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A new instability of counter-propagating waves in $BaTiO_3$?

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ABSTRACT The self-development of a strong reflection grating in a $BaTiO_3$ sample illuminated with two mutually incoherent counter-propagating light waves is reported. The two transmitted waves diffracted from this grating become partially mutually coherent.

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1 Introduction

An inherent feature of barium titanate crystals is a strong photorefractive nonlinearity based on a diffusion-driven charge redistribution. When the crystal is illuminated with two coherent light beams, a refractive-index grating is formed which is $\pi/2$-shifted with respect to the fringe pattern. This type of nonlocal nonlinear response ensures a significant gain for a properly polarized and properly oriented weak probe beam at the expense of the intensity of a strong coherent pump beam (see, e.g., [1]). The beam coupling gain was successfully used to design a vast variety of photorefractive coherent oscillators. Exposed to one or several pump beams, these devices generate new beams with a direction of propagation imposed by the cavity mirror(s) or by phase matching conditions (see, e.g., [2, 3]). Apart from a scientific interest these oscillators attract attention of researchers because they can serve as phase conjugate mirrors and they can make coherent beam clean-up possible [2].

The appearance of oscillation, which is due to nearly (or strictly) degenerate four-wave mixing, is a consequence of instability in terms of nonlinear dynamics. Above a certain critical value of the coupling strength the system becomes unstable. With no external seed any fluctuation in the direction of the oscillation wave grows nonlinearly and reaches a value that may be comparable to the pump intensity. Instabilities different from that at the oscillation threshold were revealed in photorefractive oscillators, too, like, e.g., instability of a single-frequency mode of operation accompanied with a bifurcation into a two-frequency oscillation [4, 5]. When investigating different instabilities in a semi-linear coherent oscillator with two mutually incoherent counter-propagating waves [1–4], we discovered that a high efficiency photorefractive grating may appear that couples two mutually incoherent pump waves. The abrupt and strongly nonlinear development of this reflection grating suggests the attribution of the phenomenon to a new type of instability with a critical behavior.

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2 Basic experiments

The effect described below was detected when studying a semi-linear photorefractive oscillator with two counter-propagating pump waves that involve a transmission grating recording (Fig. 1). In this geometry a photorefractive sample ($BaTiO_3$:Co) is illuminated by two counter-propagating waves, 1 and 2, formed from the same Ar^+-laser beam (TEM_{00}, output power about 300 mW at $\lambda = 514$ nm). The light waves 4 and 1 that record a transmission grating in the sample, are made mutually coherent by adjusting the path of the wave 1 to be longer than that of wave 2 exactly to the doubled distance between the sample and the conven-

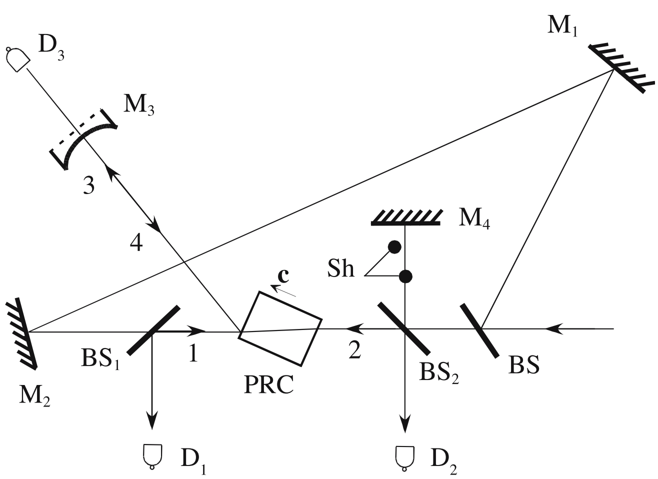

FIGURE 1 Experimental arrangement used to observe the development of the reflection gratings. PRC is the photorefractive crystal pumped as a phase conjugate mirror by the pump beams 1 and 2, M_1–M_4 are mirrors, BS, BS_1, BS_2 are beam splitters, D_1–D_3 are photodetectors, Sh is a shutter

tional mirror M_3. By this way the light of wave 2 scattered from optical imperfections of the sample in the direction of wave 3 returns back to the sample as wave 4, which is coherent to wave 1.

