Theory of self-frequency detuning of oscillations by wave mixing in photorefractive crystals

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We present a theory of the frequency-detuning properties of various oscillators formed by four-wave mixing in photorefractive crystals. It is shown that the detuning originates from the self-induced grating dynamics in the mixing crystal, governed by phase conditions of the optical paths and several other parameters, such as external and internal electric fields in the mixing crystal.

Interesting experimental findings of self-frequency-detuning effects of phase-conjugate mirrors (PCM's) were reported recently.[1–3] A dye laser in conjunction with a passive (self-pumped) phase-conjugate mirror (PPCM) caused an unexplained self-frequency scanning of the laser.[1,2] Another system of a cavity formed by two PCM's also demonstrated a nondegenerate oscillation.[3]

Recently[4] we presented a first theory that explains the self-frequency-shift properties of the ring PPCM and discussed its meaning as a new type of active interferometry with applications to optical sensors such as gyroscopes. In this Letter we present a general study of the frequency-detuning properties of various oscillators with photorefractive crystals. The results also suggest possible causes for detuning in the PPCM used in self-scanning experiments[1,2] but do not establish the mechanism conclusively; this will be done in further experimental work.

Figures 1(a)–1(d) describe the oscillators discussed in this work. In the past[5,6] these oscillators were analyzed with the assumption of degenerate frequencies. However, the consideration of complex amplitudes with the phases of the beams in the cavity plus the crystal dictates a detuning of the mixing beams. The phase contribution of the induced moving gratings written by these beams is an essential ingredient. It is due to the detuning dependence of the complex coupling constant γ of two mixing beams detuned by δ in a photorefractive crystal[4,5]:

$$\gamma(\delta) = \gamma_0/[1 + i(\tau\delta)], \quad (1)$$

where τ is the time constant of the grating buildup. The additional detuning-dependent phase from $\gamma(\delta)$ is added to the $\pi/2$ spatial phase shift, which exists between the gratings and interference fringes in diffusion-dominated photorefractive crystals.

The mathematical treatment of the nondegenerate four-wave mixing is straightforward in most systems in which the phase-mismatch factor due to the detuning is negligible, i.e., $\delta l/c \ll 1$. l is the effective width of the mixing crystal and c is the speed of light in the crystal. This assumption is valid for photorefractive crystals since $l \sim$ 1–5 mm and $\delta \lesssim 1/\tau \sim 1$–$10^4$ rad/sec. Thus we will use previous calculations for the degenerate case[5] simply by plugging in the complex $\gamma(\delta)$ with the same assumptions of plane waves, transmission gratings, and negligible absorption in the crystal.

Consider the standard nondegenerate phase conjugation in which three of the four-wave mixing beams—the two pumps and the signal—are externally supplied. If the signal's frequency is $\omega + \delta$ compared with ω of the two pumps, the reflected beam is downshifted to $\omega - \delta$. Although the magnitude of $\gamma(\delta)$ decreases for larger δ, the reflectivity may be highest for $\delta \neq 0$. This can be shown, for example, from the expression for the small-signal reflectivity[7]

$$R = \left|\sinh\left[\frac{\gamma(\delta)l}{2}\right]\Big/\cosh\left[\frac{\gamma(\delta)l}{2} + \frac{\ln r}{2}\right]\right|^2, \quad (2)$$

where r is the pump's intensity ratio. Self-oscillation ($R = \infty$) occurs for a nonzero δ and a specific r such that

$$\gamma_0 l\left[\frac{1}{1 + i(\tau\delta)}\right] + \ln r = iq\pi, \quad (3)$$

where q is an odd integer. It might have been thought[8,9] that the preference for high gain is the cause for the frequency shift of oscillators with photorefractive crystals. It will be shown, however, that for the various oscillators discussed here, the frequency detuning is dictated by phase considerations of the cavity and the crystal.

