# Critical Enhancement of Photorefractive Beam Coupling

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We show that a hybridization of the optical and material nonlinearities takes place near the threshold of the subharmonic generation in photorefractive crystals. It results in a critical (with a singularity) enhancement of the rate of spatial amplification of light waves and leads to a variety of new optical critical phenomena.

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Photorefractive nonlinear phenomena attract at present considerable research interest because of the simplicity of their realization, a wealth of physical potentialities, and prospects for applications. Formation of various spatial solitons [1] and periodic light patterns [2], beam collapse [3], and generation of spatial subharmonics [4–9], known also as period doubling, are examples of the "hot" photorefractive studies. Each of the above nonlinear phenomena, being generic, possesses, nonetheless, many important features which cannot be expected *a priori*.

Most of the nonlinear effects in photorefractive media (in particular, formation of solitons and periodic light patterns) are caused by photorefractive optical nonlinearity. Its mechanism includes two basic elements—formation of light-induced space-charge field owing to migration of photoexcited carriers and diffraction from the replica of the refractive index attributed to the linear electrooptic effect [4]. Nonlinearity of the material equations for charge separation is usually negligible or of secondary importance.

Generation of spatial subharmonics, found in cubic crystals of the sillenite family ($Bi_{12}SiO_{20}$, $Bi_{12}TiO_{20}$, $Bi_{12}GeO_{20}$) and in semiconductor CdTe [10], occupies an exceptional place among the photorefractive phenomena. In appearance, it is an optical critical effect. Above a certain threshold value of the contrast of an interference light pattern, $m > m_{\mathrm{th}}$, the response of the medium spontaneously loses its periodicity. In addition to the spatial frequency of the initiating light pattern, $K$, and the higher harmonics, $2K, 3K, \ldots$, new spatial frequencies arise in the Fourier spectrum of the space-charge field; often this new element of symmetry is the first subharmonic $K/2$; see Fig. 1. Diffraction from the spatial subharmonics gives rise to new outgoing light beams.

The peculiarity of this photorefractive phenomenon lies in the fact that it is caused by the material (not optical) nonlinearity. The subharmonic generation is qualified nowadays as the parametric excitation of weakly damped low-frequency eigenmodes, space-charge waves (SCWs) [5,6]. The threshold value of the contrast, $m_{\mathrm{th}}$, corresponds here to the balance between the rates of decay and nonlinear pumping of SCWs. Direct evidence of this concept is the observation of the subharmonic generation in a special optical configuration [7] that does not allow any diffraction processes for the initiating light beams but allows diffraction for auxiliary weak testing beams.

In the general case, the material and optical nonlinearities act together. The recognition of the leading role of the material nonlinearity has shadowed the importance of the optical nonlinear processes. The purpose of this Letter is to show for the first time that the highly different optical and material nonlinearities are strongly hybridized near the threshold of subharmonic generation. This means that the optical and low-frequency eigenmodes are mutually coupled and cannot be treated separately. The threshold behavior of SCWs leads here to new optical critical phenomena. The key point of these phenomena is the singularity of the rate of spatial amplification for light waves.

To the best of our knowledge, the found critical enhancement has no direct analog. It is somehow similar to the effect of critical opalescence [11]. The last one is distinguished, however, by its linear character. In some sense, the singularity of the spatial amplification is similar to that for phase conjugate mirrors [12]. Our effect does not require, however, counterpropagation of interacting waves and the corresponding positive feedbacks; it is compatible with forward propagation geometries.

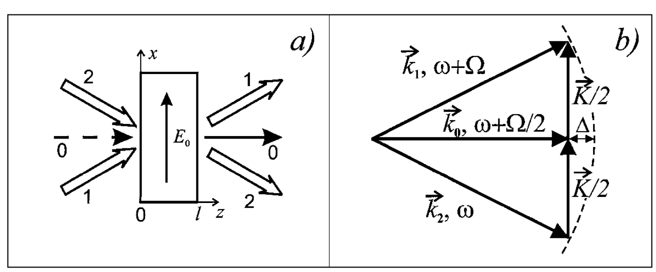


FIG. 1. (a) Schematic of subharmonic generation and critical enhancement. In the first case, only pump beams 1 and 2 are incident onto the crystal to form an initiating interference light pattern. (b) The corresponding wave vector diagram, the grating vector of the pump light pattern is $\vec{K} = \vec{k}_1 - \vec{k}_2$.

