Onset of subharmonics generated by forward wave interactions in $Bi_{12}SiO_{20}$

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The threshold conditions, under which a spatial subharmonic beam may arise when a $Bi_{12}SiO_{20}$ crystal is illuminated by two pump beams, are investigated. It is shown that a nonlinear theory based on the material equations leads to good qualitative agreement with experiments

Two-wave mixing phenomena in photorefractive media may be usually described using the model first proposed by Kukhtarev *et al.*[1,2] In this model, the variation of the amplitude of the incident light beams due to the presence of a refractive index modulation is determined by the so-called field equations, while the formation of the index modulation in response to the intensity standing wave pattern is governed by the material equations. Recently, experiments with $Bi_{12}SiO_{20}$ (BSO) have shown that, under the appropriate conditions, one or more spatially subharmonic beams may arise between the pump beams at angles that are simple fractions of the interpump angle.[3] In a previous publication,[4] these subharmonic beams have been characterized as a function of detuning and applied electric field, but as yet the mechanism responsible for the origin of these subharmonics has not been identified to any degree of certainty.

It has been proposed by Ringhofer and Solymar[5] that the mechanism responsible for the generation of subharmonics is selective amplification of a signal scattered in the right direction. In the present letter we suggest another mechanism based on the material equations and check experimentally the predictions of the model for the onset of the second subharmonic (the one appearing halfway between the two pump beams).

The basic experimental setup is shown in Fig. 1. A spatially filtered, expanded and collimated beam of light from an Ar^+ laser, emitting at 514.5 nm, was split in two. A small frequency shift was applied to one of the beams by reflecting the beam from a mirror mounted on a PZ driven by a serrodyne waveform. The two beams were then directed at the $1\bar{1}0$ face of a crystal of BSO. This face measured 6×7 mm^2 and the length of the crystal was 10 mm. In addition, collimated light from a Kr^+ laser emitting at 530.9 nm was also incident on the crystal. This light and the light from the Ar^+ laser were mutually incoherent, and hence the intensity of the light from the Kr^+ laser could be varied in order to adjust the modulation of the intensity standing-wave pattern produced by the pump beams.

The ratio of the intensities of the two beams from the Ar^+ laser was 1.2:1, and the total incident power density was maintained at 5 mW/cm^2. The beams were polarized perpendicular to the 001 face and the electric field was applied perpendicular to the 110 face of the crystal. The pump beams were angled so as to write a grating within the BSO with a period of 20 μm. The intensity of the subharmonic was measured by focusing the subharmonic beam onto a power meter using a 50 cm focal length convex lens.

With the Kr^+ laser initially switched off, the subharmonic beam intensity was recorded as a function of detuning frequency at a number of different electric fields. In this way, for each value of the applied electric field, the optimum intensity was obtained. This process was repeated with the Kr^+ laser contributing different amounts of power to the total incident intensity, and therefore varying the modulation. The total range was from 0.37 to 1 for modulation and from 1.6 to 10 kV/cm in applied electric field. We found that the subharmonic power was always higher as either the modulation or the applied electric field increased. The maximum power in the subharmonic was 15 μW for a modulation of 1 and an electric field of 10 kV/cm. The aim of this letter is to study the threshold, i.e., our interest is in the values of modulation and electric field which, at optimum detuning, cause the appearance of a subharmonic beam. There is bound to be some uncertainty concerning the measurement of these threshold values for the reason that there is always some light scattered in the direction of the subharmonic beam even in the absence of an applied electric field. Our criterion for the threshold is that the newly appearing subharmonic intensity just equals the scattered light intensity, and any small increase in electric field or modulation will cause a large increase in subharmonic power. The threshold values measured according

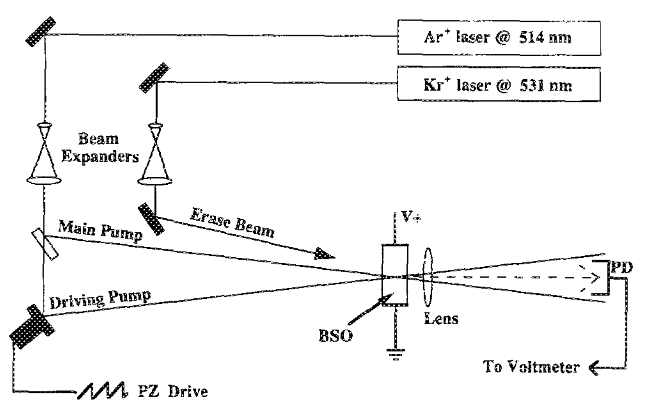

FIG. 1. Experimental arrangement used to find threshold field for the appearance of subharmonics at a given modulation.

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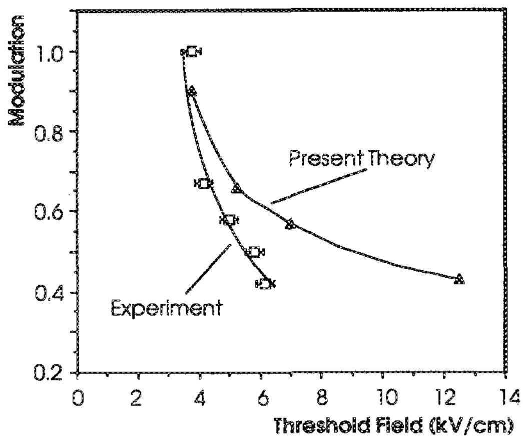

FIG. 2. Plot of threshold field for the appearance of subharmonics against modulation. (Open squares) experimental results; (filled triangles) theoretical results.

to this definition are plotted in Fig. 2. It may be seen that lower modulation will suffice for the generation of subharmonics as the electric field increases.