We use the notation and results of Ref. 5, recalculated for the complex amplitudes $A_i(z)$ of the waves rather than their intensities. Thus we define $m_1 = A_1(0)/(A^*_2(0)$, $m_2 = A^*_2(l)/(A_1(l)$, $r_1 = A_3(0)/(A^*_4(0)$, and $r_2 = 1/m_2$. For boundary conditions where $A_3(l) = 0$, valid for all the configurations of Fig. 1 except for Fig. (1d), we obtain[4]

$$m_1 = \frac{T + Q}{m_2[(\Delta + B)T + Q]}, \quad (4)$$

$$r_1 = -\frac{(\Delta + 1)T}{m_2[(\Delta T + Q)]}, \quad (5)$$

where $Q = [\Delta^2 + (\Delta + 1)^2|r_2|^2]^{1/2}$, $T = \tanh[(\gamma l/2)Q]$, $B = (1 + \Delta)|r_2|^2$, and $\Delta = [(I_2 + I_3) - (I_1 + I_4)]/I_0$ is the conserved intensity flux normalized by the total intensity $I_0 = \sum I_i \equiv \sum|A_i|^2$.

The properties of the ring PPCM [Fig. (1b)] can be obtained easily.[4] The ring's complex amplitude

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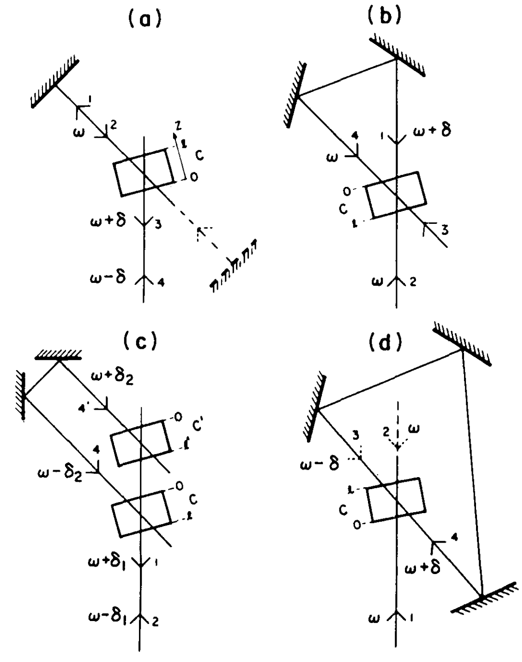

Fig. 1. The oscillators with photorefractive crystals C and C' described in this Letter. (a) Semilinear and linear (dashed lines) PPCM's. (b) Ring PPCM. (c) 2IR PPCM. Usually the two regions C and C' are in one crystal of which the faces are the mirrors. (d) Unidirectional and double-directional (dashed arrows) ring oscillators.

transmissivities for the counterpropagating beams provide the boundary conditions at the $z = 0$ surface of the mixing crystal:

$$m = A_1(0)/A_3(0), \qquad \tilde{m} = A_4(0)/A_2(0). \tag{6}$$

Realizing that $m_1/r_1 = m\tilde{m}^* = Me^{i\vartheta}$, where ϑ is a nonreciprocal phase in the ring and equating this to the ratio obtained from Eqs. (4) and (5) results in

$$\frac{(T+Q)(\Delta T + Q)}{[(\Delta + B)T + Q](\Delta + 1)T} = -Me^{i\vartheta}. \tag{7}$$

Here, $\Delta = (1 - M)/(1 + M)$ is known.[4,5] Equation (7) gives the reflectivity $|r_2|^2$ and the detuning δ with a wide linear region[4] around $\vartheta \sim 0$:

$$(\tau\delta) \approx \alpha\vartheta, \tag{8}$$

where $\alpha = [(M/(M + 1)]\sinh(\gamma_0 l)/(\gamma_0 l)$.

