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Multi-order nonlinear diffraction in second harmonic generation

S. M. Saltiel [a,b] , D. Neshev [a], W. Krolikowski [a], Ady Arie [c], Ole Bang [d], Yu. S. Kivshar [a]

a)Nonlinear Physics Center and Laser Physics Center, ANU, Canberra, Australia
b)Department of Physics, Sofia University "St. Kl. Ohridski", Sofia 1164, Bulgaria
c) School of Electrical Engineering, Tel-Aviv University, Tel Aviv 69978, Israel
d) DTU Fotonik, Department of Photonics Engineering,Technical University of Denmark,Denmark
wzk111@rsphysse.anu.edu.au

Abstract: We analyze the emission patterns in the process of second harmonic (SH) generation in $\chi^{(2)}$ nonlinear gratings and identify for the first time, to the best of our knowledge, the evidence of Raman-Nath type nonlinear diffraction in frequency doubling processes.

In the linear optics, it is well established that propagation of light beam in periodic refractive index structures leads either to Bragg diffraction when the full vectorial phase matching condition is satisfied or to Raman-Nath diffraction otherwise. In the latter case since the full vectorial phase matching is not fulfilled, a multi-order diffraction takes place similar to the case of light scattering by acoustic waves. Analogous process takes place in parametric process such as second harmonic (SH) generation in periodically poled nonlinear media. In this, so called, nonlinear Bragg diffraction [1] the SH beam is emitted at a specific angle, determined by the vectorial phase-matching condition $2\mathbf{k}_1 + \mathbf{G}_n = \mathbf{k}_2$, where $\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave-vectors of the fundamental frequency (FF) and the SH beams, respectively, and $\mathbf{G}_n$ denotes the grating vector of the periodic modulation of the sign of nonlinearity [see Fig.1(a), left]. If this vectorial condition is not satisfied, a multi-order nonlinear diffraction of the SH should be seen [Fig.1(a), right] instead, in analogy to the linear Raman-Nath diffraction. Nonlinear Bragg diffraction is well documented in the literature and has been observed in both naturally laminated crystals and artificially created nonlinear one-dimensional $\chi^{(2)}$ gratings. Conical, 13th order nonlinear Bragg diffraction has also been recently observed in annularly poled Z-cut crystals [2]. On the other hand, to the best of our knowledge the nonlinear Raman-

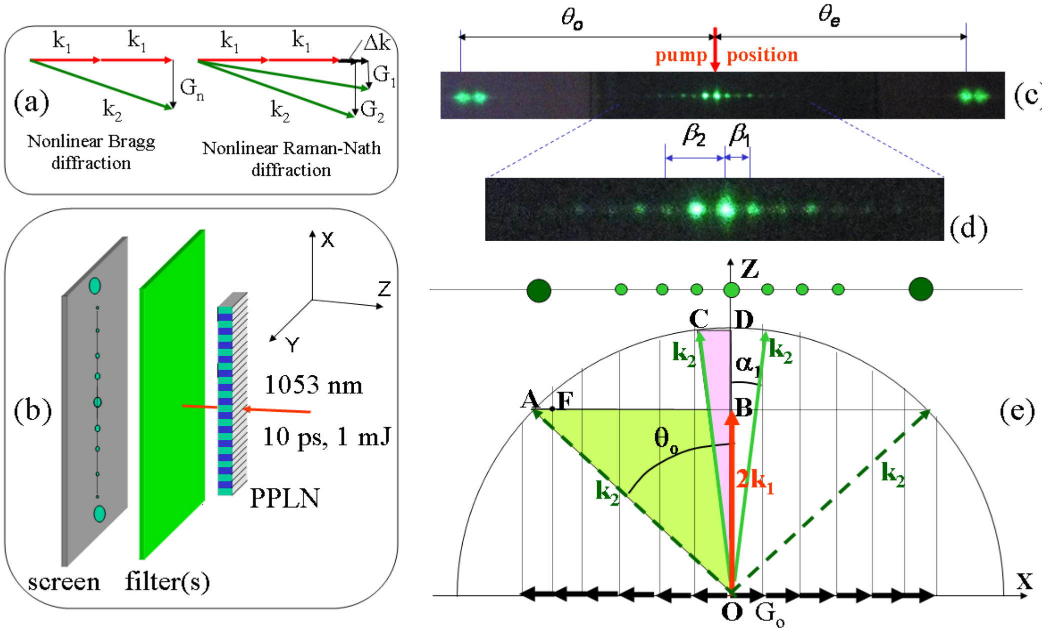

Fig. 1. (a) Phase matching conditions for nonlinear Bragg and Raman-Nath diffraction. (b) Schematic of the experiment. (c,d) Experimentally recorded nonlinear diffraction patterns generated in one dimensional QPM grating in periodically poled $LiNbO_3$ with period 14.6 μm. (e) Phase-matching diagrams for SH diffraction in z-cut periodically poled samples when pump ($2\mathbf{k}_1$) is perpendicular to QPM grating vector $\mathbf{G}_0$.

