Appl. Phys. B 73, 711–715 (2001)
DOI: 10.1007/s003400100728

Applied Physics B
Lasers and Optics

P. MATHEY[1]
P. JULLIEN[1]
S.G. ODOULOV[2,✉]
O.A. SHINKARENKO[2]

Manifestation of Curie–Weiss law for optical phase transition

[1] Laboratoire de Physique, Université de Bourgogne, BP 47870, 21078 Dijon, France
[2] Institute of Physics, National Academy of Sciences, 03650 Kiev-39, Ukraine

Received: 2 April 2001 / Revised version: 23 August 2001
Published online: 7 November 2001 •

ABSTRACT Considerable slowing down is observed for both the temporal development of the coherent oscillation slightly above the threshold and the refractive index grating decay slightly below the threshold for a semilinear photorefractive oscillator with two counter-propagating pump waves. It is shown that in the vicinity of the threshold the reciprocal characteristic time is a linear function of deviation from the threshold coupling strength. This behaviour is similar to an empirical Curie–Weiss law and points to the analogy of the oscillation threshold to a second-order phase transition.

PACS 42.65.Hw; 42.65.Sf; 42.65.P

1 Introduction

The threshold of oscillation in photorefractive coherent oscillators features similarities to phase transitions in solid-state physics and hydrodynamics [1–4]. Below the threshold the sample illuminated with the pump beam exhibits the light-induced scattering in a wide solid angle (beam fanning). This scattering is a consequence of the pump-wave diffraction from the arbitrarily oriented low-amplitude noisy photorefractive gratings which 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. Thus the system changes qualitatively: a new, highly ordered state emerges from a completely disordered state. The amplitude of the oscillation field normalized to the amplitude of the pump field may be considered as the order parameter: it is identically zero below the threshold (below the transition point) and tends to unity at saturation far above the threshold. The natural control parameter is the coupling strength, which is proportional to light-induced change of the refractive index times interaction length.

It should be noted that the similarity of the oscillation threshold in the usual lasers to the phase transition has been known since the classical work of Haken [5]. For free-running lasers the gain shows typical characteristics of a second-order phase transition [6]. Surprisingly, the number of photorefractive coherent oscillators with different cavity configurations and considerably different properties is much larger than the number of known laser classes. Some of the photorefractive oscillators exhibit the features of the second-order phase transition [2, 3] while the others show those of the first-order phase transition [4]. A priori, it is not clear to what extent one can extend conclusions formulated for any particular oscillator to all other possible oscillator geometries.

In this paper we describe the threshold behaviour of a semilinear coherent oscillator with two counter-propagating pump waves, using a $BaTiO_3$ crystal as an amplifying medium [7, 8]. Special attention is devoted to the study of the oscillation dynamics above the threshold and the dynamics of the photorefractive grating decay below the threshold: the critical behaviour is revealed, with a characteristic time going to infinity exactly at the threshold value of the coupling strength. In such a way the manifestation of the Curie–Weiss law is detected, for the first time to our knowledge, in a coherent optical oscillator. This observation confirms that within the range of experimental parameters the second-order optical phase transition is observed, as we expected from the calculated coupling strength dependence of the steady-state output intensity. The detected critical slowing down (which follows directly from the analogy of oscillation threshold and phase transition) is important for correct evaluation of the oscillation dynamics for conventional lasers that incorporate a semilinear coherent oscillator as a part.

2 Experiment

2.1 *Experimental procedure*

The schematic representation of the experimental arrangement is shown in Fig. 1. The beam of an Ar^+ laser (TEM_{00}, 0.514 μm) is used to pump a 3.6 mm thick $BaTiO_3$ sample (PRC); it impinges upon the sample at the angle of 45° in air. The transmitted pump beam is retroreflected by a beam splitter BS to generate a counter-propagating pump wave. Another mirror M forms the semilinear cavity with the sample. The cavity axis is tilted with respect to the sample normal at 25° in air so that the angle between the pump and oscillation waves is 20° in air.