In our experiments the path difference of pump waves 1 and 2 is 72 cm, i.e., much larger than the coherence length of Ar^+-laser with no etalon inside the cavity. This path difference of 72 cm was chosen also to be different from multiple round trips in the Ar^+-laser cavity to avoid accidental coherence at larger path differences.

The exactly counter-propagating pump waves enter the 6.1 mm-thick sample through opposite z-faces at angles $\geq 32°$ (and $\geq 212°$) up to $\approx 40°$ (and $\approx 220°$). The cavity axis is tilted to the z-direction by 24°. All angles are given in the air. The polar ferroelectric axis of the photorefractive crystal points into the direction of the conventional mirror of the semi-linear oscillator. This geometry ensures a high-beam coupling gain for the forward propagating oscillation wave 3 if all interacting waves are extraordinarily polarized [6]. According to the calculations reported in [7], it should ensure also a high gain for an oscillation wave propagating in roughly opposite direction if the mirror M_3 is repositioned to the other side of the sample. Such oscillation waves are coupled to the pump waves by reflection gratings.

Two uncoated glass plates BS_1 and BS_2 are put at approximately 45° to the direction of the pump waves. The beams reflected from these plates are used to monitor the intensities of the transmitted pump waves. The plate BS_2, together with the sample and the auxiliary flat mirror M_4 placed at a distance equal to that from BS_2 to the sample, forms a Michelson interferometer. The onset of high contrast fringes in this interferometer indicates the appearance of a reflection grating in the sample.

The effect that we describe manifests itself when all experimental parameters are optimized to get the largest possible coupling strength. An indirect proof of a high coupling strength $\gamma_0\ell$ is the observation of mirrorless coherent oscillation nearly in the direction of beam 3 when the conventional mirror M_3 is removed. According to our estimates [5], the threshold coupling strength which is necessary to get mirrorless oscillation cannot be smaller than $\gamma_0\ell = 2\pi$.

The first unusual observation consisted in the sudden appearance of a strong reflection of pump 2 from the sample, which occurs simultaneously with the development of oblique mirrorless coherent oscillation. Figure 2 shows light scattering tracks inside the sample illuminated successively with (a) only one pump wave (wave 2), with (b) only one pump wave (wave 1) and with (c) the two pump waves simultaneously. Note in frame c the track of a beam reflected up from the right face, which is absent in the frames a and b. It should be underlined that this beam is not visible at the very beginning of the sample exposure to the two pump waves; it appears abruptly after a certain time delay. The observation of this anomalous strong reflection track suggests a rather quick "bleaching" of the sample for the pump wave 1 that is related somehow to the onset of mirrorless oscillation.

In the next experiment the temporal evolutions of the two transmitted pump waves are measured with the detectors D_1 and D_2. During this recording the mirror M_4 is blocked by a shutter Sh and the integral intensity of mirrorless oscillation is collected with the detector D_3. Typical kinetic curves are plotted in Fig. 3. An obvious correlation can be seen in these dependences. After an exposure time of approximately 0.7 s, the intensity of mirrorless oscillation starts to grow and the intensities of the transmitted pump waves change dramatically: the initial intensity of pump 2 decreases by more than a factor of two, while the intensity of pump 1 becomes nearly 20 times larger than its initial value. As there is no end mirror M_3 in this experiment, such a strong increase of the intensity of the transmitted pump 1 is possible only if the two counter-propagating pump waves become coupled by a reflection grating.

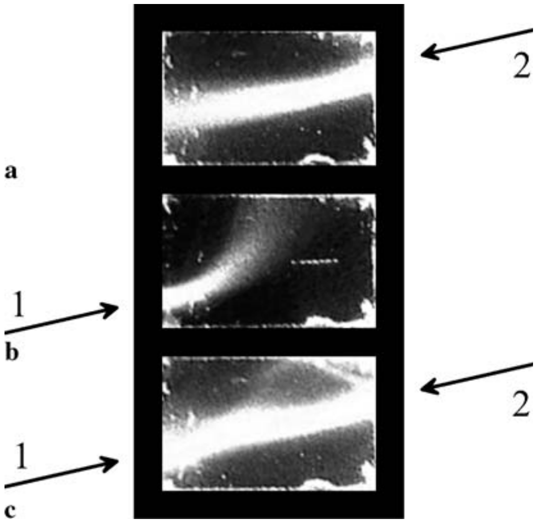

FIGURE 2 Snapshots of the beam paths inside the photorefractive crystal when (**a**) pump 2 is alone incident on the crystal, (**b**) pump 1 is alone incident on the crystal, (**c**) the two pump beams propagate and interact in the crystal

