numerically and experimentally investigated the nonlinear Raman-Nath SHG of hybrid structured FW whose phase modulation combine periodic and random structure. The SHG of both 1D and 2D hybrid structured FW have been investigated. The results show that the introduction of the random modulation to the FW can provide richer reciprocal vectors than the pure periodic structure. Consequently, more diffraction spots and higher conversion efficiency can be obtained. Besides, the total intensity of the SH is increasing with the degree of randomness of the structure. This study not only enriches the family of SHG of the structured FW, but also has potential application in dynamically controlling SH wave in arbitrary directions.

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