✉ E-mail: odoulov@iop.kiev.ua

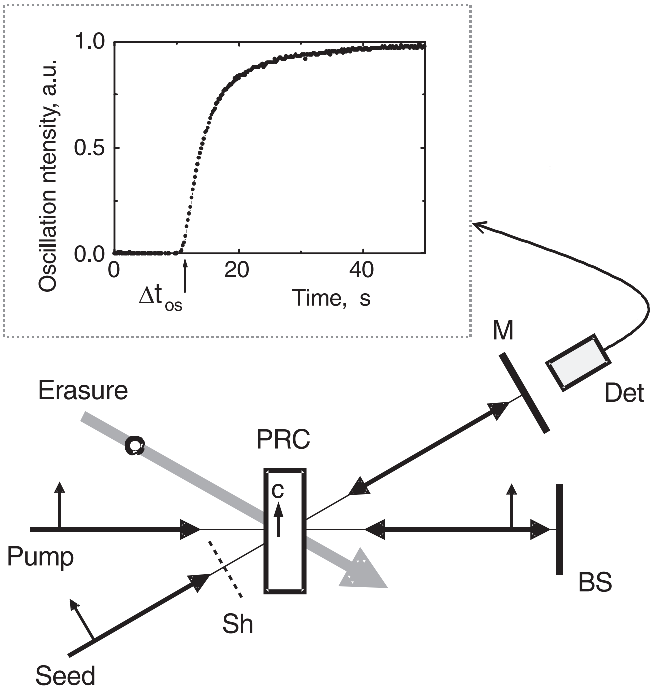

FIGURE 1 Schematic representation of the experimental arrangement. PRC is the photorefractive sample with ferroelectric axis in the plane of the drawing, M is the mirror, BS is the beam splitter, Det is the photodetector, Sh is the shutter. The *inset* shows typical temporal evolution of oscillation intensity. Coupling strength for particular angles between the beams given in the text is $\gamma\ell \simeq 2$

The supplementary oblique light beam (seed) is used to align the cavity; it serves also as a seeding beam to record the grating below the oscillation threshold. Like the pump beams this one is polarized parallel to the sample optical axis (extraordinary wave). Another orthogonally polarized light beam (ordinary wave) is used to control the coupling strength, by varying its intensity. Partially transparent mirror M ($R = 0.96$) extracts a part of the oscillation intensity, measured with the detector Det placed behind this mirror.

2.2 *Evaluation of characteristic times*

With this arrangement, the self-oscillation occurs when the seeding beam is stopped by a shutter Sh and when the intensity of the erasing beam I_E is below a certain threshold value. Typical temporal development of the oscillation intensity is shown in the inset of Fig. 1. It resembles the known one for e.g. a double phase-conjugate mirror [2] and a ring-loop oscillator [3]. For a comparatively long time the intensity of the oscillation beam is very weak, but starting from a certain delay time Δt_{os} the oscillation intensity grows nonlinearly and reaches the saturation level quickly. The time delay Δt_{os} can be considered 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 the oscillation intensity. The characteristic time Δt_{os} clearly divides the whole dynamics into two parts, one with a well-developed oscillation and the other that resembles the superluminescence of the amplifying media in the usual 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. When feedback is applied, this time constant can vary with the coupling strength. The increase of the characteristic time of the photorefractive scattering in the vicinity of the oscillation threshold has already been reported for other oscillator geometries [2, 3, 9].

In our experiment the characteristic time below the threshold is evaluated in a different way, inspired by the early studies on forced Rayleigh scattering [10]. In this technique the relaxation of an artificially 'heated' selected spatial mode is studied instead of the relaxation of the spontaneous grating-like fluctuation [11].

First, a photorefractive grating is recorded by the pump beam and auxiliary coherent light beam, sent to the sample exactly in the direction of the cavity optical axis (seed beam in Fig. 1). Then the seeding beam is blocked with the shutter Sh and the intensity of the wave diffracted from the photorefractive grating is measured with photodetector Det. Below the threshold the grating which has been recorded by the pump and seed beams is not self-supported; it decays and the intensity of the diffracted wave drops.

