Second-order optical phase transition in a semilinear photorefractive oscillator with two counterpropagating pump waves

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Soft-mode onset of coherent oscillation is revealed in a semilinear cavity with two counterpropagating pump waves. From the dynamics of the oscillation intensity and the dynamics of the grating decay with the feedback applied, critical behavior is detected: Both the characteristic time of oscillation onset and grating decay time go to infinity exactly at the threshold coupling strength. A conclusion is made about the similarity of this type of oscillator to the second-order phase transition. © 2002 Optical Society of America

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1. INTRODUCTION

Photorefractive crystals may generate new optical beams with the frequency of the pump wave if appropriate optical feedback is ensured by external mirrors or by a special selection of pump-beam orientations.[1–3] Below the threshold of oscillation, the sample illuminated with the pump beam exhibits light-induced scattering in a wide solid angle (beam fanning). This scattering is due to diffraction of the pump wave from the arbitrary oriented low-amplitude noisy photorefractive gratings that self-develop in the sample. Above the threshold, a rapid growth of the amplitude occurs for one of these gratings, which couples the pump wave to the oscillation wave. The oscillation wave has a small divergence, and its intensity may be comparable to that of the pump wave. In this way, the system changes qualitatively: A new, highly ordered state emerges from a completely disordered state. In other words, the onset of optical oscillation in photorefractive coherent oscillators is similar to the structural phase transitions in solid-state physics. This similarity has been mentioned in several publications.[4–6] In some other publications the typical features of the phase transitions were revealed in coherent oscillation dynamics, without mentioning this similarity. For example, critical slowing of fluctuations was reported when the threshold was approached from below.[7,8] Changes in correlation length of the generated fields in the threshold vicinity have been a subject of interest.[9–11]

It should be noted that similarity of the oscillation threshold in lasers to phase transition is known from the classical works of Haken.[12] For free-running lasers the gain shows typical characteristics of second-order phase transition.[13,14] As distinct from conventional lasers, the number of photorefractive coherent oscillators with different cavity configurations and considerably different properties is quite larger. Some of the oscillators exhibit the features of the second-order phase transition,[5] and the others show those of the first-order phase transition.[6] It is not clear to which extent one can extend conclusions formulated for any particular oscillator to all other possible oscillator geometries. Until now, the detailed studies have been performed for double phase-conjugate mirrors,[5] for generation of hexagonal patterns,[6] and for ring-loop coherent oscillators.[15]

In this paper we study the threshold behavior of the semilinear coherent oscillator with two counterpropagating pump waves. The oscillator under discussion was suggested by A. Yariv and D. Pepper[16]; it was then realized experimentally by R. Hellwarth and J. Feinberg, to our knowledge, as the very first photorefractive oscillator.[17] The practical interest of this configuration is related to its possible applications in lasers with the capability of intracavity distortion correction.[1,3,18] The dynamics of multimode oscillation well above the threshold was studied in this oscillator earlier to reveal the transition to temporal chaos.[8]

At the beginning, we present the results of calculation of the steady-state output characteristics for this oscillator within the approximation of nonlinear mixing of two-plus-two counterpropagating plane waves. Then the experimental implementation of this coherent oscillator with a $BaTiO_3$ crystal is described, and the results are compared with those calculated. Special attention is devoted to oscillation dynamics above the threshold and dynamics of photorefractive grating decay below the threshold: Revealed is critical behavior with a characteristic time going to infinity exactly at the threshold value of coupling strength. In this way, manifestation of the Curie–Weiss law is detected, for the first time to our knowledge, in a coherent optical oscillator.