Note that any reciprocal phase in the ring is canceled out. The effect of the detuning δ on ϑ was ignored in this calculation. It adds $\delta L/c$ to ϑ, where L is the cavity's length and c the speed of light in the cavity. For long L, however, ϑ must be renormalized.[4]

Equation (4) provides a solution for a linear PPCM [Fig. (1a)], where $(m_1 m_2) = M'e^{i\vartheta'}$ is known. It gives $\delta(\vartheta')$ and Δ, and Eq. (5) gives the reflectivity of this device, r_1. This is not exactly compatible with the linear PPCM of Fig. 1(a), since the mirrors plus the cavity provide information about A_1/A_2 and not A_1/A^*_2, which is required for m_i. Then ϑ' and δ will not be specified by Eq. (4). However, for the semilinear PPCM with only one mirror the solution is immediate: $m_1 = 0$ in Eq. (4) results in

$$T + Q = 0. \tag{9}$$

Since Δ and Q are real, this implies $\delta = 0$ and a nonshifted oscillation for any ϑ'.

The two-interaction-region PPCM (2IR PPCM) (Ref. 2) of Fig. 1(c) is a combination of a ring PPCM with a double phase conjugator (at the region C') in its feedback loop.[5] The boundary conditions for the double PCM (DPCM) with two vanishing beams at the crystal's surfaces are similar to those for the semilinear PPCM with one mirror, producing the same conditions of Eq. (9) and stationary gratings, i.e., $\delta_1 = \delta_2 = \delta$. This still permits different frequencies for the two couples of the writing beams in the region C' and moving gratings in the first region C. The precise δ will be determined by another property of the DPCM obtained by the conserved constant[5] $c = A_{1'}(z)A_{2'}(z) + A_{3'}(z)A_{4'}(z)$ in the region C'. Since $A_{3'}(l') = A_{1'}(0) = 0$ and $c(0) = c(l')$, it follows that

$$A_{1'}(l')/A_{4'}(0) = A_{3'}(0)/A_{2'}(l'). \tag{10}$$

This means that the complex amplitude transmissivities of the counterpropagating beams through the DPCM (C') are the same, and the ring is reciprocal. Therefore, as for the ring PPCM, $\delta = 0$, all the beams of the 2IR PPCM are degenerate, unless a nonreciprocal phase ϑ exists in the ring. The ϑ dependence of δ is similar to that of the ring PPCM and is given by relation (8). The apparent contradiction with the experimental findings is discussed below.

The unidirectional and double-directional ring oscillators of Fig. 1(d)[6] are different from the previous rings. The existence of a feedback loop of the oscillating beams into themselves [$A_4(0) = mA_4(l)$ and $A_3(l) = \tilde{m}A_3(0)$] results in a dependence on the reciprocal phases of the resonator paths (m and $\tilde{m}$), whereas reciprocal phases were canceled out in the previous rings, which depend on $m\tilde{m}^*$. We do not elaborate on this configuration since an analysis has already been published.[9].

Besides the explicit detuning dependence on the crystal and cavity parameters such as τ, M, and γ_0, the effect of an electric field in the crystal is interesting. An applied dc field adds a phase source in the cavity through its effect on the spatial phase between the gratings and the fringes of the mixing beams in the crystal. This affects the frequency shift of the oscillation.[10]

The electric-field dependence of γ is given by [5,6]

$$\gamma(E_0, \delta) = \gamma_0 \frac{f(E_0)}{1 + i(\tau\delta)}, \tag{11}$$

where

$$f(E_0) = a\frac{E_p(E_0 + iE_d)}{E_0 + i(E_d + E_p)}$$

and $a = (E_d + E_p)/(E_p E_d)$ normalizes $f(E_0)$ such that $\gamma(E_0 = \delta = 0) = \gamma_0$, $E_d = k_B\tilde{T}k/e$, $E_p = ep_d/\epsilon k$, k_B is Boltzmann's constant, $\tilde{T}$ is the temperature, ϵ is the dielectric constant, e is the electron charge, k is the grating's wave number, and p_d is the trap's density. Inserting $\gamma(E_0, \delta)$ into the equations that describe the various oscillators gives the detuning dependence on E_0. (We neglect here the weak τ dependence[10] on E_0.) Applying Eq. (9) for the semilinear PPCM with one mirror will dictate that $\gamma(E_0, \delta)$ be real and

