Instability of Single-Frequency Operation in Semilinear Photorefractive Coherent Oscillators

P. Mathey,[1] S. Odoulov,[2] and D. Rytz[3]

[1]*Laboratoire de Physique, Université de Bourgogne, 21078 Dijon, France*

[2]*Institute of Physics, National Academy of Sciences, 03650 Kiev, Ukraine*

[3]*Forschunginstitute für mineralische und metalische Werkstoffe, Edelsteine/Edelmetalle GmbH, 55743 Idar-Oberstein, Germany*

(Received 12 February 2002; published 15 July 2002)

The transition of the single-frequency oscillation of a semilinear photorefractive coherent oscillator for sufficiently large coupling strengths into two-frequency oscillation is predicted and is observed experimentally. The critical value of the coupling strength at which the bifurcation occurs is a function of pump intensity ratio and cavity losses. The supercritical bifurcation in the oscillation spectrum is analogous to the second-order phase transition.

DOI: 10.1103/PhysRevLett.89.053901 PACS numbers: 42.65.Hw, 05.45.–a, 42.65.Pc, 42.65.Sf

Nonlinear mixing of four coherent light waves nearly degenerate in frequency may lead to amplification of a weak signal beam [1]. This amplification, in turn, can result in self-excitation of coherent oscillation providing that the appropriate feedback is ensured [2]. The most widely used nonlinear media to study the four-wave mixing coherent oscillators are the photorefractive crystals (see, e.g., [3,4]). These oscillators were extensively studied during the last decade as suitable model systems for the investigation of spatial and temporal dynamics [5–9]. It has been pointed out, in particular, that the onset of oscillation at the threshold may be considered as an optical phase transition with the properties of either second-order [10] or first-order [11] phase transition.

Exactly at the coherent oscillation threshold only one photorefractive grating with a well defined grating vector emerges from the multitude of low-amplitude arbitrarily oriented noisy gratings responsible for light-induced (nonlinear) scattering in photorefractive crystals. The amplitude of this selected grating increases rapidly with the increasing coupling strength which is a control parameter here. The light wave diffracted from the developing grating is the oscillation wave. Its amplitude normalized to the amplitude of the pump wave is an order parameter [10,12].

In the present Letter we describe one more type of instability for already well developed oscillation in a semilinear cavity: at a certain critical value of the coupling strength $(\gamma\ell)_{cr}$ above the threshold of coherent oscillation the immobile photorefractive grating splits into two gratings with the same grating vectors but moving in opposite directions (here γ is the coupling constant [3] and ℓ is the sample thickness). Being the result of diffraction from the moving grating the oscillation wave acquires Doppler frequency shift Ω with respect to the pump wave frequency ω. As a consequence the oscillation spectrum above $(\gamma\ell)_{cr}$ consists of two lines $\omega \pm \Omega$ separated by 2Ω, instead of single-frequency ω below $(\gamma\ell)_{cr}$. The characteristic frequency shifts are of the order of reciprocal dielectric relaxation time (i.e., buildup time of photorefractive nonlinearity). For cw laser intensities used Ω changes from zero to several Hz.

The underlying physical phenomenon is related to an enhancement of the phase conjugate reflectivity of the backward wave four-wave mixing phase conjugate mirror with the diffusion-driven nonlocal photorefractive nonlinearity for the signal wave with frequency detuned from that of the pump wave [13].

Earlier the instability in temporal behavior (and therefore in spectral domain) was revealed for the semilinear photorefractive oscillator [7] and attributed to the excitation of multiple transverse modes (the Fresnel number of the cavity was chosen as a control parameter in these experiments). We show in this Letter that the spectrum instability is also an inherent feature of a single-transverse-mode semilinear oscillator. This instability of oscillation spectrum can be considered as an optical phase transition, too, that has no analogy, however, in structural phase transitions in solid state physics. Apart from the fundamental interest for nonlinear dynamics the observation of instability may be of practical importance: The semilinear photorefractive oscillators have been seriously considered as nonlinear mirrors that can reduce the divergence of conventional lasers [4].

In what follows, first the experimental observations for the semilinear photorefractive oscillator with two counterpropagating pump waves are described. Then we present a qualitative explanation and the results of our computer simulations. Finally the experimental results are compared with calculated results.

