Generation of Spatial Subharmonic Gratings in the Absence of Photorefractive Beam Coupling

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Using a new experimental geometry, we have proved for the first time that the generation of spatial subharmonic gratings in photorefractive crystals is not dependent on optical nonlinearity. We present results which confirm that the subharmonic gratings result from a parametric excitation of ultra low-frequency eigenmodes of a crystal by a time modulated fundamental grating.

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In 1988, Mallick *et al.* first reported the generation of so-called spatial subharmonic beams in photorefractive crystals [1]. In their experiment, two plane waves were incident on a crystal of $Bi_{12}SiO_{20}$ (BSO) in the conventional two wave mixing geometry: The applied electric field and the K vector of the fundamental grating arising, due to interference between the two beams, were in the [001] direction, and both beams were incident on the (−110) face. They found that under certain experimental conditions, in addition to the familiar higher-order diffracted beams, there also arose other beams which appeared between the two pump beams emerging from the crystal and which corresponded to diffraction of the incident light from integer submultiples of the fundamental grating vector.

Initial attempts at explaining the origin of these subharmonic beams were reliant on the application of familiar photorefractive concepts: the buildup of periodic space-charge fields generated by pairs of light waves, followed by light diffraction, and mediated by the linear electro-optic effect. It was suggested that the additional light beams might arise as a result of the high gain amplification of light scattered in certain directions [2–4]. Unfortunately, this approach was not able to provide predictions compatible with the accumulated experimental data [1,2,5,6].

More recently it has been predicted theoretically that the subharmonic gratings can result from a parametric excitation of weakly damped space-charge waves by a time modulated fundamental grating [7]. In the framework of this theory, the material nonlinearity (coupling together of different gratings of space charge) is responsible for the generation of new spatial frequencies including subharmonic gratings. Light diffraction and the consequent optical beam coupling (which we term here the optical nonlinearity) only enables the visualization of these gratings and does not contribute to their initial generation. The theoretical model relies on a number of assumptions concerning the nonlinear dynamics of the space-charge field and for this reason cannot be regarded as conclusive.

Currently, then, there is a dilemma: Either the subharmonic gratings are a specific manifestation of the photorefractive effect or they should be classed as a new effect of the nonlinear excitation of eigenmodes of the medium. In the latter case a wide range of semiconductors with a high value of the lifetime-mobility product (which appears to be critical for the onset of subharmonics [8]), such as GaAs, CdTe, and Ge, would be a subject of scientific interest. In particular, investigations would no longer need to be restricted to electro-optic materials.

In this Letter we provide conclusive evidence that the subharmonic generation is not in fact dependent on the photorefractive optical nonlinearity. We describe a new experimental geometry ideally suited to the study of spatial subharmonics which does not permit the pump beams to participate in any photorefractive beam coupling, while allowing direct measurement of the fundamental and subharmonic grating strengths. Experimental results are provided which show for the first time the presence of a subharmonic grating in this situation. This definitely identifies the material's nonlinearity as being responsible for the origin of subharmonics. Last, we demonstrate that the experimental data agree well with the main predictions of the theory [7,8].

The experimental arrangement is shown schematically in Fig. 1. Two linearly polarized beams of equal intensity, pump 1 and pump 2, are derived from the same expanded and collimated argon ion laser beam of wavelength 514.5 nm and are incident in a vertical plane on the (001) face (5×10 mm^2) of a $5 \times 10 \times 11$ mm^3 BSO crystal with absorption coefficient $\alpha = 1.1$ cm^{-1}. The pump beams are symmetrically angled about the normal to the face so as to write a grating with a period $2\pi/K =$ 20 μm. The electric field vectors of the two pump beams are both initially in the [−110] direction. The total pump

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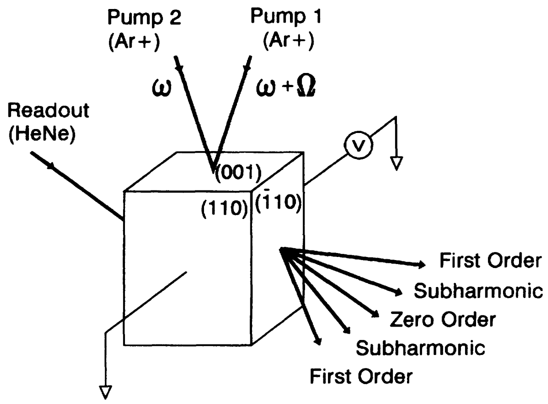

FIG. 1. Experimental setup. Only the readout beam Bragg matched to the fundamental grating and the corresponding diffracted beams are shown.

intensity I_0 was in the range 15 to 70 mW/cm^2. An electric field E_0 of up to 7 kV/cm was applied using silver loaded paint electrodes on the (110) faces (5 × 11 mm^2) of the crystal. Both the grating vector and the electric field are aligned in the [110] direction. The electro-optic properties of BSO (point group 23) are such that in this arrangement the pump light experiences no field induced changes of optical dielectric tensor and therefore is not involved in any grating-mediated wave mixing.

Pump 2 is shifted in frequency by reflection off a moving mirror, driven by a serrodyne voltage providing 28 dB (electrical) suppression of unwanted spectral components. Two readout beams are derived from a HeNe laser with a wavelength of 632.8 nm. Each beam has a diameter of 3 mm and an intensity of 1.5 mW/cm^2, and both are incident on the (−110) face of the crystal (10 × 11 mm^2) centered between the two electrodes and about 3 mm below the face through which the pump beams enter. One HeNe beam is angled so as to be Bragg matched to the fundamental grating generated by the pump beams and the other is correspondingly Bragg matched to the subharmonic grating with a grating vector $K/2$. In this way the diffraction efficiency of the respective gratings may be monitored at the same time.