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2. THRESHOLD AND OUTPUT CHARACTERISTICS OF THE OSCILLATOR

The coherent oscillator under consideration has the geometry depicted in Fig. 1. Two counterpropagating pump beams, 1 and 2, enter the sample from the opposite faces. For any signal beam this sample pumped with two waves acts as a phase-conjugate mirror with the reflectivity R_{pc},[1] therefore it can form an optical cavity with the ordinary mirror M. The condition of the self-excitation imposes, as for any other optical oscillator, that the intensity of the oscillation wave should be the same after every round trip of the cavity, i.e.,

$$RR_{pc} = 1, \tag{1}$$

with R being the reflectivity of the ordinary mirror. The steady-state phase-conjugate reflectivity can be calculated in the undepleted pump approximation from the known solution for backward-wave four-wave mixing in a photorefractive crystal[1]:

$$R_{pc} = \frac{\sinh^2\left(\dfrac{\gamma l}{2}\right)}{\cosh^2\left(\dfrac{\gamma l}{2} + \dfrac{\ln r}{2}\right)}, \tag{2}$$

where l is the sample thickness, $r = I_2(l)/I_1(0)$ is the pump-intensity ratio, γ is the coupling constant,

$$\gamma = -\frac{2\pi^2 r_{eff}\, n^3 k_B T \sin\theta}{e\lambda^2 \cos\theta}, \tag{3}$$

r_{eff} is the effective electro-optic coefficient, n is the refractive index, k_B is the Boltzmann constant, T is the absolute temperature, e is the electron charge, λ is the wavelength, and θ is the half-angle in the air between the pump wave and the oscillation wave.

Oscillation should occur if phase-conjugate reflectivity overpasses $R_{pc} \geqslant 1/R$. The threshold coupling strength $(\gamma l)_{th}$ necessary to reach self-oscillation is deduced from Eq. (2). For the optimized pump-intensity ratio, $\ln r = \gamma l$, we get

$$(\gamma l)_{th} = 2\ln\left(\frac{1 + \sqrt{1 + R}}{\sqrt{R}}\right). \tag{4}$$

With a highly reflecting cavity mirror ($R = 1$) the threshold coupling strength is $(\gamma l)_{th} \simeq 1.76$ for the pump ratio $r = 0.17$.

Linear-stability analysis predicts finite threshold coupling strength but does not indicate the type of bifurcation,[19] either supercritical or subcritical. To define the behavior above the threshold, we use the exact solution of the four-wave mixing problem, given by Cronin-Golomb *et al.*[1] The following relationships are deduced from their solutions.[1] They allow calculation of $I_3(0)$ as a function of γl, and $I_1(0)$ and $I_2(l)$ are given by the boundary conditions,

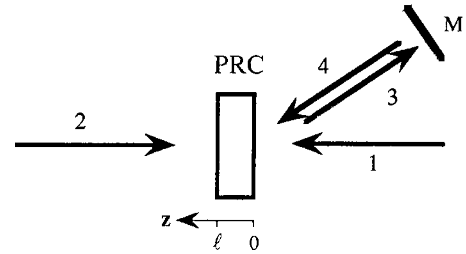

Fig. 1. Geometry of the oscillator considered.

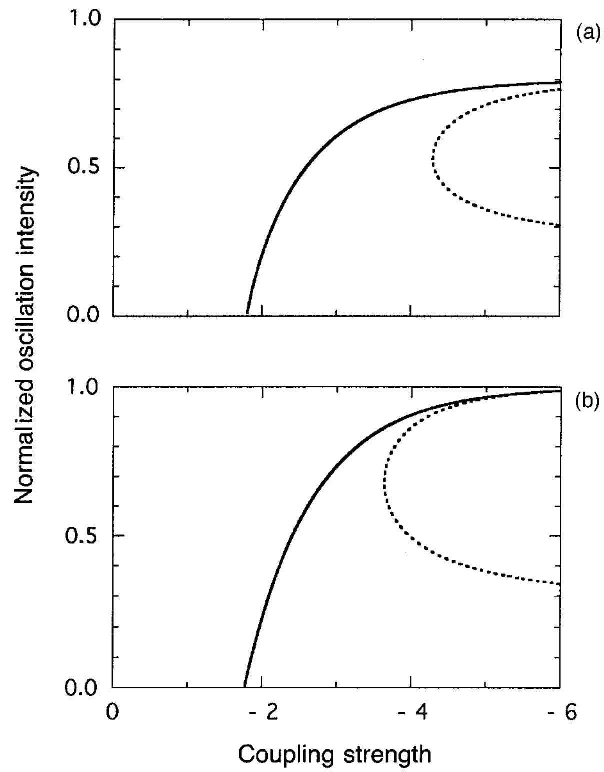

Fig. 2. Calculated coupling-strength dependence of the oscillation-wave output intensity for geometries (a) with two independent counterpropagating waves and (b) for the transmitted pump wave retroreflected into the sample.