The characteristic decay curve is shown in Fig. 2a. For comparison, Fig. 2b shows the decay curve recorded with no feedback (mirror M tilted) and counter-propagating pump wave blocked (the wave reflected from the beam splitter BS is stopped). A substantial slowing-down of the relaxation pro-

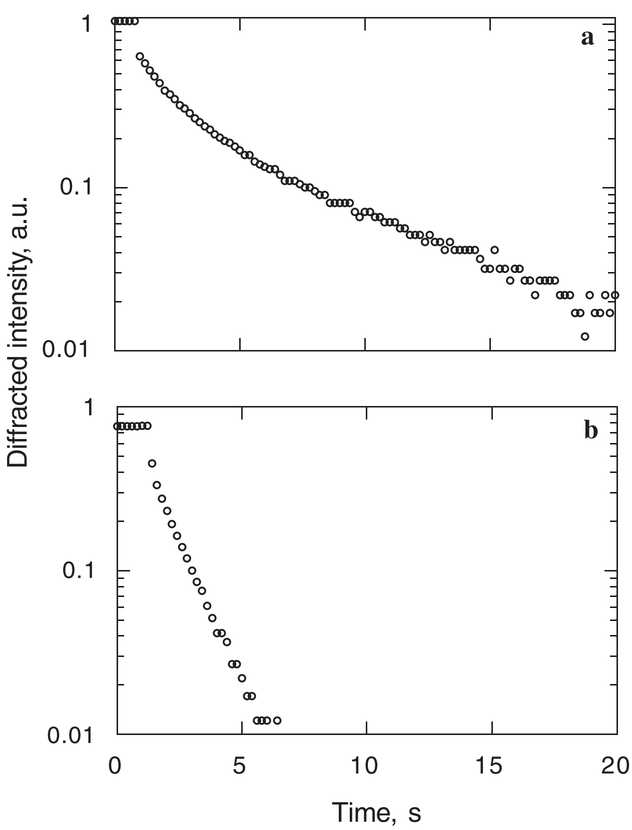

FIGURE 2 Temporal dependence of grating decay below the oscillation threshold. **a** decay with counter-propagating pump wave present, **b** decay with the beam splitter BS blocked

cess can be seen in Fig. 2a. The relaxation time is evaluated from the initial linear slope of the decay curve. It is the time when the signal drops to $(1/e)$ of its initial intensity. This time is two times smaller than the relaxation time of the refractive index because the diffracted intensity is proportional to the square of the refractive-index change Δn.

2.3 *Experimental results*

Further, we measure the oscillation intensity and the characteristic delay time as a function of the coupling strength. As a rule, the coupling strength in photorefractive crystals is independent of the 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 the recording beam I_0 and the 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

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

where $(\gamma\ell)_0$ is the initial coupling strength for $I_E = 0$. (We neglect the anisotropy of photoconductivity when writing (1) in this form.) Figure 3a shows the coupling strength dependence of the saturated oscillation intensity. It is easily seen that the oscillation intensity is increasing gradually for two orders of magnitude above the threshold value of the coupling strength $\gamma\ell \geq (\gamma\ell)_{th}$. No discontinuity is observed exactly at the threshold. This points to the soft mode of the oscillation onset from the noise level. Qualitatively this dependence corresponds to the increase of the oscillation intensity above the threshold, which can be calculated within the approach developed in [12] with the appropriate boundary conditions [13].

Figure 3b gives the plot of the reciprocal characteristic time below and above the threshold, measured as explained above. The experimental points can be fitted with a linear dependence

$$\frac{1}{\tau} \propto |\gamma\ell - (\gamma\ell)_{th}|, \tag{2}$$

separately for the downward and upward slopes. These two fits give, within a few per cent, the same value for $(\gamma\ell)_{th} \simeq 0.89(\gamma\ell)_0$, which agrees well with that measured from the coupling strength dependence of the oscillation intensity (Fig. 3a).

As described above, we use principally different techniques to measure the characteristic build-up and decay times above and below the threshold. The time measured below the threshold is close to $\tau_{\Delta n}/2$, i.e. to one-half the time of the refractive index grating decay with feedback applied. It is linearly proportional to the dielectric relaxation time of the sample for a given light intensity, but it also contains the factor depending on the overthreshold coupling strength $(\gamma\ell - \gamma\ell_{th})$.