In the experiment the $BaTiO_3$:Co sample measuring $3.7 \times 4.0 \times 6.1$ mm^3 is pumped by two loosely focused counterpropagating waves from an Ar^+ laser (0.51 nm, TEM_{00}, no etalon inside the cavity to prevent from reflection grating recording); see Fig. 1. The variable beam splitter allows one to control the pump beam intensity ratio keeping nearly constant the total intensity of light in the sample. The pump waves 1 and 2 enter the sample through opposite faces; they make an angle about 4° to the optical axis **c** inside the sample. Both pump waves are

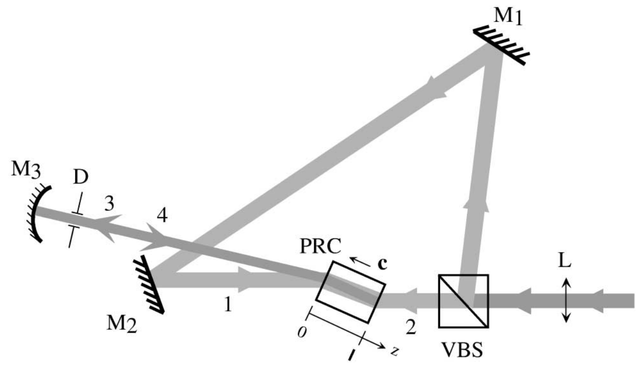

FIG. 1. The experimental setup: PRC is the photorefractive crystal; M_1, M_2, and M_3 are the mirrors; VBS is the variable beam splitter; D is the aperture, L is the lens; ℓ is the interaction length; the numbers from 1 to 4 denote the interacting light waves; the arrow indicates the direction of the crystal polar axis **c**.

extraordinarily polarized. Pumped in such a manner the $BaTiO_3$ crystal becomes a phase conjugate mirror; it ensures more than 100% reflectivity for an appropriately oriented signal wave. Together with the high-reflecting ordinary mirror M_3 (concave with 50 cm radius) the $BaTiO_3$ crystal forms a semilinear photorefractive oscillator. The cavity length is about 31 cm; the aperture of 0.5 mm diameter is placed inside to select the lowest transverse mode in oscillation. The oscillation waves that self-develop in this cavity, 3 and 4, are tilted inside the sample to about 2° with respect to the optical axis **c** to maximize the coupling strength [14].

With a similar geometry of the coherent oscillator and the same photorefractive sample we showed earlier that the onset of oscillation features the properties of a second-order phase transition, including critical behavior in the vicinity of the threshold coupling strength $(\gamma\ell)_{th}$ [12].

In the used $BaTiO_3$ sample, as in most other photorefractive crystals with a pronounced photoconductivity, the coupling strength $(\gamma\ell)$ is independent of the pump intensities. A more suitable control parameter that allows one to affect the phase conjugate reflectivity is the pump intensity ratio, $r = I_2(\ell)/I_1(0)$ (see Fig. 1). The other option for a control parameter is the reflectivity of the conventional mirror R that defines cavity losses.

We have found that depending on the pump intensity ratio the oscillation intensity is either constant in time or exhibits high contrast sinusoidal modulation with frequency 2Ω. Such a modulation proves the existence of two components in the oscillation spectrum. If a reference wave with the frequency of the pump wave ω (local oscillator) is sent to the detector together with the oscillation wave, the main beat frequency becomes equal to Ω. This leads to the conclusion that two components in the oscillation spectrum are symmetrically shifted to $\pm\Omega$ with respect to the pump frequency ω.

Figure 2 shows the dependence of the oscillation frequencies versus the pump intensity ratio r. It is obvious

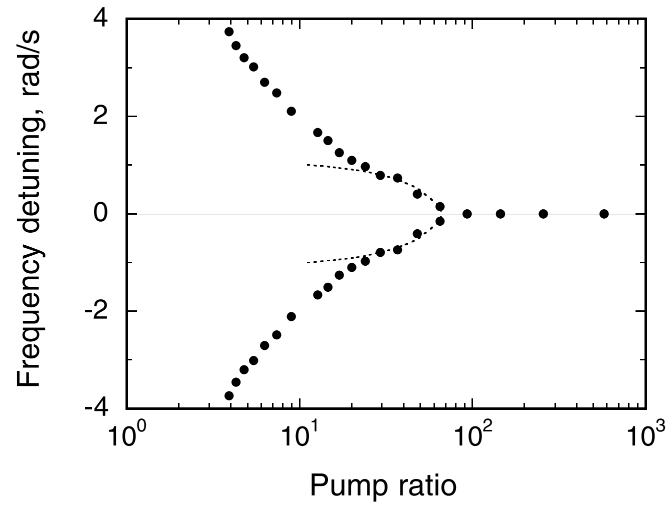

FIG. 2. Experimentally measured frequency detuning of the oscillation wave versus pump intensity ratio. The dashed line shows a fit of Eq. (1) to experimental data below the bifurcation point.

