Steady-state analysis of ac subharmonic generation in photorefractive sillenite crystals

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The stationary solution is obtained for the photorefractive subharmonic gratings excited in crystals of the sillenite family by a standing light interference pattern and an applied ac electric field. We show that the main subharmonic with doubled spatial period may become unstable against excitation of the subharmonic with quadrupled spatial period. The threshold condition for this bifurcation is found. [S1050-2947(98)05808-9]

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Excitation of parametric waves can take place in various continuous media. This type of process is known for plasma waves [1,2], spin waves [3], and optical waves [4]. The latest example of excitation of this wave type is the so-called photorefractive parametric oscillation [5–8]. The special case of excitation of parametric waves in photorefractive media named subharmonic generation is the case that has attracted most attention since the first experimental verification showed the existence of such states. Photorefractive subharmonic generation refers to the effect in which a fundamental holographic grating recorded in a photorefractive crystal becomes unstable against the excitation of additional gratings with grating vectors assuming integer fractions $K/2$, $K/3$, or $K/4$ of the fundamental grating vector K. The first generation of photorefractive subharmonics was observed in a crystal of the sillenite family, $Bi_{12}SiO_{20}$, subjected to a running light interference pattern and a dc electric field [9] (dc case). Shortly after, generation of subharmonics was demonstrated by illuminating a sillenite crystal with a standing light pattern and applying an alternating (ac) electric field (ac case) [10,11]. An analytical theory based on the concept of space-charge waves [12] has allowed a description of the linear part of the problem, including the calculation of the threshold condition for the subharmonic generation in both cases [13,14]. In spite of the great amount of accumulated experimental data only few analytical treatments of the nonlinear stage of the subharmonic excitation have been made; see, e.g., Ref. [15], which contains analysis of the steady state for the simplest dc case, the $K/2$ subharmonic. The amplitudes of the saturating parametric waves were shown to be determined largely by the nonlinear frequency shifts by which also various stationary states beyond threshold as well as the feedback from these parametric waves to the fundamental wave may be described. Moreover, the concept of renormalization of the nonlinear coupling constant was introduced with the nonlinear theory. This is very important when analyzing the stability of the nonlinear regimes.

In this paper we give a stationary solution to the problem of subharmonic generation for the case where a square-wave ac electric field is applied to a sillenite crystal. The threshold condition for excitation of secondary subharmonic gratings with wave vectors $K/4$ and $3K/4$ is also found. Moreover, we explain the existence of two types of spatial domains measured experimentally recently and show that the mechanism of stabilization of the instability in the present ac case is different from that in the dc case.

We consider a standing light interference pattern in the form $I=I_0[1+m\cos(Kx)]$, where I_0 is the average light intensity, m is the contrast, and K is the fundamental grating vector. In addition, we assume that a square-wave field, $E_{ex}(t)$, is applied in the x direction (parallel to K) and is given by

$$E_{ex}(t)=E_0 p(t),$$

$$p(t)=\begin{cases}1, & nt_0<t<(n+1/2)t_0\\ -1, & (n+1/2)t_0<t<(n+1)t_0,\end{cases}$$

where n is an integer and t_0 is the period. In this case, we can restrict ourselves to the one-dimensional problem. In what follows we also assume that the amplitude of the induced space-charge field, $E(x,t)$, is small compared with the amplitude of the externally applied electric field, i.e., $|E|\ll E_0$. This enables us to apply a perturbational approach and, thus, to start from Eq. (33) of Ref. [12], which can be rewritten in the form

$$\frac{\partial^3 E}{\partial x^2 \partial t}+\left(\omega_0+\frac{k_B T}{q}\frac{\zeta I_0}{E_0^2}\right)\frac{\partial^2 E}{\partial x^2}-\frac{\zeta I_0}{E_0}p(t)\frac{\partial E}{\partial x}-\frac{1}{\mu\tau}\frac{\zeta I_0}{E_0^2}E=\zeta I_0\frac{\partial}{\partial x}\left[m\cos(Kx)-\frac{E^2}{E_0^2}\right], \tag{1}$$

