Tunable frequency shift of photorefractive oscillators

Shmuel Sternklar, Shimon Weiss, and Baruch Fischer

Department of Electrical Engineering, Technion-Israel Institute of Technology, Haifa 32000, Israel

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We report on an experimental study of the self-frequency detuning in wave mixing oscillators with the photorefractive barium titanate crystal. We show its dependence on a dc electric field on the crystal, optical phases in the oscillator cavity, and light intensity in the crystal. This resolves many aspects of previously observed and unexplained self-frequency detuning effects with similar oscillators and indicates the existence of an internal electric field in the mixing crystal.

Wave mixing oscillators with photorefractive crystals have been shown to display self-frequency shifts of the oscillating beams. Recent reports of self-frequency scanning[1,2] with a passive phase-conjugate mirror (PPCM) coupled to a dye-laser cavity have led to some speculations concerning the mechanism that causes this nondegenerate oscillation. Self-frequency detuning has also been observed in the double phase-conjugate resonator.[3]

We have developed a theory of self-frequency detuning in these oscillators.[4–6] Here we present experimental results that resolve many aspects of self-frequency detuning, which we show is easily controllable by external means.

Two four-wave mixing configurations were studied: the semilinear PPCM [Fig. 1(a)] and the ring PPCM [Fig. 1(b)]. The ring PPCM is especially interesting, since it has been shown[5] to have similar detuning properties to those of the two-interaction-region (2IR) PPCM used in frequency-scanning experiments. A theoretical study[5] has revealed that the ring and semilinear configurations differ fundamentally in their detuning characteristics because of their different boundary conditions. In the semilinear PPCM, the detuning has a wide region of an approximately linear dependence on dc electric field along the grating wave vector, expressed by [5]

$$\tau\delta = -\beta E_{0\varphi}, \tag{1}$$

where[7] $\beta = E_p/E_d(E_d + E_p)$ and $E_{0\varphi}{}^2 \ll E_d(E_d + E_p)$. Here δ is the frequency detuning as shown in Fig. 1, and τ is the time response of the crystal. $E_p = ep_d/\epsilon_\varphi k_g$ and $E_d = k_B T\, k_g/e$, where p_d is the density of traps in the material, k_B is Boltzmann's constant, T is temperature, and k_g is the grating wave number. ϵ_φ and $E_{0\varphi}$ are the dielectric constant and the effective dc electric field, respectively, along the grating wave vector $\mathbf{k}_g$. In this device, vanishing boundary conditions for two of the beams implies no detuning dependence on the beams' phases. Therefore for $E_{0\varphi} = 0$, $\delta = 0$ also, and the oscillation is degenerate.

For the ring and 2IR PPCM's, frequency detuning can occur even for a zero electric field. Any nonreciprocal optical phase between the counterpropagating beams in the ring is accompanied by frequency detuning of oscillating beams 3 and 1 with respect to beams 2 and 4 by an amount δ. In the nearly linear detuning region [5,6]

$$\tau\delta = \alpha\vartheta - \beta E_{0\varphi}, \tag{2}$$

where $\alpha = (M/M + 1)$ $(\sinh \gamma_0 l/\gamma_0 l)$. M is the ring's intensity transmittance, γ_0 is the coupling constant for the wave mixing with zero detuning and electric field, and l is the effective crystal width. ϑ is the nonreciprocal optical phase in the ring ($-\pi \leq \vartheta \leq \pi$). The 2IR PPCM is essentially a ring PPCM with a double phase conjugator[7] in the feedback loop, which does not of itself contribute a nonreciprocal phase to the ring.[5] Therefore it will be governed by a similar detuning dependence on ϑ and $E_{0\varphi}$.

Since the semilinear PPCM is free of any optical phase dependence in its detuning property, it was a preferred starting point for our experiments. A bare $BaTiO_3$ crystal was held with pressure between two thin metal electrode plates and connected to a dc volt-