Nath diffraction has not been reported so far.

Here we report on the nonlinear diffraction of beams on a one-dimensional nonlinear $\chi^{(2)}$ grating, realized by periodic spatial alternation of the sign of the relevant components of the $\chi^{(2)}$ tensor. We observe, for the first time, multiple-order nonlinear diffraction in the SHG processes, which represent the Raman-Nath contribution to the process. In addition, we analyze the diffracted SH beams whose angular positions are governed by the longitudinal phase matching conditions only and can be classified as non-integer order nonlinear diffraction, that have never been observed before, according to our knowledge.

In our experiments we use periodically poled lithium niobate (PPLN) and stoichiometric lithium tantalate samples. To observe nonlinear diffraction, we employ the experimental setup shown in Fig. 1(a) with picosecond laser and a regenerative amplifier as a light source. The laser delivers 10 ps pulses at 1053 nm with repetition rate of 20 Hz and output pulses energy close to 1 mJ. The laser beam is focused to a spot size ~ 400 μm in the plane of the crystal and propagates along Z axis or at an angle with respect to the same axis.

In Fig. 1(c,d) we show an example of the SH diffraction patterns obtained in a one dimensional $\chi^{(2)}$ grating (period Λ = 14.6 μm) in the PPLN sample. The SH diffraction array consists of two types of patterns: (i) central diffraction spots grouped around the position of the pump and (ii) peripheral diffraction spots situated relatively far from the pump at two sides of the diffraction array – utmost left and right pairs of spots on Fig. 1(c). It has been verified that these two neighbor peripheral spots at each side of the diffraction array are orthogonally polarized. Those located further away from the pump are polarized in the x-y plane, while the other two are polarized in the x-z plane. Since the pump is ordinary polarized we identify the interactions that are responsible for these SH spots as O_1O_1-O_1 and O_1O_1-E_2, respectively. The tensorial $\chi^{(2)}$ components responsible for the O_1O_1-O_2 process are d_{22} and d_{21}, while for the O_1O_1-E_1 interaction the second-order polarization term involves additionally both d_{32} and d_{31} components. Similar O_1O_1-O_2 spots have been observed recently in periodically poled KTP [3]. However in contrast to our experiments, they were due to second-order nonlinearities existing only in the domain walls.

The observed nonlinear diffraction patterns can be explained by employing the phase-matching conditions shown in Fig. 1(e). The general vectorial phase-matching condition $2\mathbf{k}_1 + \mathbf{G}_n + \Delta k = k_2$ is valid for both types of the SHG diffraction spots (the central and the peripheral ones). Figure 2 shows one of the phase-matching diagrams (triangle OCD) that governs the first-order SH diffraction patterns, appearing close to the pump. It is constructed by the vectors k_2, $2k_1$, BD = Δk_{TPM}, and one of the grating vectors G_0. The angular positions of the SH diffraction spots in the central part are defined only by the transverse phase-matching (TPM) conditions that leads to the following relation: $\sin \beta_m = m\lambda_2/\Lambda$, (m = 1, 2,…) , which is, in fact, identical as that describing position of maxima in the Raman Nath linear diffraction process.

Let us turn now to the peripheral diffraction spots which appear far away from the pump [see Fig. 1(c)]. Our experiments with four different samples show that the angular positions of these diffraction spots θ_o and θ_e [defined in Fig.1(c)] do not depend on the poling period. This is in striking contrast to the direct dependence on Λ of the central spots. The measured angles can be explained only if we assume that the directions of these SH beams are defined uniquely by the longitudinal phase matching (LPM) conditions represented by the triangle like, for instance, OAB (see Fig.1e) constructed from vectors $\mathbf{k}_2$, $\mathbf{2}k_1$, FA=Δk_{LPM} and m numbers of grating vectors, BF = m$\mathbf{G_0}$, (m = 1, 2,…). For normal incidence, the measured angles θ_o and θ_e are found to be in accordance with those derived on the basis of LPM conditions that lead to the expressions for the SH Cherenkov radiation $\cos\theta_{o,e} = 2k_1/k_{2o,e}$. For the pump propagating at an angle γ with respect to the z axis, the phase matching conditions for the LPM diffraction spots are the same except that $2k_1$ needs to be replaced by $2k_1\cos\gamma$. Although the period of the poled grating does not appear in the equations for calculating the angles $\theta_{o,e}$ for the LPM diffraction spots, we have to point out that these strong LPM signals do not appear in homogeneous crystal without periodic poling.

In conclusion, we have observed the second-harmonic patterns generated due to multi-order nonlinear diffraction. We have found that the angular positions of the SH beams are defined only by a ratio of the wavelength to the grating period as in Raman-Nath diffraction in linear optics. The diffraction patterns include additional spots defined only by the longitudinal phase matching which are not observed without the nonlinear grating. For this reason we believe that these peripheral SH spots can be identified as non-integer order nonlinear Bragg diffraction. As it was shown recently [4] similar type SH radiation can be used for a single shot full reconstruction of the femtosecond pulses.

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