that for pump ratios below critical value $r_{cr} \simeq 70$ the single-frequency oscillation splits into symmetric double-frequency oscillation; the frequency separation is increasing with the diminishing r. The coupling strength $\gamma\ell$ is constant in this experiment; it is large enough to observe the bifurcation. All data in Fig. 2 correspond to the same total light intensity inside the sample. For any fixed pump ratio smaller than the critical value r_{cr} it is found that the frequency separation of the two oscillating modes is scaling with the light intensity I nearly linearly, $\propto I^{0.83}$, as one can expect for a medium with the relaxation of nonlinearity governed by photoconductivity (see, e.g., [3]). The exponent different from unity is typical for crystals with shallow traps [15]; the measured value (0.83) coincides with that reported for this sample earlier [16].

In close vicinity below r_{cr} the frequency shift behaves as

$$\Omega \propto \sqrt{(\ln r_{cr} - \ln r)/\ln r_{cr}}\,; \qquad (1)$$

see the dashed line in Fig. 2.

The coherent oscillation in the considered geometry self-develops from the noise and its frequency is not imposed by any specific frequency in the broad and smooth spectrum of the seeding radiation. On the other side, the semilinear oscillator belongs to oscillators with unclosed cavities for which the spectrum of cavity longitudinal modes is discrete but the eigenfrequencies are not fixed [15]. The cavity mode frequency does not affect the oscillation frequency because of the self-adaptable nature of the phase conjugate mirror. It is obvious also that the conventional mirror of the cavity cannot possess spectral selectivity within the region of several Hz. One should look therefore for a reason for the bifurcation in the oscillation spectrum that is related to gain, i.e., to spectra of amplified phase conjugate reflectivity.

It is known that for large values of the coupling strength the maxima of the phase conjugate reflectivity $R_{pc} = I_3(0)/I_4(0)$ correspond to certain frequency detuning $\Omega \neq 0$ while for small coupling $R_{pc}(\Omega)$ is bell-shaped with the maximum exactly at $\Omega = 0$. This experimental observation with the passive phase conjugate mirror was supported by calculation of the spectra of phase conjugate reflectivity [13].

Suppose that waves 1 and 4 are recording a transmission grating in the photorefractive sample (Fig. 1) and wave 2 is reading out this grating giving rise to conjugate wave 3. These two waves, 2 and 3, are recording also a photorefractive grating that appears to be out of phase (π shifted) with respect to the initial grating. If degeneracy in frequency is removed the in phase component appears in the secondary grating and its amplitude is increasing with frequency detuning [3,4]. The diffraction efficiency of the resulting grating is therefore increasing, too. This gives a hint as to why the phase conjugate reflectivity may become larger for signal wave 3 shifted in frequency with respect to pump waves 1 and 2.

The above description allows one also to explain why the pump ratio can serve as a control parameter here. For strong disbalance of the pump intensities the amplitudes of two partial gratings (one recorded with waves 1 and 4 and the other recorded with waves 2 and 3) are quite different. The destructive superposition of two gratings in this case is not dangerous: Even in the case $\Omega = 0$ the overall grating can have a sufficiently large amplitude. This is why for large values of r the oscillation may be frequency degenerate.

For decreasing r approaching to unity the amplitudes of two out-of-phase gratings become comparable and their compensation effect for $\Omega = 0$ becomes stronger. The phase conjugate reflectivity drops down for $\Omega = 0$, while it may still be large for the frequency shifted signal wave. This results in the bifurcation in the oscillation spectrum below r_{cr}.

In what follows we present the results of calculation of oscillation spectra for the considered semilinear photorefractive oscillator. A simple model of nonlinear mixing of four plane waves is considered within the undepleted pump approximation [2,13]. The plane wave approximation is well justified because the oscillation wave and pump waves are all plane waves with the intensity profiles close to Gaussian. Within the undepleted pump approximation the results of calculation are valid, strictly speaking, for coupling strengths only slightly higher than the threshold values. The comparison will show, however, a rather good qualitative agreement between calculated and measured data even for well developed oscillation, i.e., far above the threshold.

We do not impose in advance any restrictions to the frequency of the oscillating wave except the requirement that the spectrum should be symmetric with respect to $\Omega = 0$. This is an inherent property of any oscillator with a phase conjugate mirror [17]: Every time when the oscillation wave is reflected from the phase conjugate mirror the sign of its frequency shift Ω is inverted. This is the reason why the field inside the cavity is self-reproduced only after two cavity round-trips [1,17], and the field possesses symmetrically shifted components $\omega \pm \Omega$.