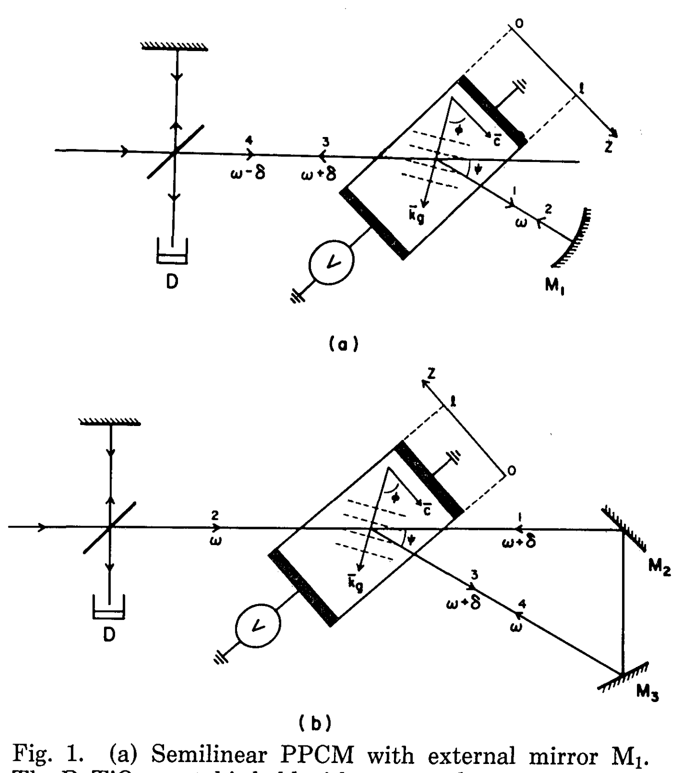

Fig. 1. (a) Semilinear PPCM with external mirror M_1. The $BaTiO_3$ crystal is held with pressure between two electrodes and connected to a dc voltage supply. A portion of output beam 3 is combined with the reference beam on the detector D. (b) Ring PPCM.

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age-supply source. We set up a semilinear PPCM, as shown in Fig. 1(a). Light from the 488-nm line of an argon-ion laser without an étalon was focused with a beam spot diameter of approximately 1 mm in the crystal. The angle ψ in the crystal between the pump beam 4 and oscillation beams 1 and 2 was about 7°, and $\varphi \approx 74°$ between $\mathbf{k}_g$ and the c axis of the crystal. A portion of phase-conjugate beam 3 and a reference beam were combined on detector D. The applied voltage formed an electric field in the crystal in the range $|E_A| \leq 1.3$ kV/cm. Note that $E_{A\varphi} = E_A \sin\varphi$ is the component of E_A along the grating wave vector. There exists the possibility, however, that the actual voltage drop across the interaction region in the crystal was somewhat different from this value because of a nonuniform voltage in the crystal. Photoconductivity in the interaction region, in particular, would contribute to this nonuniformity. A strong background irradiation on the crystal, however, did not change our results. In all our experiments, the applied electric field was perpendicular to the crystal's c axis. This has important implications for our experimental results and is discussed below.

A sample of typical plots of the measured detuning dependence on an electric field is shown in Fig. 2(a) for slightly varying crystal orientation or input power density. The intensity of the oscillation did not change appreciably for different E_A; thus I_0 was approximately constant. For the plot marked with circles, the input power density was 15 mW/mm^2. For this run, we noticed instabilities in the detuning for certain values of E_A, especially at ±0.6 kV/cm. For the other graphs, the power density was 25 mW/mm^2. No instabilities were noticed in these runs. Invariably, we observed small positive detuning for a zero applied field, which decreased to zero in the region $E_A = E_i \sim 0.2$ kV/cm. These graphs indicate that an internal electric field $\mathbf{E}_{\text{IN}}$ exists within the crystal and has a component $E_{\text{IN}\varphi} = -E_{i\varphi}$ along $\mathbf{k}_g$, where $E_{i\varphi} = E_i \sin\varphi \simeq 0.19$ kV/cm. This field may be due to the bulk photovoltaic effect that causes an electric field to form along the c axis (or the z coordinate). We can estimate this internal photovoltaic field $\mathbf{E}_{\text{IN}} = \hat{z}E_{i\varphi}/\cos\varphi = \hat{z}E_i \tan\varphi \simeq 0.7\hat{z}$ kV/cm. The actual electric field may be different, as explained earlier. The lack of data points around zero detuning ($E_A \simeq E_i$) reflects the small $\delta(E_A)$ sensitivity in this region and a poorly defined point of absolute zero detuning in some graphs. One reason for this may be the presence of a set of grating wave vectors, for a set of oscillation angles between the crystal and the external spherical mirror. The components of the internal field $E_{\text{IN}}\hat{z}$ along these different grating wave vectors cannot be canceled out simultaneously by the externally applied field $E_A\hat{x}$. In this regard, an applied field parallel to the c axis and the internal field, so that $\mathbf{E}_A = E_A\hat{z}$, should be more efficient because it would cancel out the vectorial internal field.