The characteristic time of the oscillation onset above the threshold also depends linearly on the dielectric relaxation time and is a function of $(\gamma\ell - \gamma\ell_{th})$. In addition it depends

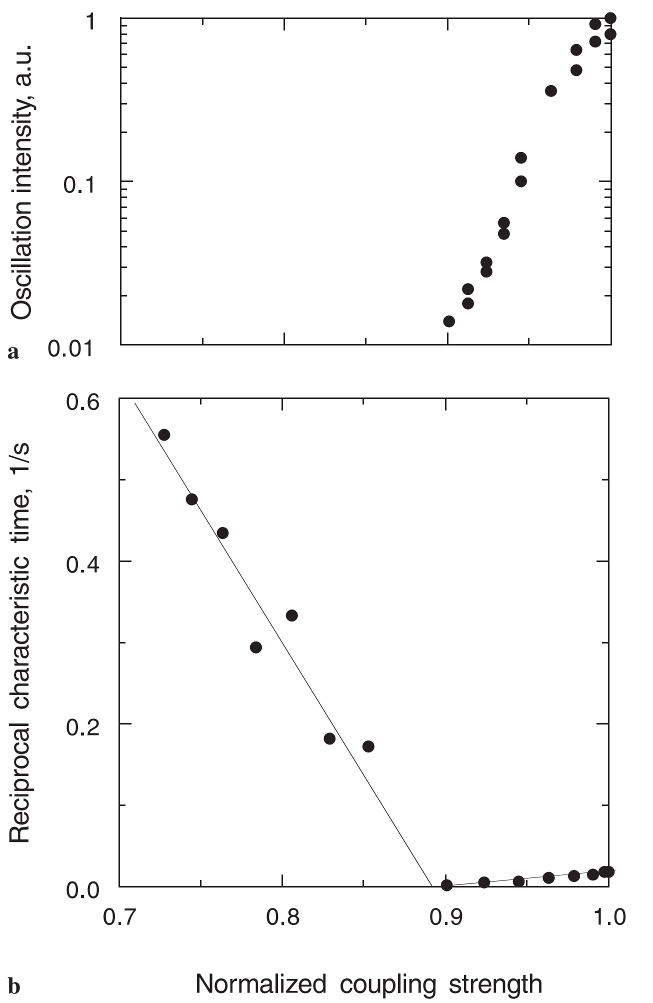

FIGURE 3 Coupling strength dependence of the oscillation intensity (**a**) and the inverse characteristic time (**b**)

strongly on the initial level of scattered light which is seeding the oscillation. The results of numerical simulation for a double phase-conjugate mirror [14] and a semilinear phase-conjugate mirror [15], which are close relatives of the considered semilinear oscillator with two pump waves, show that the delay time of the oscillation onset may vary by orders of magnitude when the initial seed amplitude is changing considerably.

The dependence in Fig. 3b therefore represents the values proportional to a "real" relaxation time but with the proportionality factors that are different for two branches, below and above the threshold. This is why one can make conclusions from this figure about the linear dependence of $1/\tau$ on $(\gamma\ell - \gamma\ell_{th})$ and about particular values of $\gamma\ell$ where $1/\tau$ diverges, but it is impossible to compare the absolute values of the relaxation time itself and of the slopes of the two branches.

3 Discussion

The oscillation onset in the coherent oscillator considered is similar to the second-order phase transition. Both the results of calculation [13] and the experimental data (Fig. 3a) show the soft mode of oscillation onset: the intensity of the oscillation wave is zero exactly at threshold and is increasing gradually till the saturation level is reached with the increasing coupling strength. Similarly to the usual lasers the oscillation intensity is increasing linearly with the over-threshold pumping, i.e. $I_{\mathrm{osc}} \propto (\gamma\ell - \gamma\ell_{\mathrm{th}})$. This means that the amplitude of the oscillation field $E_{\mathrm{osc}} \propto \sqrt{\gamma\ell - \gamma\ell_{\mathrm{th}}}$ and therefore the derivative $\mathrm{d}E_{\mathrm{osc}}/\mathrm{d}(\gamma\ell)$ becomes infinite exactly at $\gamma\ell = \gamma\ell_{\mathrm{th}}$. This justifies the choice of normalized amplitude $E_{\mathrm{osc}}/E_{\mathrm{pump}}$ as an order parameter for this system. Another option would be to consider the normalized amplitude of the self-developing refractive-index grating which couples the pump and oscillation waves, $\Delta n/\Delta n_{\mathrm{max}}$, as an order parameter. Here Δn_{max} is the ultimate refractive-index change that can be achieved at a given experimental condition (limited by the diffusion field, applied external field or inherent photovoltaic field of the sample). In fact, both possible order parameters are not independent because the amplitude of the diffracted beam is directly proportional to the amplitude of the refractive-index grating. We choose the normalized amplitude of the oscillation wave as an order parameter because it can be experimentally measured.