where k_B is the Boltzmann constant, T is the absolute temperature, q is the elementary charge, and $\tau\mu$ is the lifetime mobility product for free electrons. The parameters ω_0 and ζ are given by $\omega_0=sI_0N_D/N_A$ and $\zeta=sqN_D/\varepsilon_0\varepsilon_s$, where s is the photoexcitation constant, N_D is the density of donors, N_A is the density of acceptors, and $\varepsilon_0\varepsilon_s$ is the permittivity of the crystal. The left-hand side of Eq. (1) is linear in E and determines the linear characteristics of the space-charge waves. The first term on the right-hand side is the effective driving force whereas the second term governs the nonlinear coupling.

By introducing the Fourier representation for E,

$$E(x,t)=\sum_{\kappa} E_0 e_\kappa(t)\exp(i\kappa x),$$

where $e_\kappa(t)$ is the dimensionless field amplitude and the summation is assumed over all physically actual values of the grating vectors κ, we can represent Eq. (1) in the form

$$\frac{\partial e_k}{\partial t}+[\gamma_k+i\omega_k p(t)]e_k=-i\omega_k\frac{m}{2}(\delta_{k,K}+\delta_{k,-K})+i\omega_k\sum_{k'} e_{k'}e_{k-k'}. \tag{2}$$

Here $\delta_{k,K}$ is the Kronecker symbol. The frequency ω_k and the damping coefficient, γ_k entering Eq. (2) are given by

$$\omega_k=\omega_0\frac{E_q(k)}{E_0},$$

$$\gamma_k=\omega_0\frac{E_q(k)E_M(k)+E_q(k)E_D(k)+E_0^2}{E_0^2}, \tag{3}$$

where $E_q(k)=qN_A/k\varepsilon_0\varepsilon_s$, $E_D(k)=k_BTk/q$, and $E_M(k)=1/k\mu\tau$ are the characteristic photorefractive fields. Throughout this paper we consider the case of high quality factor, $Q_k=|\omega_k/\gamma_k|\gg 1$, when the subharmonics can be generated [12]. This means that the inequalities $|E_M(k)+E_D(k)|\ll E_0\ll|E_q(k)|$ have to be fulfilled. Actually, these inequalities have already been used in deriving Eqs. (2) and (3). In the most interesting case for subharmonics generation, the temporal period of the ac field is much smaller than the grating formation time and much larger than the electron lifetime. Thus, we can adopt the period averaging procedure to Eq. (2); see, e.g., Ref. [16]. The equation for the amplitude $\overline{e_k}$, averaged over a period t_0, i.e., $\overline{e_k}(t)=\int_{t-t_0/2}^{t+t_0/2}e_k(t')dt'/t_0$, is given by

$$\frac{\partial\overline{e_k}}{\partial t}+\gamma_k\overline{e_k}=-i\omega_k\frac{m}{2}(\delta_{k,K}+\delta_{k,-K})+i\omega_k\sum_{k'}\overline{e_{k'}}\,\overline{e_{k-k'}}, \tag{4}$$

where the fast oscillating part of the field amplitude has been neglected since it is small, of the order $\omega_K t_0$, in comparison with $\overline{e_k}$.

Within the linear approximation Eq. (4) has the following steady state solution for the fundamental grating amplitude:

$$\overline{e_K}=-i\frac{m}{2}\frac{\omega_K}{\gamma_K}=-i\frac{mQ_K}{2},\quad \overline{e_K^*}=i\frac{mQ_K}{2},\quad K>0, \tag{5}$$

where the asterisk denotes the complex conjugate. This solution characterizes the initial periodic state for a sufficiently low contrast m, when the higher spatial harmonics, $2K$, $3K,\ldots$, are negligible [12,16]. According to Eq. (5) the fundamental grating has the amplitude $mQ_K/2$ and is shifted by $-\pi/2$ with respect to the light interference pattern.