Note that the temporal behaviors presented in Fig. 3 are typical if all index gratings in the photorefractive sample, remaining from previous exposures, are erased with incoherent light before recording. If the erasure is done with one of the two (coherent) pump waves, the growth of mirrorless oscillation intensity is delayed in time and becomes much steeper. The correlation with the intensity changes of the transmitted pump waves becomes even more pronounced in the last case.

To prove the hypothesis that a reflection grating appears in the sample, the dynamics of the fringe visibility is studied. The shutter Sh unblocks the mirror M_4, while a screen is put instead of detector D_2 and a CCD camera monitors the intensity distribution on the screen. Two beams are coming to the screen, one from the pump wave 2 that is reflected by BS_2 and then by the mirror M_4, and the other reflected by BS_2 from pump 1. At the beginning of exposure these beams are incoherent and the intensity distribution on the screen in bell-shaped. A central area of the light spot on the screen is shown in Fig. 4a. This uniform distribution is transformed into a regular fringe pattern when strong variations in the transmitted pump intensities occur. The fringe contrast is growing rapidly, reaching its maximum value approximately after 2 s of exposure time (Fig. 4b). This confirms that a reflection grating self-develops inside the sample, which diffracts pump wave 2 exactly in the opposite direction. This grating now acts as a second mirror of the Michelson