$$\gamma l = \left(\frac{I_0}{\sqrt{\Delta^2 + 4|c|^2}}\right)\ln\left[\frac{|c|^2 - I_1(0)I_2(l) + I_3(0)I_2(l) - I_3(0)\sqrt{\Delta^2 + 4|c|^2}}{|c|^2 - I_1(0)I_2(l) + I_3(0)I_2(l) + I_3(0)\sqrt{\Delta^2 + |4|c^2}}\right], \tag{5}$$

$$|c|^2 = [\sqrt{I_1(0)I_2(l) - RI_1(0)I_3(0)} \pm I_3(0)\sqrt{R}]^2, \tag{6}$$

$$\Delta = I_2(l) - I_1(0) - RI_3(0), \tag{7}$$

$$I_0 = I_2(l) + I_1(0) + RI_3(0). \tag{8}$$

When deriving these equations, the coupling strength was assumed to be real (i.e., $\pi/2$ phase shift of the photorefractive grating with respect to fringes, which means a diffusion-driven charge transport and interaction strictly degenerate in frequency). As a consequence, phase difference of four interacting waves is considered to be independent of the propagation coordinate.

Figure 2(a) represents the calculated dependence of oscillation-wave intensity $I_3(0)$ normalized to I_0 [see Eq. (8)] as a function of coupling strength γl. The beam-intensity ratio is taken to be $r \simeq 0.17$, which is close to the experimental value, as described below. The soft-mode onset of oscillation is obvious, with the gradual increase of the oscillation-wave intensity from zero to saturated value for large coupling strength. The branch of the solution with the smallest threshold is single valued. The trivial solution with zero intensity for arbitrary coupling strength also exists. A double-valued solution shown by dots in Fig. 2(a) appears above a given coupling-strength value. This last branch is similar to that known for a semilinear oscillator with only one pump wave.[1] The lowest threshold coupling strength coincides, of course, with that calculated in the undepleted pump approximation.

One can see from Fig. 2 that in the vicinity of the threshold the intensity of the oscillation wave increases linearly with overthreshold coupling strength, $I_{\mathrm{osc}} = I_3(0) \propto [\gamma l - (\gamma l)_{\mathrm{th}}]$. Oscillation field amplitude $E_3(0)$ is therefore increasing as $E_3(0) \propto [\gamma l - (\gamma l)_{\mathrm{th}}]^{1/2}$, i.e., the derivative $\mathrm{d}E_3(0)/\mathrm{d}(\gamma l)$ becomes infinite at threshold. The similarity to a conventional free-running laser is evident: In lasers the output intensity I_{osc} is linearly increasing with over-threshold pump $I_{\mathrm{osc}} \propto (P - P_{\mathrm{th}})$, whereas oscillation field amplitude grows as $(P - P_{\mathrm{th}})^{1/2}$.

Thus we conclude that oscillation starts by supercritical bifurcation[19] of the order parameter that is the electric field of the oscillation wave for this optical phase transition. Consequently, the photorefractive oscillator considered manifests behavior typical for the second-order phase transition. Below, we present experimental confirmation of this behavior and give additional arguments in favor of second-order optical phase transition based on a study of the temporal dynamics.

It should be noted that the solution of Eqs. (5)–(8) can be extended for the particular practically useful geometry where the pump $I_1(0)$ is the wave $I_2(0)$ reflected by the beam splitter BS (see Fig. 3). Only one input wave $I_2(l) = 1$ is considered in this case, and the intensity of the second pump is given by an additional boundary condition:

$$I_1(0) = [I_2(l) - I_3(0)]R_p\,, \tag{9}$$

where R_p is the reflectivity of end beam splitter BS. Reflectivity R_p should account also for losses that are due to the Fresnel reflection from the sample face and for the sample absorption.