Let us analyze the stability of the fundamental grating against the excitation of subharmonics with grating vectors k_1 and k_2 meeting the spatial synchronism condition $k_2=K-k_1$. Their amplitudes obey the following coupled equations:

$$\frac{\partial\overline{e_{k_1}}}{\partial t}+\gamma_{k_1}\overline{e_{k_1}}=2i\omega_{k_1}\overline{e_K}\,\overline{e_{K-k_1}^*},$$

$$\frac{\partial\overline{e_{K-k_1}^*}}{\partial t}+\gamma_{K-k_1}\overline{e_{K-k_1}^*}=-2i\omega_{K-k_1}\overline{e_K^*}\,\overline{e_{k_1}}. \tag{6}$$

To find the threshold for growth of subharmonics let us seek a solution to Eq. (6) in the form $\overline{e_{k_1}},\overline{e_{K-k_1}}\propto\exp(\nu t)$. From the condition of solvability of the resultant algebraic system we obtain two solutions for the increment of the instability $\nu_\pm$:

$$\nu_\pm=-\frac{\gamma_{k_1}+\gamma_{k_1-K}}{2}\pm\left[\left(\frac{\gamma_{k_1}-\gamma_{k_1-K}}{2}\right)^2+4\omega_{k_1}\omega_{k_1-K}|e_K|^2\right]^{1/2}. \tag{7}$$

If ν_+ is positive, the amplitudes of the subharmonic gratings experience exponential growth starting from noise level. The threshold equation for this instability reads $\nu_+=0$ from which, using Eq. (5), we find the threshold value of the contrast, $m_{\rm th}=Q_K^{-1}\sqrt{Q_{k_1}^{-1}Q_{K-k_1}^{-1}}$. This defines $m_{\rm th}$ as a function of k_1: The minimum of this function specifies the lowest threshold of the contrast for a given K. Using Eq. (3) for the quality factor one can show that the minimum takes place at $k_1=K/2$ if the inequality $E_0^2<4(2+\sqrt{5})E_M(K)E_q(K)-E_D(K)E_q(K)$ is fulfilled. In this case the threshold of subharmonic instability is given by [12]

$$m_{\rm th}=\frac{1}{Q_KQ_{K/2}}. \tag{8}$$

Otherwise, the minimum is reached at k_1 between 0 and K. Since Q_K and $Q_{K/2}$ are both much greater than unity, the threshold of the contrast is very small.

When the contrast exceeds the threshold value, Eq. (5) for $\overline{e_K}$ is no longer valid and we have to take into account the feedback from the subharmonics to the fundamental grating. In this case Eq. (4) leads to the following generalization of Eq. (5):

$$\overline{e_K}=-iQ_K\left(\frac{m}{2}-\overline{e_{K/2}^2}\right). \tag{9}$$

Together with Eq. (6) this equation has two steady-state solutions given by

$$\overline{e_K}=-i\frac{1}{2Q_{K/2}},\quad \overline{e_{K/2}}=\overline{e_{K/2}^*}=\pm\left(\frac{m-m_{\rm th}}{2}\right)^{1/2}. \tag{10}$$

One can see from Eqs. (5) and (10) that the amplitude of the fundamental grating grows linearly as a function of m for $m<m_{\rm th}$ and remains constant for $m>m_{\rm th}$. The phase $\varphi_K=\arg(\overline{e_K})$ remains equal to $-\pi/2$ above threshold. The subharmonic amplitude is zero below the threshold, $m<m_{\rm th}$;