Turning to the ring PPCM, we measured $\delta(E_A)$ for this device with the setup shown in Fig. 1(b). The input power density was 25 mW/mm^2, $\varphi \approx 74°$, and $\psi \approx 7°$. Typical plots are shown in Fig. 2(b) for slightly varying crystal or external ring orientations. The difference between these plots and those for the semilinear PPCM is striking: here the $\delta(E_A)$ dependence shifts vertically along the detuning axis, with both positive and negative detuning observed for a zero applied field. This is easily explainable through the $\alpha\vartheta$ term in detuning expression (2) for the ring PPCM. A nonreciprocal phase ϑ may be due to incomplete

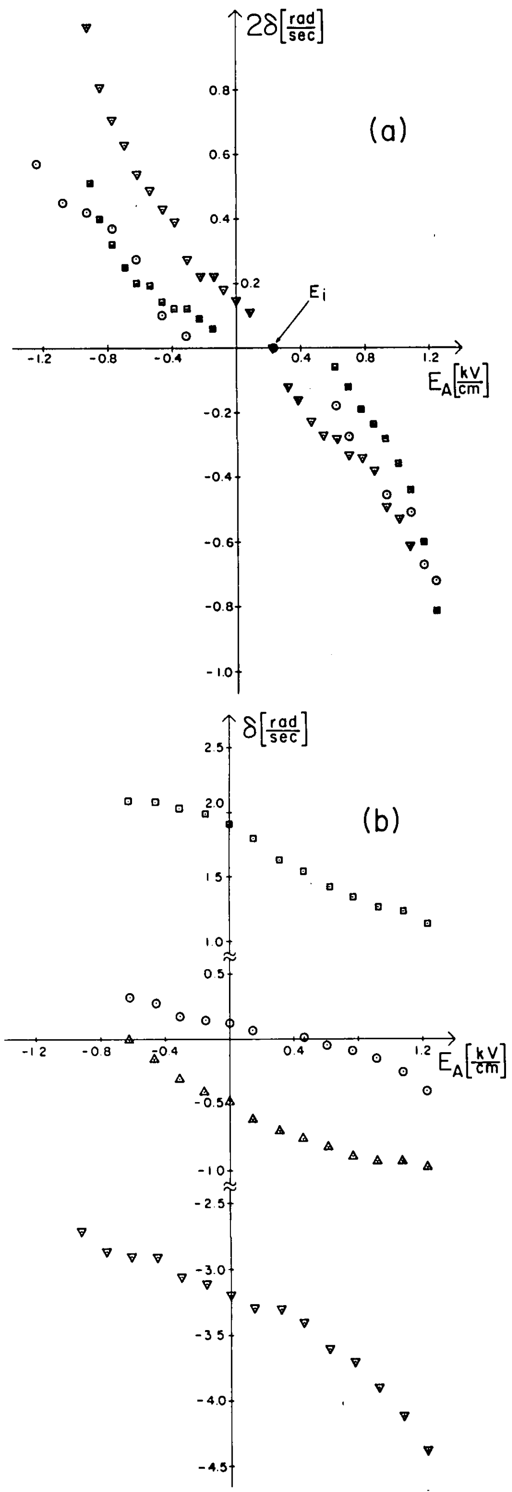

Fig. 2. (a) Experimental data of detuning versus applied electric field for the semilinear PPCM. (b) Experimental data of detuning versus applied electric field for the ring PPCM.

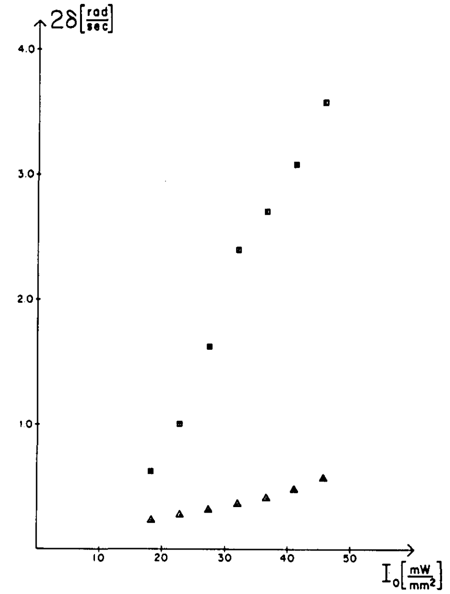

Fig. 3. Experimental data of detuning versus optical power density in the crystal for the semilinear PPCM with zero applied field. The data with the triangles were taken with the crystal used in the other experiments in this paper, and the squares with another $BaTiO_3$ crystal.