In the undepleted pump approximation the phase conjugate reflectivity reads [3,4]

$$R_{pc} = \frac{\sinh^2(\frac{\gamma''\ell}{2}) + \sin^2(\frac{\gamma'\ell}{2})}{\cosh^2(\frac{\gamma''\ell - \ln r}{2}) - \sin^2(\frac{\gamma'\ell}{2})}, \tag{2}$$

with the coupling strength that becomes complex in the nondegenerate case

$$\gamma\ell(\Omega) = \frac{\gamma_0\ell}{1 + i(\Omega\tau)^2} + i\,\frac{\Omega\tau\gamma_0\ell}{1 + i(\Omega\tau)^2} = \gamma''\ell + i\gamma'\ell, \tag{3}$$

τ being the refractive index decay time, and $\gamma_0\ell$ being the crystal coupling strength for $\Omega = 0$.

To calculate the spectra of R_{pc} for different initial coupling strengths $\gamma_0\ell$ and pump ratios r it was sufficient to solve Eqs. (2) and (3) together [4,13]. In order to find the oscillation frequencies for the considered coherent oscillator one should find the maxima of $R_{pc}(\Omega)$, i.e., to require that

$$\frac{dR_{pc}}{d\Omega} = 0, \tag{4}$$

and take into account also the amplitude condition of oscillation. The latter postulates that the oscillation wave is self-supported if cavity losses (represented by reflectance R of the conventional mirror) are compensated for by amplified reflection from the phase conjugate mirror R_{pc}:

$$RR_{pc} = 1. \tag{5}$$

Equations (2)–(5) have been solved numerically to find the dependences of threshold coupling strength $(\gamma\ell)_{th}$ and threshold frequency detuning Ω_{th} versus pump intensity ratio r with the cavity mirror reflectivity R taken as a parameter.

Figure 3 represents the calculated threshold frequency detuning to be compared with the experimentally measured frequency detuning. Three bifurcating curves are plotted for the cavity mirror reflectivities $R = 1$, 0.1, and 0.01.

A satisfactory qualitative agreement between the experimental and calculated dependences is obtained. Quantitative agreement is less good, however; the experimental curve measured with a highly reflecting end mirror $R = 1$ is closer to that calculated for $R = 0.1$ (highlighted curve in Fig. 3). This discrepancy is most probably due to other cavity losses not taken into account in calculation (Fresnel reflections from sample face, light absorption in the sample, and incomplete overlap of oscillation wave and pump wave inside the sample) and also due to the obvious violation of undepleted pump approximation (what strongly affects the pump ratio inside the sample).

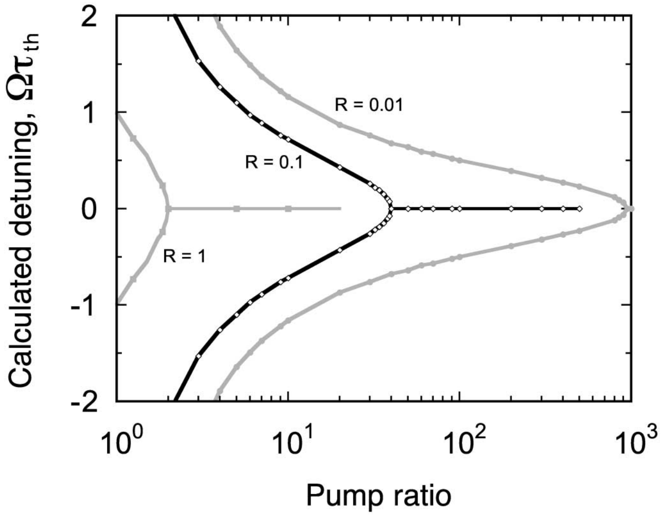

FIG. 3. Calculated frequency detuning of the oscillation wave versus pump intensity ratio for three different values of end mirror reflectivity.

The computer simulations confirmed the supercritical type of bifurcation in frequency spectra, Eq. (1). A similar equation can be derived for Ω with the coupling strength $\gamma''\ell$ taken as a parameter. The simulations show that the onset of double-frequency oscillation occurs not far away from the parameters defined by the condition $\gamma''\ell \simeq \ln r$ (usually this condition optimizes the phase conjugate reflectivity [4]). Taking into account the definition of $\gamma''\ell$, Eq. (3), we arrive at

$$\Omega\tau \propto \sqrt{\frac{(\gamma_0''\ell) - (\gamma''\ell)_{cr}}{(\gamma''\ell)_{cr}}}. \tag{6}$$

Such a type of bifurcation suggests that the discovered spectrum instability is analogous to the second-order optical phase transitions. The frequency detuning can be considered as an order parameter: It is zero at the bifurcation point and is increasing until the ultimate saturated value that is limited by the necessity to maintain a sufficiently large amplitude of the refractive index grating. Therefore the normalized frequency detuning is changing from zero at the splitting threshold to unity in saturation.