The analysis is based on the theoretical papers of Au and Solymar[6,7] in which the nonlinear material equations are solved by a numerical method yielding the Fourier components of the electron density, ionized space-charge density, and space-charge field.

We must assume in the calculation that there is an input modulation both at the fundamental component m_1 and at the subharmonic component $m_{1/2}$. For most values of the parameters, it is found that the fundamental components have negligible influence upon the subharmonic components. But at certain values of m_1 when the applied electric field increases we find that, for a small increase of the applied electric field there is a large increase in the subharmonic space-charge field. Taking a concrete case, we find for example that for a certain set of materials parameters (given below) and for a fundamental modulation of $m_1 = 0.9$ and applied field = 3.5 kV/cm, the subharmonic space-charge field is 3.3×10^{-4} kV/cm, whereas for an electric field of 4 kV/cm the subharmonic space-charge field increases to 1.5 kV/cm, an increase by a factor of 5000. This is a clear indication of a sudden onset of interaction between the fundamental and the subharmonic component of the space-charge field. We claim then that the threshold applied electric field is 4 kV/cm.

The numerical calculations are rather laborious to perform. In order to calculate accurately the subharmonic component of the space-charge field as many as 26 higher harmonics are required in the Fourier expansion. It can take as long as 4 h of CPU time on our DEC VAX 11/780 computer to obtain the maximum value of the space-charge field for a given electric field.

Figure 2 shows the relationship between the threshold electric field and m_1, using the following crystal parameters: Ionized acceptor density, 10^{22} m^{-3}; refractive index, 2.62; electro-optic constant, 3.4×10^{-12} m/V; temperature, 293 K; recombination coefficient, 1.6×10^{-17} m^3/s; electron mobility, 10^{-5} m^2/V s; photoionization constant, 2×10^{-5} m^2/J; donor density, 10^{25} m^{-3}. The total incident power density was 5 mW/cm^2, the wavelength 514.5 nm, and the grating spacing 20 μm. Examination of Fig. 2 shows that the theory is in good qualitative agreement with the experimental results.

We believe that there are two distinct mechanisms responsible for the emergence of subharmonics. One is based on the field equations concerned with the coupling of the waves as described in Ref. 5. The other one is based upon the materials equations, the subject of the present letter. In practice, both mechanisms are bound to be present. The latter one is responsible for the sudden rise of the subharmonic and the former one causes its further amplification. When we consider the threshold conditions only then, we believe, the materials equations will suffice, and this seems to be confirmed by the good qualitative agreement between theory and experiment.

As mentioned before, the numerical calculations are rather demanding so the chances seem to be rather low that the theory, as it stands, could accurately predict the subharmonic output. We can, nevertheless, indicate by a particular example the mechanism by which we believe the subharmonic arises.

Let us assume that at the input of the crystal we have some scattering in the direction of the subharmonic beam. We take the scattered power as 5×10^{-13} times smaller than the input power leading to a modulation (between the subharmonic beam and one of the pump beams) of 10^{-6}. Let us further assume that the conditions are right for the onset of subharmonic interaction, the applied electric field being 4 kV and the parameters as given previously. Then, due to the interaction between the fundamental grating and the subharmonic grating, we have an enormous intensity gain coefficient, calculated from the space-charge field as $\Gamma = 6729$/cm. Assuming that this gain coefficient remains constant for a distance of 6.8 μm, the subharmonic power increases by a factor of 100 and the subharmonic modulation increases by a factor or 10 to $m_{1/2} = 10^{-5}$. It is a very important characteristic of the interaction mechanism discussed that the interaction decreases as the intensity of the subharmonic component increases. For $m_{1/2} = 10^{-5}$, the gain coefficient may be calculated as $\Gamma = 2128$/cm. We need now a distance of 21.6 μm for the modulation to increase to $m_{1/2} = 10^{-4}$. The corresponding gain coefficient is now $\Gamma = 673$/cm and a distance of 68.4 μm is needed for a further increase in modulation to $m_{1/2} = 10^{-3}$. Thus, in a distance of less than a tenth of a mm, we predict an amplification of the subharmonic beam of six orders of magnitude. As $m_{1/2}$ rises further the fundamental-subharmonic interaction disappears, and we are left eventually by the usual gain coefficient which in our present case amounts to about 20/cm.

As it turns out the results of the calculation are sensitive to the choice of the parameters particularly to the value of mobility. Considering further the large number of parameters, the uncertainty about their exact value, and the complexity of the numerical calculations we feel that further experimental work needs to be done which might give clues for simplifying the theoretical approach. Assuming that the crystal would produce a subharmonic, the ideal experiment that could shed a lot more light upon the mechanism is as follows. Measure both the threshold conditions and the subharmonic output as a function of detuning for a set of BSO crystals grown under identical conditions and having thicknesses of (say) 1 mm, 2 mm, 3 mm, 5 mm, and 10 mm. If our hypothesis is correct, that is, the threshold conditions are determined by the materials equations only, then these threshold values will be more or less the same for each member of the set. The maximum subharmonic output would, on the other hand, be strongly dependent on the thickness of the crystal since the field equations predict an exponential increase with thickness.

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