  

The essence of the case under study can be explained with the help of Fig. 1. Two coherent pump beams, 1 and 2, detuned in frequency by $\Omega$, propagate symmetrically near the $z$ axis in a photorefractive crystal allowable for the subharmonic generation [5,6]. An electric field $E_0$, necessary for weak damping of SCWs, is applied along the $x$ axis. Additionally, to the pump beams a weak signal beam 0, detuned by $\Omega/2$, propagates along the $z$ axis. A small deficit of its wave vector, $\Delta$, shown in Fig. 1b, can be treated within the approximation of slowly varying amplitudes. Pump beams 1 and 2 form a running fundamental grating of space-charge field with the grating vector $\vec{K}$ and the period $\Lambda = 2\pi/K$. The light pairs 1, 0 and 0, 2 both contribute to a buildup of a $\vec{K}/2$-grating running with the same velocity, $\Omega/K$, as the fundamental one.

Let now the detuning $\Omega$ meet the condition of the parametric resonance $\Omega = 2\omega_{K/2} = 4\omega_K$, where $\omega_K \propto I_0/KE_0$ is the eigenfrequency of SCW with a wave vector $\vec{K}$ parallel to the applied field and $I_0$ is the total pump intensity [5]. This condition corresponds to excitation of the subharmonic $K/2$ owing to the material nonlinearity.

In addition to the material nonlinearity, we take into account the relevant optical nonlinear processes, namely diffraction of pump beams 1 and 2 from the subharmonic $K/2$ into the central beam 0. Redistribution of the energy between the pump beams owing to diffraction from the fundamental grating is a secondary process in our case because this grating is nearly in phase with the pump interference pattern [4,5]; in addition, this redistribution can be diminished by choosing a sufficiently thin crystal.

Within the conventional description of SCWs and diffraction in photorefractive crystals [4–6], the governing equations for the dimensionless amplitude of the subharmonic, $e_{K/2} = E_{K/2}/E_0$, and the dimensionless amplitude of the central light beam, $a_0$, have the form

$$(\partial_t + \gamma_{K/2})e_{K/2} = i|\omega_{K/2}| \times \left(\frac{2}{3} a_1 a_2^* e_{K/2}^* - a_0 a_2^* - a_1 a_0^*\right), \tag{1}$$

$$(\partial_z - i\Delta)a_0 = -i\kappa(a_1 e_{K/2}^* + a_2 e_{K/2}), \tag{2}$$

with $\gamma_{K/2}$ the damping constant ($\gamma_{K/2} \ll |\omega_{K/2}|$), $\kappa = \pi n^3 r E_0/\lambda$, $n$ the refractive index, $r$ the effective electrooptic coefficient, $\lambda$ the light wavelength, and $a_{1,2}$ the dimensionless amplitudes of the pump waves. All the light amplitudes $a_{0,1,2}$ are normalized to the square root of the pump intensity so that $|a_1|^2 + |a_2|^2 = 1$ and the contrast of the pump interference pattern $m = 2|a_1 a_2^*|$.