above the threshold it grows as the square root of $m-m_{\mathrm{th}}$. The phase of the subharmonic grating, $\varphi_{K/2}=\arg(\overline{e_{K/2}})$, meets the phase matching condition $2\varphi_{K/2}=\varphi_K+\pi/2$, which can be found from the steady-state version of Eq. (6) with $k_1=K-k_1=K/2$. Since phase is indeterminate by a factor of 2π, the subharmonic phase is either 0 or π, which is seen from Eq. (10). The choice between 0 and π is arbitrary and, consequently, two types of spatial domains with these two values of phase can coexist simultaneously. Such domains have been found experimentally [17]. The phase matching condition elucidates the mechanism of stabilization of the instability for the ac case. Since $\overline{e_{K/2}^2}$ is positive, the nonlinear term in Eq. (9) tends to decrease the amplitude of the fundamental grating. When $|\overline{e_K}|$ is reduced down to its threshold value $(2Q_{K/2})^{-1}$, the increment of the subharmonics goes to zero, i.e., $\nu_+=0$ and, as a result, the growth of subharmonics is stopped. Freezing the fundamental grating amplitude at the threshold level owing to the feedback from the subharmonics is the essence of the stabilization mechanism in the ac case. This mechanism is also known from other fields of nonlinear science [18] and it differs basically from the mechanism of stabilization for the dc case.

Increasing the contrast can result in a new instability against excitation of additional gratings with wave vectors k_1', $K-k_1'$, $K/2-k_1'$, and $K/2+k_1'$. From the above analysis we have learned that the threshold of the subharmonics instability is usually lowest for the $K/2$ grating and we therefore assume that the threshold of the subsequent instability is lowest for the grating with $k_1'=K/4$. This is also supported by experiments [10]. Using this assumption and Eq. (4) one can obtain the coupled equations for the amplitudes $\overline{e_{K/4}},\overline{e_{3K/4}}$ in the form

$$\frac{\partial\overline{e_{K/4}}}{\partial t}+\gamma_{K/4}\overline{e_{K/4}}=2i\omega_{K/4}(\overline{e_{K/2}}\overline{e_{K/4}^*}+\overline{e_{3K/4}}\overline{e_{K/2}^*}+\overline{e_K}\overline{e_{3K/4}^*}),$$

$$\frac{\partial\overline{e_{3K/4}}}{\partial t}+\gamma_{3K/4}\overline{e_{3K/4}}=2i\omega_{3K/4}(\overline{e_K}\overline{e_{K/4}^*}+\overline{e_{K/2}}\overline{e_{K/4}}). \tag{11}$$

The complex conjugate of this system gives the necessary equations for $\overline{e_{K/4}^*}$ and $\overline{e_{3K/4}^*}$. By inserting the ansatz $\overline{e_{K/4}},\overline{e_{3K/4}},\overline{e_{K/4}^*},\overline{e_{3K/4}^*}\propto\exp(\nu_1 t)$ into Eq. (11), we arrive at the following two solutions for the increment:

$$\nu_1^{\pm}=-\left(\frac{\gamma_{K/4}+\gamma_{3K/4}}{2}\mp\omega_{K/4}|\overline{e_{K/2}}|\right)+\left[\left(\frac{\gamma_{K/4}-\gamma_{3K/4}}{2}\mp\omega_{K/4}|\overline{e_{K/2}}|\right)^2+4\omega_{K/4}\omega_{3K/4}(|\overline{e_K}|^2-|\overline{e_{K/2}}|^2)\right]^{1/2}. \tag{12}$$

Changing the sign in front of the square root to minus gives the third and fourth solutions for ν_1. By combining Eq. (12) with the expressions for $\overline{e_K}$ and $\overline{e_{K/2}}$ given by Eq. (10) and setting the real part of the increment, ν_1^+, equal to zero one can obtain the second threshold value of the contrast M_{th}:

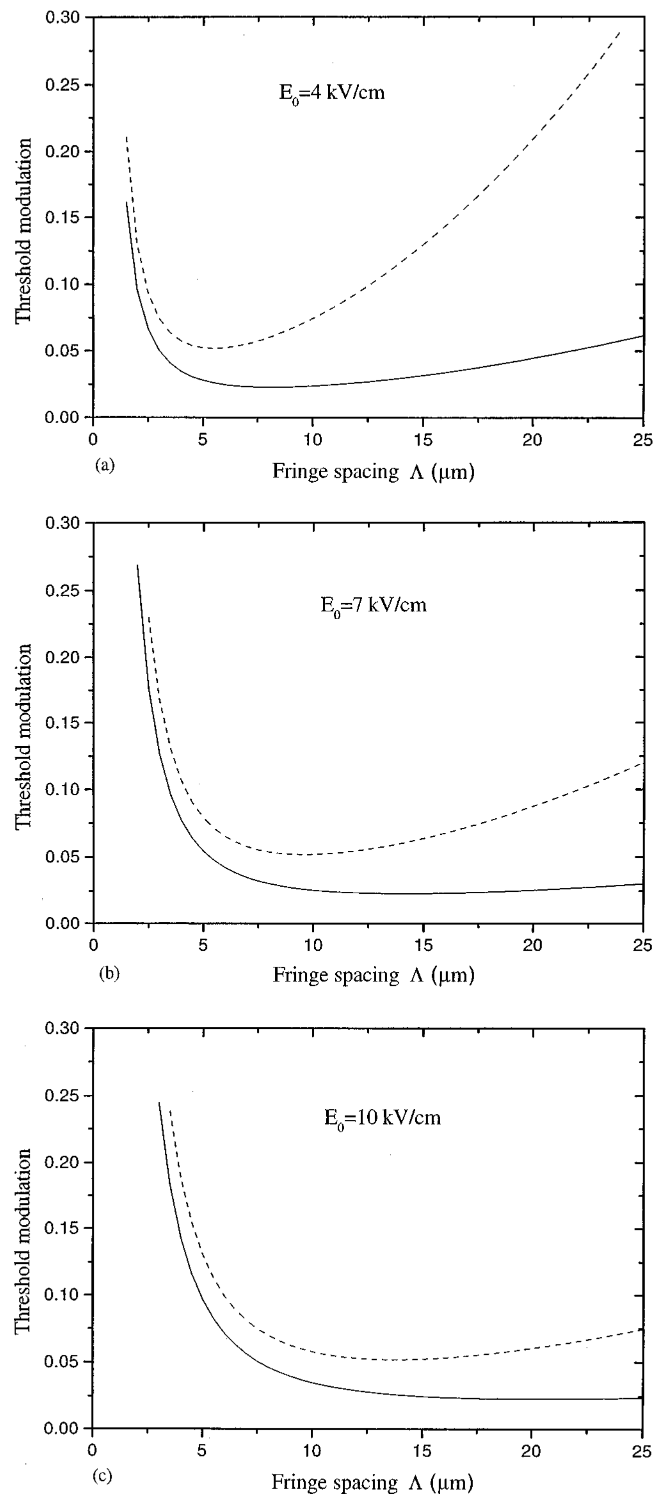

FIG. 1. The threshold values of the contrast m_{th} (solid line) and M_{th} (dashed line) as functions of the fringe spacing Λ for different values of the amplitude E_0.

$$M_{\mathrm{th}}=\frac{1}{Q_K Q_{K/2}}+\frac{(1+\gamma_{3K/4}/\gamma_{K/4})^2}{2Q_{K/4}^2}. \tag{13}$$

Figure 1 shows the threshold values of the contrast m_{th} from Eq. (8) and M_{th} from Eq. (13) as functions of the fringe spacing, $\Lambda=2\pi/K$, for different values of the amplitude of the ac field, E_0. The curves are plotted for parameters relevant to $Bi_{12}SiO_{20}$ crystals [19]. It is seen that threshold of modulation for both the $K/2$ and the $K/4$ subharmonics decreases with increasing ac field amplitude at fixed fringe spacing. Moreover, the difference between these two thresholds is reduced.

Further increasing the contrast can result in new instabilities for next subharmonic gratings, and a cascade of thresholds can be found by repeating the procedure outlined here. The presence of higher spatial harmonic gratings can also influence the amplitude of both the fundamental grating and subharmonic gratings, leading to a growth in dissipation.

In conclusion, we have obtained the steady-state solution for the photorefractive subharmonics with grating vector $K/2$ excited by an ac field. It is also shown that, with increasing contrast, the $K/2$ subharmonic becomes unstable against excitation of the $K/4$ subharmonic grating. Experiments are being planned to verify the theoretical predictions of this paper.

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