Figure 2(b) represents coupling-strength dependence of oscillation intensity for $R_p = 0.17$. Qualitatively, this dependence remains similar to that shown in Fig. 2(a); the threshold value of coupling strength is exactly the same as in the case of two independent pump waves because the pump ratio is not affected by diffraction from the grating at the threshold. The obvious advantage of a geometry with a retroreflected pump lies in better pump utilization.

The absorptionless case is considered in this section for the sake of simplicity. It is known that amplified phase-conjugate reflectivity (and therefore the coherent mirrorless oscillation) can be achieved also in nonlinear media with linear absorption.[20] Qualitatively, the output characteristics of the oscillator with linear-absorption losses remain similar to those shown in Fig. 2, but the threshold coupling strength becomes larger.

One more important assumption is a plane-wave approximation with a zero seed in the oscillation-wave direction. We used this approximation to explain qualitatively the underlying physical effect, profiting from the analytical solution for a simple model. For a more rigorous description of real coherent oscillators one should use the approach developed in Ref. 21 and numerical calculations with a seeding beam of finite spatial content.

3. EXPERIMENT

Schematic representation of the experimental arrangement is shown in Fig. 3. The beam of an Ar^+ laser (TEM_{00}, 0.514 μm) is used to pump a 3.6-mm-thick $BaTiO_3$ sample (photorefractive crystal, PRC). The transmitted pump beam is retroreflected by beam splitter BS to generate a counterpropagating pump wave. Another mirror M forms a semilinear cavity with the sample. The supplementary oblique light beam is used to align the cavity; it serves also as a seeding beam to record the grating below the oscillation threshold. It is polarized parallel to the sample optical axis, in the same way as pump beams. One more light beam, orthogonally polarized, is used to control the coupling strength, with its intensity varied. Partially transparent mirror M ($R = 0.96$) extracts part of the oscillation intensity measured with the detector Det placed behind this mirror.

With this arrangement, self-oscillation occurs when the seeding beam is stopped by shutter Sh and when the intensity of erasing beam I_E is below a certain threshold value. Typical temporal development of the oscillation intensity is shown in Fig. 4. Qualitatively, it resembles

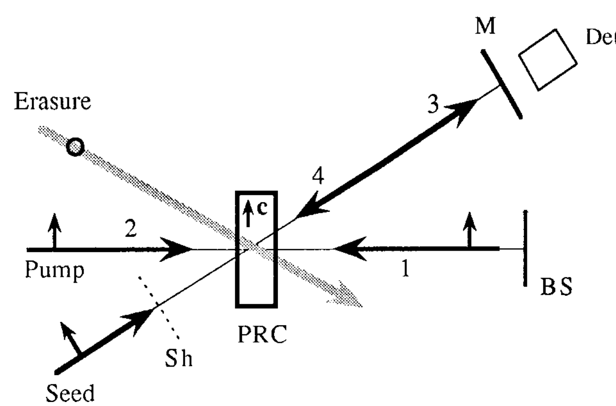

Fig. 3. Schematic representation of experimental arrangement: PRC is the photorefractive crystal sample with the ferroelectric axis in the plane of drawing, M is the mirror, BS is the beam splitter, Det is the photodetector, and Sh is the shutter.

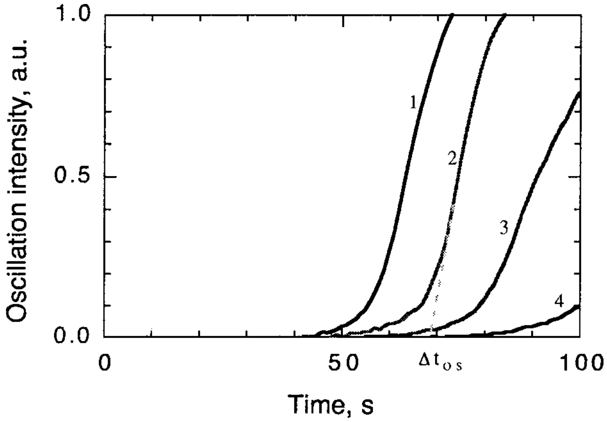

Fig. 4. Temporal evolution of oscillation intensity. Curves 1, 2, 3, and 4 correspond to the decreasing coupling strength. The dashed curve in curve 2 shows how oscillation delay time Δt_{os} is evaluated.