phase conjugation, which would result in different complex amplitude transmissivities for the counterpropagating beams in the ring. In general, we observed that optimization of the oscillation through careful alignment resulted in a smaller detuning. This suggests a possible method for determining the degree of phase conjugation with this phase-conjugate mirror. Another factor may be the presence of reflection gratings in the crystal. These gratings might give rise to a dependence also on reciprocal phases and the rings' optical-path-dependent phase. These gratings were absent in our experiments since the beams' coherence length was smaller than the ring path. In the 2IR PPCM, however, the small internal ring is conducive to the formation of reflection gratings.

From Eq. (1) or (2) and recalling that the time constant is intensity (I_0) dependent as[7] $\tau \approx A/I_0$, we may write

$$\delta(E_{0\varphi}, I_0) \approx -(\beta/A)E_{0\varphi}I_0, \quad (3)$$

where $E_{0\varphi} = E_{A\varphi} + E_{\mathrm{IN}\varphi}$. This permits control of the detuning by using I_0. We checked this dependence on the power density with two crystals and obtained the plots shown in Fig. 3. For this experiment, we used the semilinear PPCM configuration with zero applied field. The observed detuning dependence on I_0 was nearly linear. The deviation in the slopes for different crystals is due to different values for $E_{\mathrm{IN}\varphi}$ or β, which are strongly dependent on the particular wave mixing configuration within the crystal. $E_{\mathrm{IN}\varphi}$, E_p, E_d, and τ are ψ and φ dependent through k_g and ϵ_φ.

We fitted all plots in Figs. 2 and 3 to straight lines and normalized the slopes δ versus E_A in Fig. 2 to $I_0 = 25$ mW/mm^2. This resulted in an average slope of $\beta I_0/A = 0.46$ (rad/sec)/(kV/cm) for both the semilinear and ring PPCM's, as expected. In Fig. 3, the slope δ versus I_0 for the same crystal is $\beta E_{\mathrm{IN}\varphi}/A = 0.005$ (rad/sec)/mW/mm^2. Assuming that $E_p \simeq 5$ kV/cm and $E_d \simeq 1$ kV/cm in our experiments gives $\beta \simeq 0.7$ cm/kV for $k_g = 2\pi/(1.5\ \mu\mathrm{m})$, a dielectric constant along $\mathbf{k}_g$ of $\epsilon_\varphi \simeq \epsilon_x \sin^2\varphi \approx 4000$ ($\epsilon_x \gg \epsilon_z$), and $p_d \sim 5 \times 10^{16}/\mathrm{cm}^3$. This results in $A = 37$ sec mW/mm^2 for the $\delta(E_A)$ experiments and $A = 26$ sec mW/mm^2 for the $\delta(I_0)$ experiment (where we assume that $E_{\mathrm{IN}\varphi} = -0.19$ kV/cm). This implies that $\tau \sim 1$–1.5 sec in the $\delta(E_A)$ experiments with $I_0 = 25$ mW/mm^2.

The hypothesis that the internal electric field has a bulk photovoltaic origin is supported by observed photocurrents in $BaTiO_3$.[8,9] An analysis[5] shows that it affects the detuning by modulating the nonuniform space charge field and by a uniform dc field that is formed. In an open circuit along the c axis, as is the case in our experiment, the effect of the dc field almost cancels the overall detuning. Thus either some assumptions used in the derivation of the photovoltaic effect on the coupling constant are incorrect or the crystal is effectively partially short circuited internally. The latter possibility has support from other works.[10]

We have experimentally resolved many aspects of self-frequency detuning. For the ring or 2IR PPCM, the combination of a nonreciprocal phase in the feedback ring and an internal field are the sources of the self-detuning.[11] The amount of detuning and its sign (positive or negative), and so the scanning rate and direction when coupling to a dye-laser cavity, are dependent on the crystal orientation. This enters through the detuning dependence on γ_0, τ, the optical phase ϑ, β, and the component of the internal field along $\mathbf{k}_g$. The semilinear PPCM, however, is not sensitive to optical phase in the oscillation path for its detuning and should therefore prove to be a more useful detuning device in certain applications. The self-frequency detuning is easily controllable through an applied dc electric field or the light intensity in the crystal. This will permit external biasing of interferometers such as the ring passive phase-conjugate gyroscope[4] and optical frequency and intensity control and modulation.

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