International Journal of Nonlinear Optical Physics
Vol. 2, No. 2 (1993) 221–227




# HIGH-ORDER DIFFRACTION IN PHOTOREFRACTIVE SBN:Ce DUE TO NON-SINUSOIDAL GRATINGS FORMED BY BEAMS OF COMPARABLE INTENSITY

HONG-JUN ZHANG,* ZE-SUN TANG,** THOMAS R. MOORE and ROBERT W. BOYD
*The Institute of Optics, University of Rochester, Rochester, New York 14627, USA*

Received 25 June 1992

We have observed multiple-order diffraction from photorefractive gratings formed by two input beams of comparable intensity that interfere in a single crystal of SBN:Ce. High-order diffraction occurs both in the direction of two-beam coupling gain and in the opposite direction. These results are in good agreement with the predictions of a theoretical model that interprets high-order diffraction as a form of Raman-Nath scattering from a highly anharmonic diffraction grating that is formed by a light intensity distribution having a large depth of modulation.

## 1. Introduction

The appearance of spatial subharmonics and high-order spatial harmonics in photorefractive scattering excited by two incident beams has recently received considerable attention.[1–5] In particular, several groups have studied the consequences of the use of light with an intensity having a large depth of modulation to form the photorefractive grating, and they conclude that scattering should occur in a number of integer orders, both towards and away from the direction of photorefractive gain.[6–9] These predictions are in contrast to that of Ref. 4, which describes a mechanism by which high-order scattering should occur only in directions in which the scattered light experiences gain by means of photorefractive two-beam coupling. Here we report our observation of high-order scattering, in both the gain and non-gain directions, due to non-sinusoidal photorefractive gratings created by a sinusoidal intensity pattern with a large depth of modulation. Our results are in good agreement with the predictions of a theoretical model we describe below.

*Permanent address: Institute of Physics, Chinese Academy of Sciences, Beijing 100080, China.
**Permanent address: Department of Physics, University of Aeronautics and Astronautics, Beijing 100080, China.

## 2. Experiment

The experimental apparatus used in our investigation is shown in Fig. 1. A beam of light at a wavelength of 514 mm from an argon ion laser was split into two parallel beams of nearly equal intensity (1:1.06) by a plane-parallel plate. The beams were then passed through a single focusing lens of long focal length causing them to cross within the photorefractive crystal. The two beams were linearly polarized in the plane of incidence, which also contained the $c$-axis of the crystal. The confocal parameter of each beam was significantly greater than the thickness of the crystal and the total incident intensity at the crystal was $\sim$ 200 mW/cm$^2$. The crystal was a 1.85 mm thick piece of SBN, doped with 0.1% Ce. The maximum value of the refractive index variation resulting from the photorefractive effect was measured by two-wave mixing experiments and found to be $\Delta n = 2.3 \times 10^{-5}$. The total transfer of energy between the beams due to two-beam coupling was less than 10%. The beams transmitted through the crystal exhibited the effects of high-order diffraction. Integer orders were clearly visible but we saw no evidence of subharmonics. The power in each diffracted order was measured in the far field by a power meter and a lock-in amplifier. The results of such a measurement are shown in Fig. 2 for the case in which the grating period had a value of 200 $\mu$m. In plotting this graph, we have normalized all reflectivities to the value obtained for the first diffracted order in the gain direction of the two-beam coupling process; we arbitrarily call this order the *positive* first order of diffraction. During our investigation we saw no nonlinear dependence of the intensities of the diffracted orders on the incident intensity; however, we intentionally maintained a very low laser power in order to minimize thermal effects.