the coupling-strength dependence of the output intensity: For a comparatively long time the intensity of the oscillation beam is too weak to be seen in the shown plot, and then, after a certain delay time Δt_{os}, oscillation intensity grows nonlinearly and reaches the saturation level. Time delay Δt_{os} can be considered, in fact, as a temporal threshold of oscillation. Below Δt_{os} the light scattered in the direction of the future oscillation wave is gradually amplified, but it remains much smaller than the saturation value of oscillation intensity. Characteristic time Δt_{os} clearly divides the whole dynamics into two parts, one with well developed oscillation and the other resembling superluminescence of the amplifying media in conventional lasers.

Below the oscillation threshold, the light scattered in the direction of the oscillator optical axis is also amplified, and the amplification process can be characterized by a relevant time constant. With the feedback present, this time constant can vary with the coupling strength. An increase of the characteristic time of photorefractive scattering in the vicinity of the oscillation threshold was reported before in Ref. 5 for the double phase-conjugate mirror.

In our experiment, a characteristic time below the threshold is extracted in a different way, inspired by early publications on forced Rayleigh scattering.[22–24] In this technique the relaxation of an artificially "heated" selected spatial mode is studied instead of the relaxation of spontaneous gratinglike fluctuation.

First, the photorefractive grating is recorded by an auxiliary coherent light beam, sent to the sample exactly in the direction of the cavity optical axis and pump beam. Then the seeding beam is blocked with a shutter, and the intensity of the wave diffracted from the photorefractive grating is measured with the photodetector Det (see Fig. 3). Below the threshold, the recorded grating is not self-supported as happens above the threshold; therefore it decays, and the intensity of the diffracted wave drops. This is the measurement of the decay that provides information on the characteristic time below the threshold.

A characteristic decay curve is shown in Fig. 5(a). As distinct from the decay curves recorded with no feedback and pump wave 1 blocked [Fig. 5(b)], the temporal behavior is not exponential. The above makes it more complicated to evaluate the appropriate time constant: If taken at the beginning of erasure, it can be a few times smaller than at the tail of the erasure. We noted that the tails of the decay curves were less reproducible as compared with the initial part of the decay; quite often, even oscillatory behavior was observed for the remaining intensity of the weak diffracted beam. For this reason the initial linear slope of the decay curve is selected to extract relaxation time, i.e., the time when the signal drops to $(1/e)$ of its initial intensity. Substantial slowing of the relaxation process is clearly seen by comparing Figs. 5(a) and 5(b).

Further, we start measurements of oscillation intensity and characteristic delay time as a function of the coupling strength. As a rule, the coupling strength in photorefractive crystals is independent of pump intensity. To control the coupling strength, an additional light beam is sent to the sample, which partially erases the recorded grating to the level that depends on the intensity ratio of recording beams I_0 and erasing beam I_E. The erasing beam is ordinarily polarized to avoid recording of additional photorefractive gratings by a supplementary extraordinarily polarized beam. The coupling strength of the crystal is therefore

$$\gamma l = \frac{(\gamma l)_0}{1 + (I_E/I_0)}, \tag{10}$$

where $(\gamma l)_0$ is the initial coupling strength for $I_E = 0$.