$$(\tau\delta) = -\frac{E_pE_0}{E_0^2 + E_d(E_d + E_p)} \approx -\left(\frac{E_p}{E_d(E_d + E_p)}\right)E_0$$

$$= -\beta E_0 \qquad (12)$$

for $E_0^2 \ll E_d(E_d + E_p)$.

A similar procedure for the ring PPCM with an electric field, using Eq. (7), results in[10]

$$(\tau\delta) \approx \alpha\vartheta - \beta E_0 \qquad (13)$$

in the linear region, where

$$\alpha = [M/(M + 1)] \sinh(\gamma_0 l)/(\gamma_0 l), \qquad \beta = \frac{E_p}{E_d(E_d + E_p)}.$$

For the 2IR PPCM, the detunings in the two regions are determined by Eq. (12) and relation (13), giving

$$\tau_1(\delta_1 + \delta_2) = \alpha\vartheta - \beta(E_0)_1, \qquad \tau_2(\delta_1 - \delta_2) = -\beta(E_0)_2, \qquad (14)$$

where τ_i and $(E_0)_i$ are the time response and electric fields, respectively, in the two regions.

Even in the absence of an applied electric field, an internal field can activate a detuning. The bulk photovoltaic effect, for example, can cause a dc electric field in the crystal and also influences the nonuniform space-charge field. We carried out a detailed calculation to this effect on γ, assuming the photovoltaic current to be of the form of[11–13]

$$\bar{J}_{pv} = vnp\hat{c}, \qquad (15)$$

where n and p are the densities of the mobile electrons or holes and the ionized donors or acceptors, $\hat{c}$ is a unit vector along the crystal's c axis, and v is a constant. Assumptions similar[10] to those for the derivation of expression (12) give

$$\gamma = \gamma_0 f'/[1 + i(\tau\delta)], \qquad (16)$$

where

$$f' = \frac{aE_p(E_0 + E_{pv} + iE_d)}{E_0 + i(E_d + E_p)},$$

such that $\gamma(E_0 = E_{pv} = \delta = 0) = \gamma_0$, $E_{pv} = v(\hat{c}\cdot\bar{k})\, p_d/(e\mu)$, μ is the mobility, and $\bar{k}$ is the wave vector of the grating. The photovoltaic effect will modify Eqs. (12)–(14) such that

$$-\beta E_0 \rightarrow -\beta E_0 - (1/E_d)E_{pv}. \qquad (17)$$

The dc field E_0 in the crystal is determined by the electrical circuitry.

Our analysis shows that an internal electric field alone cannot satisfactorily explain the up-and-down shift of the frequency in the same system. It must be accompanied by some nonreciprocal phase in the ring. Such nonreciprocity may originate from different paths of the counterpropagating beams in the ring not being exactly phase-conjugate waves or may result from some noise or instability. We note that an experimental possibility exists of forming reflection gratings, which may cause an additional reciprocal phase dependence of the detuning, as in the unidirectional and double-directional ring oscillators. The reflection gratings may be particularly important in the compact 2IR PPCM, since a limited coherence length does not wash out these gratings.

In conclusion, we have presented a basic analysis of the frequency-shift behavior of oscillators with photorefractive crystals, which opens the way for resolving the unexplained spontaneous detuning effects and a systematic experimental evaluation of these oscillators.

Note added in proof: Following the submission of this Letter, an experimental study of the detuning properties of various oscillators was carried out by the author and his colleagues.[14]

Results of this work were presented at the annual Israel Physical Society meeting, April 1985.

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