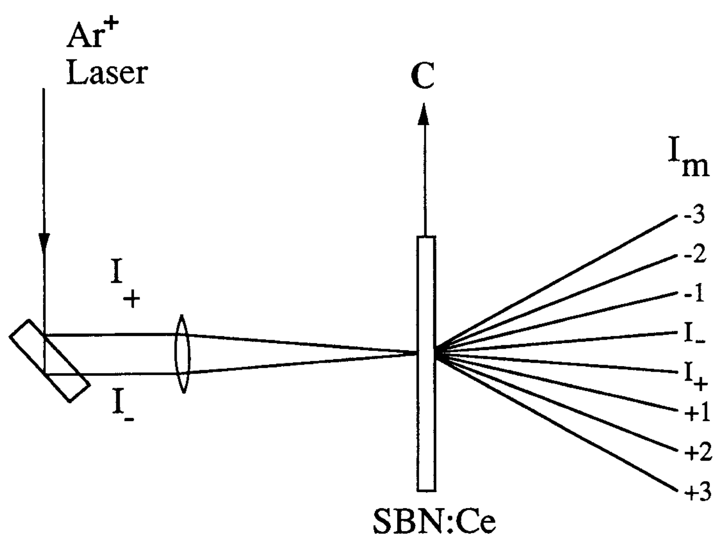


**Fig. 1.** Experimental arrangement.

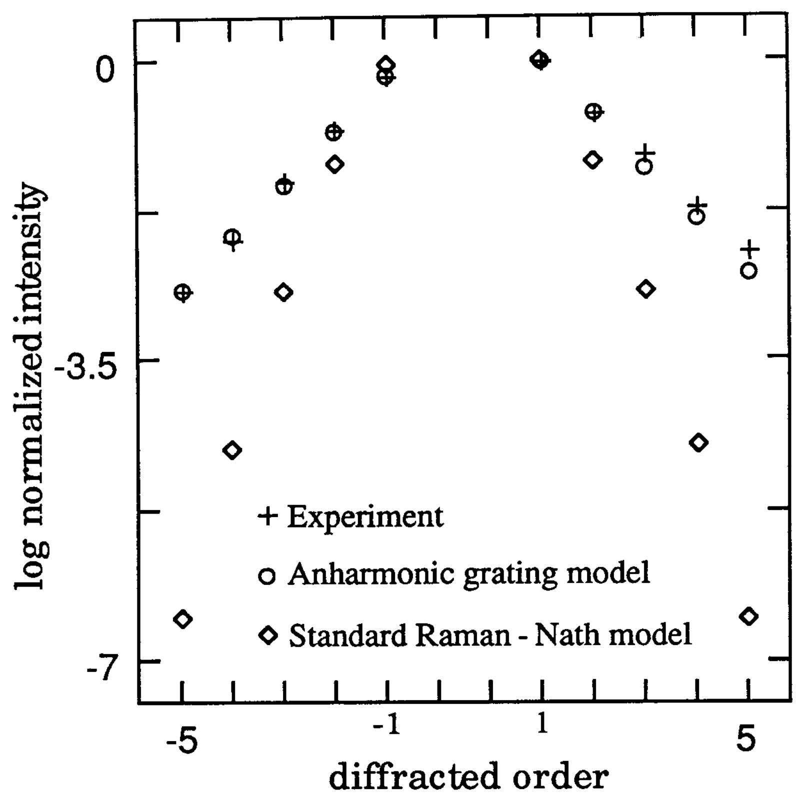


**Fig. 2.** Relative intensities of the diffracted orders normalized to that of the first positive order. The crosses represent experimental results for a grating period of ~ 200 $\mu$m, circles represent the theory presented in the text for the case $E_D/E_q = 0.25$, and diamonds represent the predictions of theory for a purely sinusoidal index grating (i.e. Raman-Nath scattering).

## 3. Theory

We begin by assuming that two coherent beams of intensities $I_+$ and $I_-$ intersect at some small angle $\theta$ within a thin photorefractive crystal. The total light intensity within the crystal can then be represented as

$$I(x) = I_+ + I_- + 2\sqrt{I_+I_-}\cos(k_g x)\,, \tag{1}$$

where $k_g = 2nk_0 \sin(\theta/2)$ is known as the grating wave number and $k_0$ is the vacuum wave number of the incident light. Equation (1) may also be written as

$$I(x) = I_0[1 + m\cos(k_g x)]\,, \tag{2}$$

where $I_0 = I_+ + I_-$ denotes the sum of the intensities of the incident waves and where $m = 2(I_+I_-)^{1/2}/I_0$ denotes the modulation index.

We next consider the form of the photorefractive response for the intensity distribution given by Eq. (2). We use the band transport model of Kukhtarev *et al.*[10] This model describes the interaction of light with the crystal in terms of the continuity equation, the carrier rate equation, the current equation, and Poisson's equation, which are given respectively by

$$\frac{\partial n}{\partial t} = \frac{\partial N_D^+}{\partial t} + \frac{1}{e}\frac{\partial j}{\partial x}, \tag{3a}$$

$$\frac{\partial N_D^+}{\partial t} = sI(x)(N_D - N_D^+) - \gamma_r n N_D^+, \tag{3b}$$

$$j = e\mu n E - k_B T \mu \frac{\partial n}{\partial x}, \tag{3c}$$

$$\frac{\partial E}{\partial x} = \frac{-4\pi e}{\varepsilon}(n + N_A - N_D^+). \tag{3d}$$

Here we have assumed that electrons are the dominant charge carrier, that $n$ is the number density of charge carriers, $N_D$ is the total number density of donor sites, $N_D^+$ is the number density of the ionized donors, $j$ is the current density, $s$ is the photoionization cross section, $\gamma_r$ is the recombination rate, $-e$ is the charge of an electron, $\mu$ is the electron mobility, $E$ is the local electric field within the crystal, $k_B$ is Boltzmann's constant, $T$ is the temperature, $\varepsilon$ is the dielectric constant of the crystal, and $N_A$ is the number density of charges that compensate $N_D^+$ when the crystal is in the dark. In writing the rate equation we have assumed that $N_A \ll N_D$ and that the rate of spontaneous excitation is small compared to that of photo-excitation.