The alternative technique to control coupling strength consists of changing the polarization of the incident pump beam.[5] The ordinary-polarization component appearing

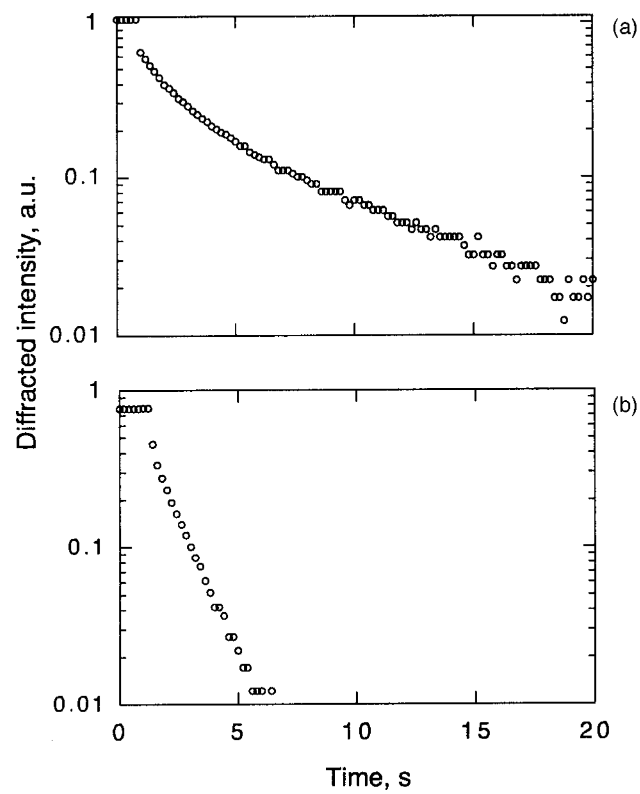

Fig. 5. Temporal dependence of grating decay below oscillation threshold. (a) Grating decay with counterpropagating pump-wave present; (b) decay with the beam splitter BS blocked.

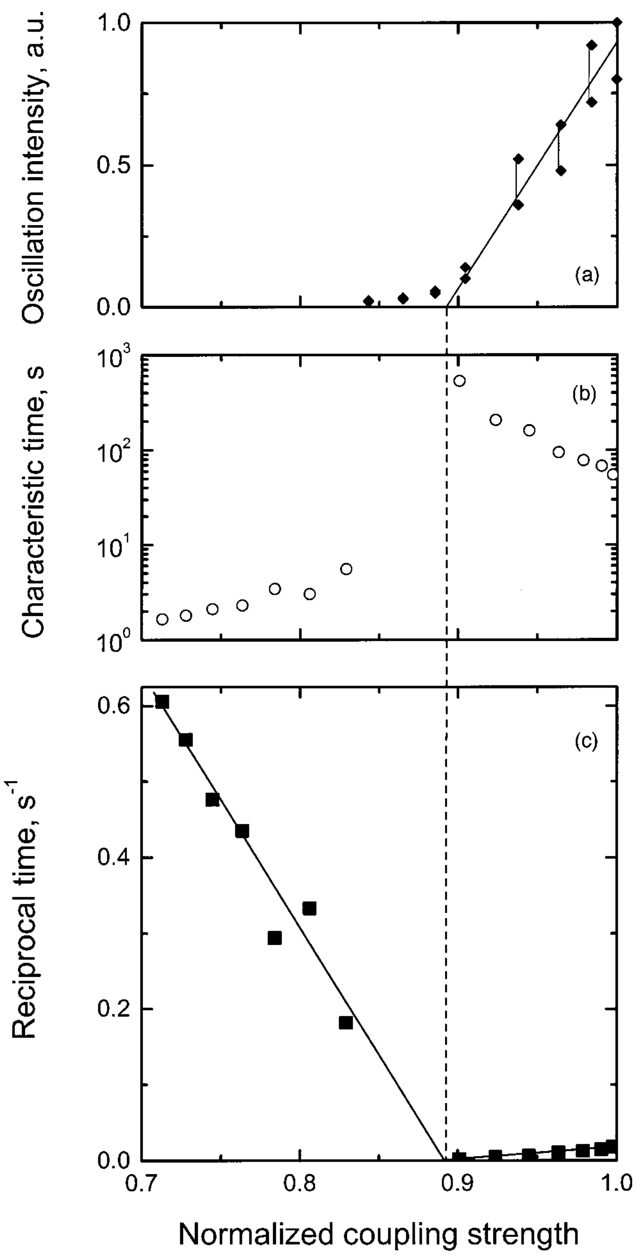

Fig. 6. Coupling-strength dependence of (a) oscillation intensity, (b) characteristic relaxation time, and (c) inverse characteristic time.