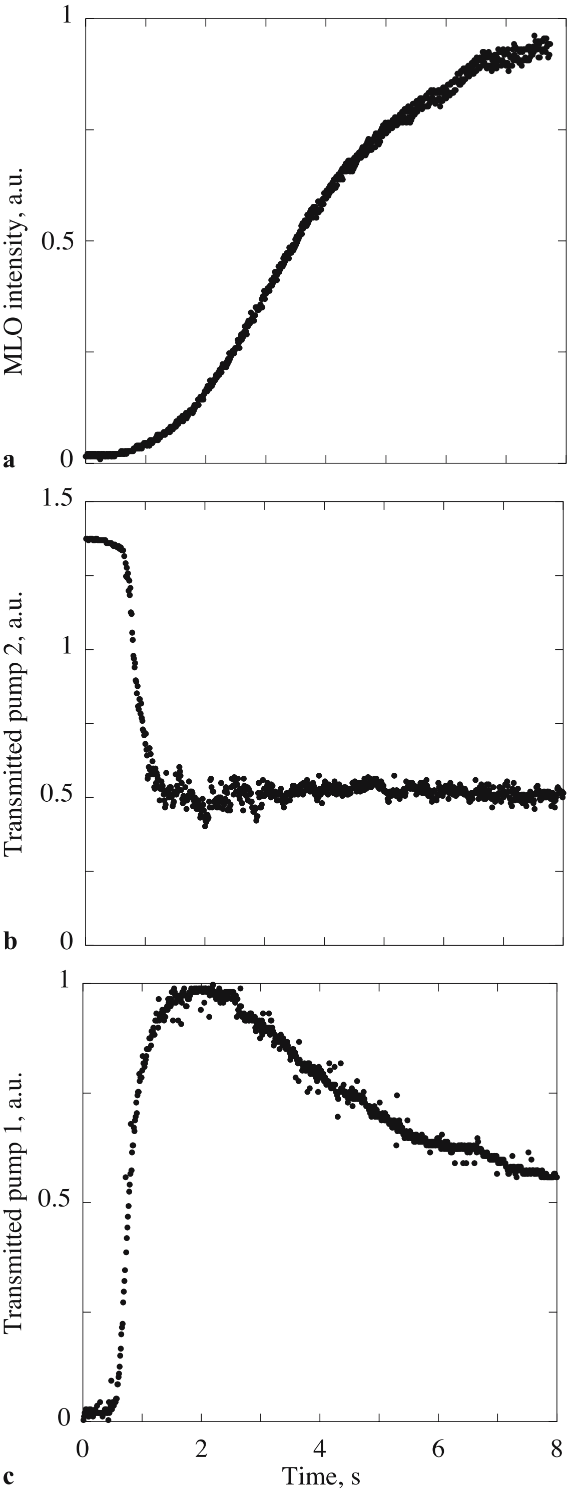

FIGURE 3 Temporal recording of the intensities for (**a**) mirrorless oscillation, (**b**) the transmitted pump 2, (**c**) the transmitted pump 1

interferometer formed by BS_2 and mirror M_4. As the distance from BS_2 to M_4 is nearly the same as the distance from BS_2 to the $BaTiO_3$ sample, the fringes appear on the screen and their spacing can be controlled by a slight tilt of the mirror M_4. This means also that the two pump waves leaving the $BaTiO_3$ sample acquire a certain degree of mutual coherence.

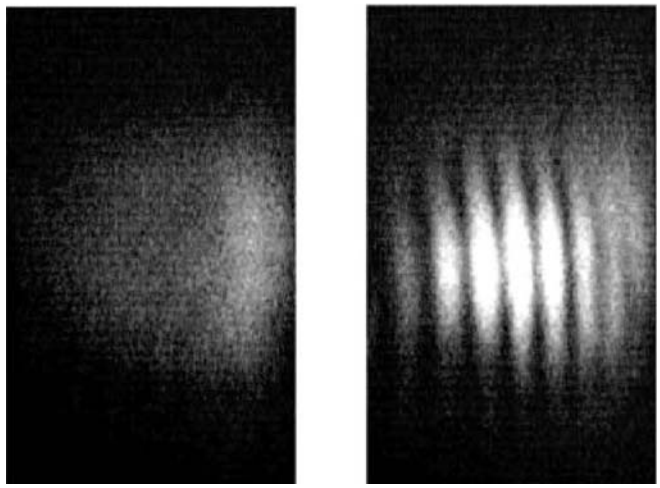

FIGURE 4 Central area of a light spot on a screen replacing detector D_2: (**a**) at the beginning of exposure, (**b**) after 2 s of exposure time

3 Discussion and verifying experiments

Four processes have been considered in the past that may result in the development of the reflection grating and in the onset of partial coherence.

1. In the geometry of the coherent oscillator we deal with a part of the Ar^+-laser output beam reflected back into the laser cavity. This might modify the losses of the longitudinal modes of the Ar^+-laser and impose an additional spectral selectivity. As a consequence, the Ar^+-laser may switch to a single frequency oscillation with a coherence length much higher than the path difference of the two counter-propagating pump waves of the photorefractive coherent oscillator (see, e.g., [8]).
2. In the sample with all faces polished oscillation with total internal reflections from polished faces (Feinberg's cat-conjugator [9]) can occur. It is clear, taking into account the small dimensions of the sample, that a phase conjugate wave should be mutually coherent to the incident pump and therefore may record a reflection grating in the sample.
3. If initially the two recording beams have a small nonzero degree of mutual coherence, it might grow during grating recording: The photons of one beam diffracted from the grating increase the coherent part in the other recording beam so that the mutual coherence of the two transmitted beams increases [10–12].
4. Mirrorless oscillation may appear in samples with a high coupling strength [5], which, as we will show, may be spatially degenerate, i.e., with new beams appearing, that are collinear to the pump beams.

To check whether the first of the above reasons may play a role in our experiments, we put a Faraday isolator at the output of our Ar^+-laser to prevent possible unwanted feedback from the photorefractive coherent oscillator to the Ar^+-laser. The described development of the reflection grating did not disappear. This allows one to conclude that the observed increase of the mutual coherence is an inherent feature of the considered oscillator and is not an artifact related to the pump laser.