By itself, each of the terms in Eqs. (1) and (2) is known. The first term on the right-hand side of Eq. (1) accounts for the nonlinear coupling between the subharmonic $K/2$ and the fundamental grating whose dimensionless amplitude is $e_K \simeq a_1 a_2^*/3$. This term is responsible for the subharmonic generation above the threshold, $m > m_{\mathrm{th}}$, where $m_{\mathrm{th}} = 3/Q$ and $Q = |\omega_{K/2}|/\gamma_{K/2}$ is the quality factor for SCW with a wave vector $K/2$ [5]. The last two terms describe the excitation of the $K/2$ grating by the light pairs 0, 2 and 1, 0. They were taken into account (without the nonlinear term) in the early studies of the subharmonics [13]. The structure of Eq. (2) is typical for optical parametric 4W processes [14] but these were never combined with the resonance low-frequency nonlinear interactions. Taken together, Eqs. (1) and (2) describe mutual coupling of the amplitudes $a_0, a_0^*, e_{K/2}$, and $e_{K/2}^*$; we can speak therefore of an interference (hybridization) of two different parametric processes related to the light waves and to SCWs.

Let us consider the consequences of the found hybridization. In steady state we have from Eq. (1), for the subharmonic amplitude,

$$e_{K/2} = -\frac{iQ}{1-\xi^2}\left[a_0 a_2^*\left(1 - \frac{2}{3}iQ|a_1|^2\right) + a_0^* a_1\left(1 - \frac{2}{3}iQ|a_2|^2\right)\right], \tag{3}$$

where $\xi = m/m_{\mathrm{th}}$. One sees that this amplitude increases drastically when approaching the threshold of the subharmonic instability. Expression (3) indeed yields an optical coupling between the amplitudes $a_0$ and $a_0^*$ in Eq. (2). In what follows, we restrict ourselves to the case $m < m_{\mathrm{th}}$, i.e., to $\xi < 1$. In the opposite case, the subharmonic $K/2$ and the output central beam 0 exist without any optical seed.

By substituting $e_{K/2}$ given by Eq. (3) into Eq. (2) and assuming that $a_0, a_0^* \propto \exp(\Gamma z)$, we obtain for the rate of optical spatial amplification, $\Gamma$,

$$\frac{\Gamma_\pm}{\kappa Q} = \frac{W}{1-\xi^2} \pm \sqrt{\left(\frac{W\xi}{1-\xi^2}\right)^2 - \frac{\Delta}{\kappa Q}\left(\frac{6}{Q}\frac{\xi^2}{1-\xi^2} + \frac{\Delta}{\kappa Q}\right)}, \tag{4}$$

where $W = |a_1|^2 - |a_2|^2 = \pm\sqrt{1-m^2}$ is the normalized difference of the pump intensities. The occurrence of two solutions for $\Gamma$ is due to the mentioned coupling between $a_0$ and $a_0^*$; the signs + and − correspond to two different values of the phase of the complex amplitude $a_0$. To return to the conventional photorefractive coupling, we should put $\xi = 0$. A positive sign of the product $\kappa W$ favors strongly the spatial amplification. This sign is controlled by the pump ratio and polarization. Below we focus our attention on this optimum case.

The most important feature of Eq. (4) is that $\Gamma_+(m)$ grows infinitely when approaching the threshold. In the close vicinity of the threshold the mismatch $\Delta$ becomes unimportant and we have

$$\Gamma_+ \simeq \frac{\kappa Q\sqrt{1-m_{\mathrm{th}}^2}}{1-\xi}. \tag{5}$$

Physically, this limit corresponds to the Bragg diffraction of the pump beams to the central beam.

It is remarkable that the effect of the mismatch, $\Delta = \pi\lambda/4n\Lambda^2$, on the rate $\Gamma$ is negative when $\kappa > 0$, $W > 0$ and it is positive when $\kappa < 0$, $W < 0$, $\Delta(1 - \xi^2) < 6|\kappa|\xi^2$. In the second case, the mismatch $\Delta$ compensates for the nonlinear correction to the wave vector $\vec{k}_0$. Optimization of the effect of the mismatch can always be performed using the dependence of the sign of the effective electro-optic coefficient $r$ on the pump polarization in cubic crystals [15,16].