It should be noted that even in a single-transverse mode operation the temporal instabilities may appear because of other reasons as, for example, symmetry breaking [18–20] or thermally induced changes of the cavity length [20]. We believe, however, that these sources of instability are much less critical in the considered oscillator with the unclosed semilinear cavity than in oscillators with classical closed ring cavities.

To summarize it is shown that a photorefractive coherent oscillator with two counterpropagating pump waves features frequency bifurcation that is related to deformation of phase conjugate reflectivity spectra at sufficiently high values of coupling strength. In the future it will be interesting to find out to what extent this bifurcation may affect the pattern dynamics in photorefractive oscillators with closed cavities and high Fresnel numbers [19,20]. The similarity to the phase transitions raises the question about the particularities of temporal dynamics near the threshold of bifurcation and analogies to critical behavior below the threshold.

The authors are grateful to H. R. Jauslin and A. Shumelyuk for helpful discussions.

[1] A. Yariv, *Quantum Electronics* (John Wiley & Sons, New York, 1989), pp. 495–527.
[2] D. Pepper and A. Yariv, Opt. Lett. **1**, 16 (1977).
[3] L. Solymar, D. J. Webb, and A. Grunnet-Jepsen, *The Physics and Applications of Photorefractive Materials* (Clarendon Press, Oxford, 1996).
[4] S. Odoulov, M. Soskin, and A. Khyzhnyak, *Optical Coherent Oscillators with Degenerate Four-Wave Mixing* (Harwood Academic Publishers, Chur, London, 1991), pp. 37–39.
[5] F. T. Arecchi, G. Giacomelli, P. L. Ramazza, and S. Residori, Phys. Rev. Lett. **65**, 2531 (1990); **67**, 3749 (1991).
[6] W. Królikowski, M. Belić, M. Cronin-Golomb, and A. Błędowski, J. Opt. Soc. Am. B **7**, 1204 (1990).
[7] S. Liu and G. Indebetouw, J. Opt. Soc. Am. B **9**, 1507 (1992).
[8] K. Staliunas, M. F. H. Tarroja, G. Slekys, C. O. Weiss, and L. Dambly, Phys. Rev. A **51**, 4140 (1995); K. Staliunas, V. Taranienko, G. Slekys, R. Viselga, and C. O. Weiss, Phys. Rev. A **57**, 599 (1998).
[9] T. Honda, H. Matsumoto, M. Sedlatschek, C. Denz, and T. Tschudi, Opt. Commun. **133**, 293 (1997); C. Denz, M. Schwab, M. Sedlatschek, T. Tschudi, and T. Honda, J. Opt. Soc. Am. B **15**, 2057 (1998).
[10] D. Engin, S. Orlov, M. Segev, G. Valley, and A. Yariv, Phys. Rev. Lett. **74**, 1743 (1995).
[11] S. Odoulov, M. Goul'kov, and O. Shinkarenko, Phys. Rev. Lett. **83**, 3637 (1999).
[12] P. Mathey, P. Jullien, S. Odoulov, and O. Shinkarenko, Appl. Phys. B **73**, 71 (2001).
[13] K. R. McDonald and J. Feinberg, Phys. Rev. Lett. **55**, 821 (1985).
[14] Y. Feinman, E. Klancnik, and S. H. Lee, Opt. Eng. **25**, 228 (1986).
[15] J. D. Mahgerefteh and J. Feinberg, Phys. Rev. Lett. **64**, 2195 (1990).
[16] P. Mathey and B. Mazué, P. Jullien and D. Rytz, J. Opt. Soc. Am. B **15**, 1353 (1998).
[17] P. A. Belanger, A. Hardy, and A. Siegman, Appl. Opt. **19**, 602 (1980).
[18] A. Mazur and S. Odoulov, IEEE J. Quantum Electron. **26**, 963 (1990).
[19] D. Hennequin, L. Dambly, D. Dangoisse, and P. Glorieux, J. Opt. Soc. Am. B **11**, 676 (1994).
[20] Z. Chen, D. McGee, and N. B. Abraham, J. Opt. Soc. Am. B **13**, 1482 (1996).