To avoid the formation of a total internal reflection loop (reason 2), the sample is aligned in a way that the amplified light propagates only between the pump wave 2 and the crystal c-axis. Then the scattered light that points into the direction of the corner between the two faces of the crystal is depleted. Therefore we do not observe an oscillation that builds up from a total internal reflection loop with neither one of the two pump waves. The probability is rather small that the threshold of such an oscillation with two waves may become smaller than that for each individual pump wave.

In spite of the fact that the third reason from the list cannot be completely ignored the particular temporal dynamics of the onset of the reflection grating can be hardly explained within this model. The abrupt step-like changes in the transmitted intensities of the pump waves (Fig. 3) resemble much various absolute instabilities in photorefractive crystals, i.e., processes with a well defined threshold. The fact that the appearance of reflection gratings is observed only at large coupling strengths, supports the existence of a threshold value of the coupling strength $\gamma_0\ell$ (here ℓ is the interaction length inside the sample and γ_0 is the crystal coupling constant, see, e.g., [1]). This is an argument in favor of the last reason mentioned above.

The direct measurement of the coupling strength threshold value which is, as we expect, higher than $\gamma\ell_{\rm th} = 2\pi$ is not a realistic task: A weak probe beam should be amplified more than 10^5 times with such a coupling strength and other nonlinear effects like, e.g., beam fanning, will reduce the measured gain. More reliable are relative measurements that can say how far above the threshold the system is. With a $\lambda/2$ phase retarder (not shown in Fig. 1) placed in front of the beam splitter BS we can decrease the coupling strength by tilting the pump waves polarization to a certain angle α as

$$\gamma\ell = \gamma_0\ell\cos^2 2\alpha\,.$$

By increasing α the coupling strength $\gamma\ell$ can be decreased down to the threshold value $\gamma\ell_{\rm th}$ that corresponds to the disappearance of the instability.

We performed such measurements at the pump ratio $r = (I_2/I_1) \approx 6.8$ and a sample tilt angle in air of 40°. We found that reflection gratings appear spontaneously only within the range $|\alpha| \le 23°$, i.e., $\gamma\ell_{\rm th} = 0.84\gamma_0\ell$. Keeping the same pump ratio, similar measurements were done for the oscillation in the external cavity (mirror M_3 unblocked). The range of values for α where the oscillation in the external cavity exists appeared to be larger, $|\alpha| \le 47°$ thus indicating a smaller threshold value for the cavity oscillation: $\gamma\ell_{\rm th} = 0.47\gamma_0\ell$. It was also found that there exists a pump ratio threshold $r_{\rm th}$ for the spontaneous development of reflection gratings. For the particular conditions of the described experiment, it is equal to $r_{\rm th} \approx 8.5$. These experiments confirm the existence of a threshold for the discovered process and show that the coupling strength threshold is larger than that for the cavity oscillation, in full agreement with our expectations.

The considered hypothetic instability differs from the known ones by the degeneracy in space: It assumes that in the field of the two counter-propagating waves 1 and 2, there are two more waves 1′ and 2′ that appear with the same wavevectors and each of them is coherent to its relative counter-propagating pump wave (wave 1′ is coherent with wave 2 and wave 2′ is coherent with wave 1). It is known that backward-wave four-wave mixing allows for mirrorless oscillation, i.e., for an absolute instability, both with transmission gratings [5] and/or with reflection gratings [13]. The mirrorless oscillation with the transmission gratings cannot be spatially degenerate: the spatial frequency of transmission gratings tends to zero in this case and the diffusion field that governs the charge redistribution becomes zero, too. At the same time for reflection gratings, the grating spacing (and diffusion field) depends very slightly on the angle between the two waves that enter the sample from the opposite faces of the crystal and the diffusion field is largest for an exact counter-propagation of the incident waves. An obvious advantage of a strict spatial degeneracy is also the best possible overlap of the beams with limited transverse dimensions. We believe that this or a similar four-wave mixing process is responsible for the appearance of the reflection gratings in our experiment.