The solid lines 1, 2, 3, and 4 in Figs. 2a and 2b show the dependencies $\Gamma'_\pm(m) = \Re\Gamma_\pm(m)$ for $\lambda = 514$ nm, $E_0 = 7$ kV/cm, the representative parameters of $Bi_{12}SiO_{20}$ crystals, $n_0 = 2.6$, $|r| = 4.6$ pm/V, $Q = 6$, and two values of the grating period $\Lambda$. The curves 1, 2 and 3, 4 refer to the cases $\kappa, W < 0$ and $\kappa, W > 0$, respectively. The dashed curves show the dependence $\Gamma'(m)$ with the neglected effect of the material nonlinearity.

One sees that the curves 1, 2, 3, and 4 coincide for sufficiently small $m$, when the square root in Eq. (4) is imaginary. With increasing contrast, the dependence $\Gamma'_\pm(m)$ exhibits a bifurcation; the point of bifurcation is closer to zero for the case $\kappa, W < 0$ most favorable for the spatial amplification. After the bifurcation, the upper branch experiences a pronounced rise and then tends to infinity. Increasing $\Lambda$ shifts the bifurcation point towards zero and makes the influence of the Bragg mismatch less pronounced.

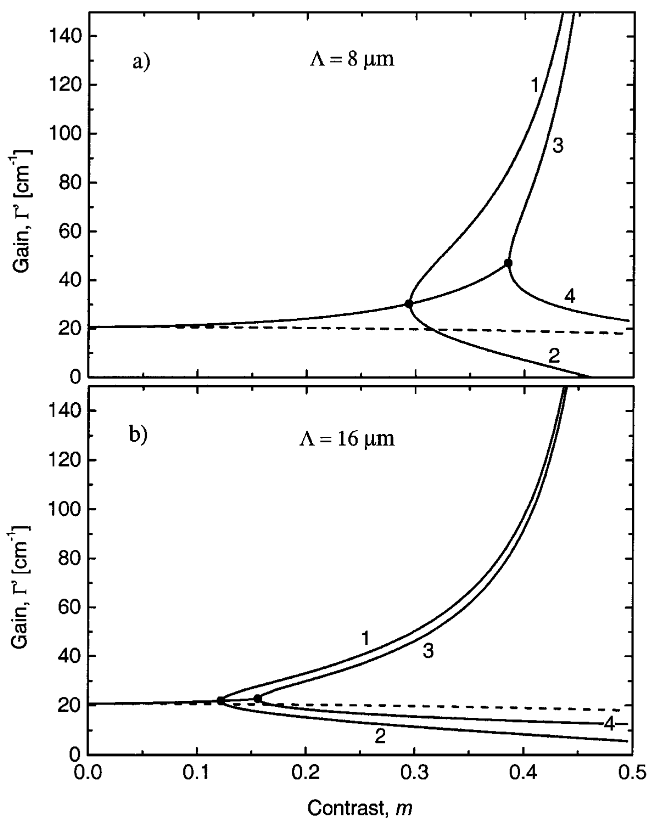


FIG. 2. Dependences $\Gamma'_\pm(m)$ for $\Lambda = 8$ $\mu$m (a) and 16 $\mu$m (b). The curves 1, 2 and 3, 4 correspond to the cases $\kappa < 0, W < 0$ and $\kappa > 0, W > 0$, respectively. The dashed curves are plotted by setting $\xi = 0$ in Eq. (4).

It is clear that the critical growth of $\Gamma(m)$ near the threshold has to be saturated on a sufficiently high level. Within the linear approximation in $a_0$, the relevant limitation is $\Gamma\Lambda < 1$. It means that the effective thickness of the recorded hologram, $\sim\Gamma^{-1}$, cannot be much smaller than the distance between the interference fringes. Within this restriction, the rate $\Gamma$ saturates at the level of $\sim 10^3$ cm$^{-1}$, which exceeds the rate of spatial amplification attainable in photorefractive materials without the critical enhancement.