in the pump wave erases in part the grating, thus reducing γl. This technique is simpler and easier to implement, but it cannot be applied in the considered case: With two counterpropagating waves of mixed polarization, another type of mirrorless coherent oscillation develops,[25,26] and this affects the measurements considerably. That is why the technique with an additional Bragg-mismatched erasing beam is chosen to control the coupling strength.

Figure 6(a) shows the coupling-strength dependence of oscillation intensity. It is easily seen that oscillation intensity increases gradually above the threshold value of coupling strength $\gamma l \geqslant (\gamma l)_{\mathrm{th}}$. No discontinuity is observed exactly at the threshold. This points to the soft mode of oscillation onset from the noise level. Qualitatively, this dependence corresponds to the increase of oscillation intensity above the threshold shown in Fig. 2.

Figure 6(b) gives the plot of characteristic time below and above the threshold measured as explained above, and Fig. 6(c) shows the same dependence replotted for reciprocal relaxation time. The experimental points can be fitted with a linear dependence

$$\frac{1}{\tau} \propto |\gamma l - (\gamma)_{\mathrm{th}}|, \tag{11}$$

separately for the downward and upward slopes. These two fits give, within a few percent, the same value for $(\gamma l)_{\mathrm{th}} \simeq 0.89(\gamma l)_0$ that agrees well with that measured from the coupling-strength dependence of the oscillation intensity [Fig. 6(a)].

Dependences similar to the one shown in Figs. 6(a) and 6(b) were measured for different initial values of coupling strength $(\gamma l)_0$ controlled by the angle between the oscillator cavity-axis and pump-wave directions. The linear dependence given by Eq. (11) is always observed (see, e.g., our preliminary data in Ref. 27). Besides, for larger $(\gamma l)_0$ we were able to follow gradual variation of oscillation intensity near the threshold within four orders of magnitude.

Note that the slopes of linear dependences in Fig. 6(b) differ several times from each other. It is not, however, possible to compare these derivatives; no other conclusion can be made from these dependences apart from that τ^{-1} is a linear function of $\gamma l - (\gamma l)_{\mathrm{th}}$ and vanishes to zero exactly at threshold coupling strength $\gamma l = (\gamma l)_{\mathrm{th}}$ for both branches, below and above the threshold. The absolute values of τ [and the derivatives $d\tau/d(\gamma l)$] below and above the threshold cannot be compared because they are measured in essentially different ways; one is characteristic (nearly exponential) decay time of the diffraction efficiency, and the other one is effective (essentially nonexponential) buildup time. It is known also that oscillation buildup time depends strongly on the initial level of the seeding radiation,[21,28] whereas the decay time of the prerecorded grating is supposed to be independent of the initial diffraction efficiency of this grating.

4. DISCUSSION

Our calculations and experimental data confirm the similarity of onset of coherent oscillation in a semilinear photorefractive oscillator and second-order phase transition. The soft mode of oscillation onset is observed, with a gradual increase of oscillation intensity from the scattering noise up to saturation. The latter corresponds to a continuous increase of the order parameter (normalized amplitude of the oscillation-wave electric field) from zero level to a certain saturation level, smaller or equal to one for order–disorder phase transitions. Note also that the derivative of the order parameter is infinite exactly at the threshold, as it should be for a second-order transition.