The threshold coupling strength $(\gamma_0\ell)^{\rm ml}_{\rm th}$ for the development of mirrorless oscillation (superscript ml) was calculated in [5]:

$$(\gamma_0\ell)^{\rm ml}_{\rm th} = \ln r\left[1+\left(\frac{\pi}{\ln r}\right)^2\right], \tag{1}$$

where the coupling strength is introduced as

$$\gamma_0 = \frac{\pi}{n}\frac{k_{\rm B}T}{e}\frac{r_{\rm eff}}{\lambda}\frac{1}{1+(\ell_{\rm s}/\Lambda)^2}, \tag{2}$$

with the refractive index n, the Boltzmann constant $k_{\rm B}$, the electron charge e, the fringe spacing Λ and the Debye screening length $\ell_{\rm s}$

$$\ell_{\rm s}^2 = \frac{k_{\rm B}T\varepsilon\varepsilon_0}{e^2 N_{\rm eff}}. \tag{3}$$

Here $\varepsilon\varepsilon_0$ stands for the dielectric constant $N_{\rm eff}$ and for the effective trap density.

It follows from (1) that the smallest threshold $(\gamma_0\ell)^{\rm ml}_{\rm th} = 2\pi$ is reached for a pump ratio $r = \exp(\pi) \approx 23$. To calculate the coupling constant one should know the angular dependences of the effective electrooptic constant and of the dielectric constant (to have an estimate for the Debye screening length). The expressions for the effective electrooptic constant and for the dielectric constant are as follows

$$r_{\rm eff} = \left(-n_0^4 r_{13}\cos^2\theta + 2n_e^2 n_0^2 r_{42}\sin^2\theta - n_e^4 r_{33}\sin^2\theta\right)\cos^2\theta,$$
$$\varepsilon = \varepsilon_{11}\sin^2\theta + \varepsilon_{33}\cos^2\theta\,, \tag{4}$$

where r_{13}, r_{33} and r_{42} are the components of the electrooptic tensor (contracted indices) while ε_{11} and ε_{33} are the components of the permittivity tensor, θ is the tilt angle of pump waves with respect to the sample's c-axis (inside the sample).

With reasonable assumptions about the effective trap density $N_{\rm eff} \approx 2\times10^{17}\,{\rm cm}^{-3}$ and with handbook values for r_{13}, r_{33}, r_{42}, ε_{11} and ε_{33} we can get, following Honda's evaluation [14], $\gamma_0\ell$ larger than 2π for a tilt angle θ of the pump waves inside the sample starting from 10° (26° in air). In this estimate a linear absorption of the crystal of about $7\,{\rm cm}^{-1}$ is taken into account. In the experiment we observe the described instability at angles larger than 12° (32° in air) which is not surprising taking into account a possible incomplete poling of our sample, that reduces r_{eff}.

As it was already mentioned the appearance of the reflection grating is synchronized in time with the development of mirrorless oscillation with oblique beams (compare time traces b, c with a in Fig. 3). We do not know at present how these two processes are interrelated. Is the appearance of oblique mirrorless oscillation a prerequisite for recording of the reflection grating or vice versa? Both hypotheses have some pro and contra. For example, in line with the last explanation, it is possible to argue that reflection gratings change the pump ratio in a way to bring it closer to the value $r = \exp(\pi) \approx 23$ that ensures the minimum threshold for mirrorless oscillation [5]. On the other hand, the depletion of pump wave 2 which is due to an onset of mirrorless oscillation increases the poor contrast (if there is any) of the two counter-propagating pump waves with a very low initial degree of mutual coherence.

Whatever is the origin of the described development of the reflection gratings it has an important consequence in bringing a partial coherence to the two initially incoherent waves. The advantage of this type of wave coupler as compared with, e.g., a semitransparent mirror, is its self-adaptive nature: with two counter-propagating pump waves there is no need to adjust the reflection grating, if it appears it sends the diffracted beam exactly in the direction of the second beam.

4 Conclusions

To conclude, we described a spontaneous development of reflection gratings that couple two mutually incoherent counter-propagating waves in a photorefractive Co-doped $BaTiO_3$ crystal. We attribute the observed phenomenon to a new kind of absolute instability in four-wave mixing of spatially degenerate backward waves. It features a rather sharp threshold and a strongly nonlinear temporal behavior typical for all instabilities. At the same time it is frequency degenerate and self develops in a pump ratio range that disagrees with predictions of a simple theory [5].

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