With this geometry, as expected, it was not possible to obtain any sign of beam coupling involving the pump beams and no subharmonic beam appeared between them. Nevertheless, it was possible to detect the presence of both fundamental and subharmonic gratings using the HeNe probe beam. This excludes the possibility that the photorefractive optical nonlinearity contributes to the origin of the subharmonic grating and conclusively demonstrates the fundamental role of the material nonlinearity. Figures 2 and 3 show the diffraction efficiency of the fundamental and subharmonic gratings as a function of pump beam angular detuning frequency Ω. The error bars indicate the repeatability of the experiment. From the two figures it may be seen that the subharmonic exists only within a restricted interval of the frequency detuning. The subharmonic grating strength is maximized by a detuning frequency Ω_s, approximately three times larger than the detuning frequency Ω_f necessary to maximize the fundamental grating strength. Near to its maximum the subharmonic grating dominates the fundamental one.

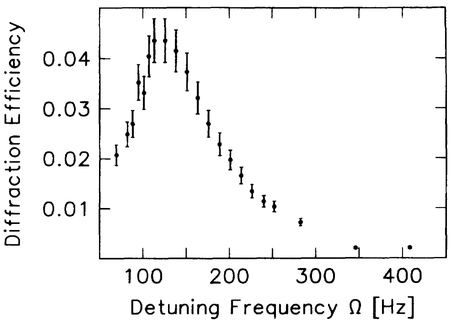

FIG. 2. Diffraction efficiency of the fundamental grating as a function of pump beam angular detuning frequency Ω in a BSO crystal. $I_0 = 20$ mW/cm^2, $E_0 = 7$ kV/cm. For details see the text.

It was also experimentally observed that the optimal detuning frequencies Ω_s and Ω_f are directly proportional to the total pump intensity I_0 over the intensity range quoted earlier. At the same time, they are inversely related to the external field E_0 beginning from the value 2 kV/cm where the subharmonic grating first appears.

Let us compare the experimental data with the theory of Refs. [7] and [8]. This theory provides the following dependence of the (angular) frequency of space-charge waves ω_k on the wave vector k:

$$\omega_k = \frac{e}{\varepsilon\varepsilon_0 k E_0}\,\frac{\alpha I_0}{h\nu}\,. \tag{1}$$

Here e is the absolute value of the electron charge, $\varepsilon\varepsilon_0$ the dielectric constant, and $h\nu$ the energy of a pump photon. The optimum of an excitation of the fundamental grating with wave vector K corresponds to the condition of linear resonance and occurs at a detuning frequency

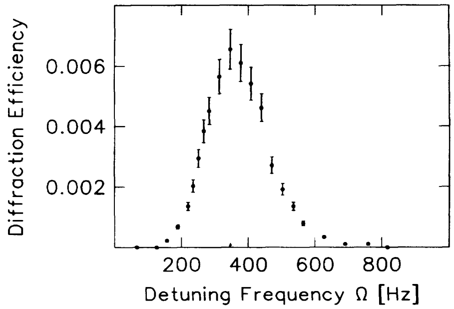

FIG. 3. Diffraction efficiency of the subharmonic grating as a function of pump beam angular detuning frequency Ω in a BSO crystal. $I_0 = 20$ mW/cm^2, $E_0 = 7$ kV/cm. For details see the text. Note the difference in scale on the horizontal axis in comparison with Fig. 2.

given by $\Omega_f = \omega_K$. The optimum detuning frequency Ω_s for the excitation of a subharmonic grating with wave vector $K/2$ takes place when the parametric resonance condition, $\Omega_s = 2\omega_{K/2}$, is fulfilled. According to Eq. (1), $\Omega_s = 4\Omega_f$.

Using Eq. (1) and the experimental parameters relating to Figs. 2 and 3, one can estimate Ω_s and Ω_f to be 350 and 88 Hz, respectively. The first value agrees excellently with the experimental result while the second one is exceeded slightly by the experimental value of 115–125 Hz. This difference may be explained by a positive nonlinear frequency shift for the fundamental grating (see Ref. [9]), the peak amplitude of which exceeds considerably that of the subharmonic grating. As is clear from Eq. (1), the eigenfrequency is directly proportional to the total pump intensity and inversely proportional to the external field. These features are in full agreement with the experimental behavior of the frequency detunings Ω_s and Ω_f. The absence of the subharmonic generation for sufficiently low external fields correlates with the theoretically predicted decrease of the quality factor of the eigenmodes with decreasing applied field.

It may be seen that our experimental results not only identify the material nonlinearity as being responsible for the generation of subharmonics but also make clear the generation mechanism: parametric excitation of weakly damped space-charge waves by the moving fringe pattern. This mechanism, involving electron transport in a space-charge field, is not restricted to photorefractive materials—it may occur in materials that do not exhibit the linear electro-optic effect. In particular, the appearance of subharmonics or related phenomena should occur in a wide class of semiconductors with long drift length.

In conclusion, using a new experimental arrangement, we have proved that the generation of subharmonic gratings in photorefractive crystals is not related to the optical nonlinearity and is caused by the direct nonlinear interaction between a moving fundamental grating and space-charge waves. We propose to apply the new optical configuration to a detailed study of different subharmonic regimes in BSO crystals (using, in particular, an alternating applied field [10]) and also to new promising materials such as CdTe and GeAs.

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