The above results admit two important generalizations. First, instead of the subharmonic $\vec{K}/2$ one can consider two nonlinearly coupled SCWs with the wave vectors $\vec{K}_1$ and $\vec{K}_2 = \vec{K} - \vec{K}_1$ meeting the phase-matching condition $\Omega = \omega_{\vec{K}_1} + \omega_{\vec{K}_2}$. This case corresponds to a range of the frequency detuning where $\Omega > 4\omega_K$ [5]. Instead of a single central beam 0 one has to consider here two weak light beams with wave vectors $\vec{k}_1 + \vec{K}_{1,2}$; compare with Fig. 1b. Therefore, a wide interval of frequency detuning and a range of propagation directions are available for the critical enhancement. Second, instead of the moving grating technique one can exploit the so-called ac technique with an alternating applied field and a static pump interference pattern [15]. This method is also useful for parametric excitation of SCWs [4,10] and is fully compatible with the critical enhancement.

Now we turn to a discussion of optical manifestations of the critical enhancement and its connection with available experimental data. Experiments with $Bi_{12}SiO_{20}$ in the optical configuration shown in Fig. 1a were performed about ten years ago without any relation to critical phenomena and to the subharmonic generation [17]. The authors obtained surprisingly high (up to $10^4$) values of the amplification factor in spite of the strong negative effect of spatial inhomogeneity [18] reducing the net interaction length. One can expect (but cannot prove) that the critical enhancement was involved there in the spatial amplification. Special experiments with thin samples are needed to obtain direct evidences of the critical behavior.

To show the impact of the critical enhancement on the photorefractive optical phenomena, we remind one that high values of the spatial amplification lead usually to optical oscillations because of various optical feedbacks between output and input [12]. Often, these oscillations are accompanied by the phase conjugation. Actually, a great part of the expected applications of the photorefractive nonlinearity is based on optical oscillations [4]. We expect that the critical enhancement can be successfully implemented in many oscillation schemes resulting in new optical critical phenomena based on the hybridized optical-material nonlinearity.

To make the above assertion clearer, we consider a simple example relevant to the subharmonic generation.

Let the optical feedback be caused by the successive reflections of the 0 beam from the rear and front crystal faces perpendicular to the $x$ axis; see Fig. 1a. In this case, the condition for an optical oscillation is $\Gamma > \Gamma_{\mathrm{th}} = \alpha + l^{-1} \ln R^{-1}$, where $\alpha$ is the light absorption coefficient, $l$ is the crystal thickness, and $R$ is the reflection coefficient. This condition means that the optical losses during a round trip in the cavity are covered by the spatial amplification. With $\Gamma \gtrsim 10^2\ \mathrm{cm}^{-1}$ it can easily be fulfilled even for very thin samples. No optical seed is necessary for the operation of such a ring oscillator.

Within the above model, the central beam appears at the output spontaneously below the genuine threshold of the subharmonic generation ($m < m_{\mathrm{th}}$) and it looks like an apparent manifestation of the subharmonic $K/2$. The apparent threshold of the subharmonic generation (related to the optical oscillation) is sensitive indeed to the sign of $W = |a_1|^2 - |a_2|^2$. This feature explains the corresponding experimentally detected difference [19]. We expect that, by distinguishing between the apparent and genuine thresholds of the subharmonic generation, one can explain other still unclear features of this phenomenon [8,9].

The found critical enhancement has clear merits for the practical use. Extremely high values of the gain enable one to use thin samples instead of thick ones in order to meet the necessary conditions for spatial amplification. This, in turn, removes the problems related to rotation of the polarization plane owing to optical activity and to attenuation of light owing to volume absorption [18]. Miniaturization of the samples allows one to increase additionally the light intensity and to reduce therefore the time of the photorefractive response.

In conclusion, we have predicted the effect of hybridization of the optical and material nonlinearities in photorefractive crystals that leads to a dramatic enhancement of the rate of spatial optical amplification near the threshold of parametric excitation of SCWs. This critical phenomenon promises considerable advantages for the use of photorefractive materials in optical amplifiers and oscillators. It elucidates also previously unclear features of the subharmonic generation.

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