Critical behavior is detected in the vicinity of the threshold, confirming the results of Ref. 8. Reciprocal relaxation time is shown to be proportional to $|\gamma l - (\gamma l)_{\mathrm{th}}|$. Such dependence is typical for the second-order phase transitions, for which different parameters increase with temperature as $|T - T_c|^{\zeta}$, with ζ standing for the critical exponent and T_c being a transition temperature (Curie temperature). For example, for ferromagnetic phase transition the temperature dependence of magnetic susceptibility χ is described by the Curie–Weiss law,[29] i.e., χ is proportional to $|T - T_0|^{-1}$, with T_0 standing for Curie–Weiss temperature. Curie–Weiss law holds also for some ferroelectric crystals, for temperature dependence of dielectric constant ϵ, and for piezoelectric and elasto-optic properties above the transition temperature.[30] Thus the observed coupling-strength dependence of τ^{-1} can be considered as an optical analogy of the Curie–Weiss law.

It should be emphasized that the Curie–Weiss temperature coincides with the transition temperature for the second-order phase transitions but it does not for the first-order phase transitions.[30] Thus the coincidence of the coupling-strength value at which the reciprocal characteristic times diverge with the threshold coupling strength also points to the second-order phase transition.

In the vicinity of the threshold, the critical exponent approaches 1, both for temporal development of scattering below the transition and for development of oscillation above the transition. Critical slowing below the threshold of mirrorless oscillation has been reported for photorefractive $Bi_{12}SiO_{20}$ crystals[7] and $BaTiO_3$ crystals.[8] The critical exponent equal to one has been measured for $Bi_{12}SiO_{20}$. For the temporal dynamics of some other optical processes such as switching in optically bistable étalons,[31,32] the critical exponent can be 1/2, i.e., significantly different from 1.

Considering the Landau theory of second-order phase transitions, one could expect asymmetry in slopes of linear dependences $\tau_0{}^{-1} = \tau^{-1}[\gamma l - (\gamma l)_{\mathrm{th}}]$ below and above the transition point to occur; in the ideal case of no fluctuations, the universal ratio of the slopes equal to 2 should hold. We do observe the difference in prefactors of linear dependences of the reciprocal relaxation times on coupling strength, as seen in Fig. 6(b) (as the authors of Ref. 5 have observed for the double phase-conjugate mirror), but the unknown seed level and different techniques used for evaluation of τ above and below the threshold prevent us for the moment from a correct comparison with theoretical predictions.

The arguments given above prove, in our opinion, that the threshold of oscillation in a semilinear cavity with two counterpropagating waves can be considered as a second-order phase transition within the investigated range of parameters. There is still a question of whether this similarity is valid for any semilinear oscillator with two pump waves or not. The background of this question is related to the known property of a semilinear oscillator with only one pump wave: This oscillator exhibits subcritical bifurcation and is therefore close to a first-order phase transition.[1,33]

Additional computer simulations of oscillation intensity versus coupling strength (as shown in Fig. 2) revealed a change of bifurcation type at $r \simeq 0.02$. For $r = 0.01$ the width of the expected hysteresis loop in the coupling strength (the difference in the threshold coupling strength for increasing coupling and decreasing coupling, normalized by the threshold coupling strength itself) becomes approximately 0.1. This value is large enough to be measured experimentally, but the absolute value of coupling strength $(\gamma l)_{\mathrm{th}} \simeq 2.47$ is too high to be reached with the $BaTiO_3$ sample available.

5. CONCLUSIONS

The conclusion about the second-order optical phase transition in a semilinear coherent photorefractive oscillator is based on the following observations: (1) a soft mode of oscillation onset (a gradual increase of the oscillation field from zero to saturated value), (2) an infinite derivative $\mathrm{d}E_{\mathrm{osc}}/\mathrm{d}(\gamma l)$ at the threshold coupling strength, (3) detection of the inverse power law for grating relaxation and buildup, with a critical exponent equal to 1 (analogous to the Curie–Weiss law), and (4) the coincidence of Curie–Weiss coupling strength and threshold coupling strength. Thus the formal analogy of the oscillation threshold in the oscillator considered to second-order phase transition is quite evident, but the question still remains of how far this analogy may go. The open fundamental questions are, e.g., in which way free energy for optical phase transition can be introduced[5] and interpreted, and what is an equivalent of the susceptibility for optical phase transition. The answers for these and other related questions will be found, we hope, in future work on this subject.

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