Under steady state conditions with no externally applied electric field and assuming that $n \ll N_D^+$, these equations reduce to the set

$$EI(x)\left[\frac{(N_D - N_D^+)}{N_D^+}\right] + \frac{E_D}{k_g}\frac{d}{dx}\left[\frac{I(x)(N_D - N_D^+)}{N_D^+}\right] = 0 \tag{4a}$$

and

$$\frac{dE}{dx} = \frac{4\pi e}{k_g \varepsilon}(N_D^+ - N_A), \tag{4b}$$

where we have introduced the diffusion field $E_D = k_B T k_g / e$. There are no known closed-form solutions to Eqs. (4), and approximate solutions are typically found by assuming that the modulation index $m$ is much smaller than unity. Under these conditions, the induced electric field $E(x)$ consists of only one harmonic component. Recently, however, these equations have been numerically integrated for certain specific cases, and predictions have been made concerning the form of the induced electric field under conditions of high modulation depth.[6,9,11] In particular, Vachss and Hesselink,[9] expanding on the work of Moharam *et al.*,[6] have numerically shown that for the case in which the diffusion field is much weaker than the maximum space-charge field the electric field can be approximated by the expression

$$E(x) = E_D\left(\frac{m\sin(k_g x)}{1 + a\cos(k_g x)}\right) \tag{5a}$$

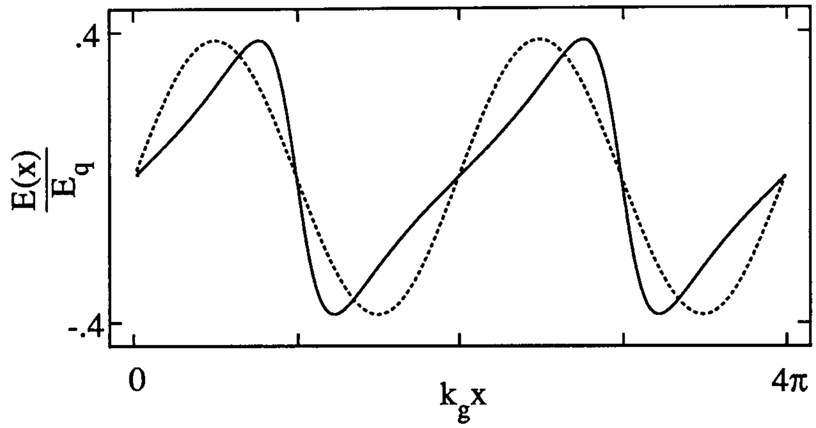


**Fig. 3.** Spatial variation of the static electric field as predicted by Eqs. (5) for $m = 1$ and $E_D/E_q = 0.25$. The dashed line is a plot of $\sin(k_g x)$ and is intended as an aid in noting the harmonic distortion present in the actual photorefractive grating.

where

$$a = m - \frac{E_D}{E_q}(2m-1)\,, \tag{5b}$$

and where $E_q$ is the maximum space-charge field given by

$$E_q = \frac{4\pi e}{k_g \varepsilon}\left[\frac{N_A(N_D - N_A)}{N_D}\right]. \tag{5c}$$

Figure 3 shows the spatial variation of the electric field predicted by Eqs. (5) for the case (which closely approximates that of our experiment) in which $E_D/E_q = 0.25$ and in which $m = 1$. For purposes of comparison, a purely sinusoidal electric field pattern is also shown in Fig. 3. Clearly evident is a significant increase in the higher Fourier components compared to the purely sinusoidal modulation that occurs in the limit of small modulation.

We next consider the nature of the modification of the incident light waves in propagating through a photorefractive crystal in which the induced space charge field is of the form given by Eqs. (5). Under our experimental conditions, the grating formed in the photorefractive crystal is a thin grating in the sense that it satisfies the inequality $L \tan\Phi \gg \Lambda$, where $L$ is the crystal thickness, $\Phi$ is the angle of incidence relative to the lines of constant refractive index, and $\Lambda$ is the grating period. For this reason, the scattering process occurs under the same conditions as Raman-Nath scattering, although our situation differs from that considered by Raman and Nath in that in our case the disturbance in refractive index is not purely harmonic. In particular, the local variation in refractive index is proportional to $E(x)$ and is given explicitly as

$$\Delta n(x) = -(1/2)n^3 r_{eff} E(x)\,, \tag{6}$$

where $n$ is the index of refraction of the crystal and $r_{eff}$ is the effective electro-optic coefficient. Consequently, in propagating through the crystal each of the incident beams of light, of amplitude $A_{\pm}(x) = A_{\pm}^{0} \exp[i(\mathbf{k}\cdot\mathbf{r} - \omega t)]$, will acquire the additional phase $\phi(x) = \Delta n(x)\omega L/c$, and consequently each transmitted beam will be described by

$$A_{\pm}(x) = A_{\pm}^{0} \exp[i(\mathbf{k}\cdot\mathbf{r} - \omega t + \phi(x))]\,. \tag{7}$$

The angular distribution of the scattered light in the far field can be determined by calculating the Fourier transform of the near-field distribution $A(x) = A_{+}(x) + A_{-}(x)$. The predictions of such a procedure, with the maximum value of $\Delta n$ taken as our measured value of $2.3 \times 10^{-5}$, are also presented in Fig. 2 and are seen to be in excellent agreement with the experimental results. In performing this comparison, we have taken the ratio $E_D/E_q$ to be a free parameter and have found that the best agreement is obtained using a value 0.25.

If we had assumed that the photorefractive grating contained only a single harmonic component, as is assumed by many elementary theories of photorefractive response, the theoretical predictions would reduce to those of the normal Raman-Nath treatment of scattering from a thin grating. In such a case, since $\phi(x)$ contains a single harmonic component, Exp. (7) for $A_{\pm}(x)$ can be expanded explicitly in a Fourier series, where the square of each Fourier coefficient gives the relative power of each scattered component of each incident wave. Since the far-field intensity pattern has contributions from each incident wave, the total far field pattern is proportional to

$$I_{\pm n} = I_{\pm 0}(0)J_n^2(F) + I_{\mp 0}(0)J_{n+1}^2(F)\,. \tag{8}$$

where $F = 2\pi\Delta nL/\lambda$, $\lambda$ being the wavelength of the incident beams.

It is possible to include the effects of truncating the photorefractive grating at the beam edges by numerically calculating the Fourier coefficients. However, using our experimental parameters, the inclusion of the truncation of the grating resulted in no significant change in the predicted intensities of the first five diffracted beams from those predicted by Eq. (8). The predictions of Eq. (8) are also shown in Fig. 2, and clearly are not in agreement with the experimental results.

## 4. Conclusions

We have observed high integer-order diffraction from thin, photorefractive gratings induced by the anharmonic electric field distribution produced by a sinusoidal intensity pattern with a high depth of modulation. Our experimental results agree well with theoretical predictions.

## References

1. D. J. Webb and L. Solymar, Observations of spatial subharmonics arising during two-wave mixing in BSO, *Opt. Comm.* **74**, 386 (1990).
2. S. Mallick, B. Imbert, H. Ducollet, J. P. Herriau and J. P. Huignard, Generation of spatial subharmonics by two-wave mixing in a nonlinear photorefractive medium, *J. Appl. Phys.* **63**, 5660 (1988).

3. F. Vachss and L. Hesselink, Selective enhancement of spatial harmonics of a photorefractive grating, *J. Opt. Soc. Am.* **B5**, 1814 (1988).
4. L. B. Au and L. Solymar, Higher diffraction orders in photorefractive materials, *J. Quantum Electron.* **24**, 162 (1988).
5. L. B. Au and L. Solymar, Amplification in photorefractive materials via a higher order wave, *Appl. Phys.* **B45**, 125 (1988).
6. M. G. Moharam, T. K. Gaylord and R. Magnusson, Holographic grating formation in photorefractive crystals with arbitrary electron transport lengths, *J. Appl. Phys.* **50**, 5642 (1979).
7. E. Ochoa, F. Vachss and L. Hesselink, Higher-order analysis of the photorefractive effect for large modulation depths, *J. Opt. Soc. Am.* **A3**, 181 (1986).
8. L. B. Au and L. Solymar, Space-charge field in photorefractive materials at large modulation, *Opt. Lett.* **13**, 660 (1988).
9. F. Vachss and L. Hesselink, Nonlinear photorefractive response at high modulation depths, *J. Opt. Soc. Am.* **A5**, 690 (1988).
10. N. V. Kukhtarev, V. B. Markov, S. G. Odulov, M. S. Soskin and V. L. Vinetskii, Holographic storage in electro-optic crystals. I. Steady state, *Ferroelectrics* **22**, 949 (1979).
11. A. Bledowski, J. Otten and K. H. Ringhofer, Photorefractive hologram writing with modulation 1, *Opt. Lett.* **16**, 672 (1991).