The analogy to the control parameter which is quite often the temperature for structural order–disorder phase transitions is in our case the inverse coupling strength, $1/\gamma\ell$. We take the reciprocal value of the coupling strength to obtain the transition into the ordered state for the decreasing control parameter (similarly to the transition into the ordered state for decreasing temperature).

The following conclusions from the analysis of steady-state oscillation confirm the second-order phase transition: (i) no discontinuity is observed in the evolution of the oscillation intensity from zero value to saturation, (ii) just above the threshold the oscillation intensity depends linearly on the overcritical coupling strength, i.e. the derivative of the order parameter exactly at the threshold is infinite. Several additional arguments supporting the analogy to the second-order phase transition will be deduced from the analysis of oscillation dynamics.

We found experimentally a power-law dependence of the reciprocal relaxation times below and above the threshold, $1/\tau \propto |\gamma\ell - \gamma\ell_{\mathrm{th}}|^{\zeta}$, with critical exponent ζ equal to unity. Such dependence is typical for the second-order phase transitions for which different parameters increase with the temperature as $|T - T_{\mathrm{c}}|^{\zeta}$, with ζ standing for the critical exponent and T_{c} being a transition temperature (Curie temperature). For example, for a ferromagnetic phase transition the temperature dependence of the magnetic susceptibility χ is described by the Curie–Weiss law [16], i.e. χ is proportional to $|T - T_0|^{-1}$, with T_0 standing for the Curie–Weiss temperature. The Curie–Weiss law is valid also for some ferroelectric crystals, for temperature dependence of the dielectric constant ϵ, piezoelectric and elasto-optic properties, etc. above the transition temperature (see e.g. [17]). Thus, the observed coupling strength dependence of τ^{-1} can be considered as an optical analogy of the Curie–Weiss law.

It should be underlined that the Curie–Weiss temperature coincides with the transition temperature for the second-order phase transitions, while for the first-order phase transitions it is different from the transition temperature [15]. 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, for both the temporal development of the scattering below the transition and the development of the oscillation above the transition. A critical slowing down below the threshold of mirror-less oscillation has been reported for photorefractive BSO ($Bi_{12}SiO_{20}$) crystals [9] and $BaTiO_3$ crystals [2]. A critical exponent equal to unity has been measured for BSO. For the temporal dynamics of some other optical processes, like e.g. switching in optically bistable étalons [18, 19], the critical exponent can be 1/2, i.e. significantly different from 1.

Summarizing, additional arguments in favour of the second-order phase transition which could be extracted from the study of oscillation dynamics are as follows: (iii) a power-law dependence of relaxation times near the threshold, (iv) the same critical value of the control parameter at which $1/\tau$ diverges for both branches, below and above the threshold, (v) coincidence of the “Curie–Weiss” temperature and the transition temperature extracted from the measurements of the order parameter.

We believe that all the arguments given above prove that the threshold of oscillation in a semilinear cavity with two counter-propagating waves can be considered as a second-order phase transition within the investigated range of parameters. At the same time one can ask how far this analogy can go. The fundamental question is what are the formal analogies in optical systems with phase transitions to thermodynamic functions and to susceptibility. Another question under discussion is in what way the fluctuations could affect the optical phase transition. Additional studies are necessary, both theoretical and experimental, to obtain clear answers to these questions.

It should be underlined that apart from fundamental interest the discussed analogy of the oscillation threshold to the phase transition leads to important practical consequences, for example for hybrid lasers [20, 21] which possess the self-pumped phase-conjugate mirrors or semilinear coherent oscillators as their inherent parts. The described critical slowing down near the oscillation threshold will affect strongly the dynamics of oscillation onset of such hybrid lasers. To design a hybrid laser with a realistically short onset time one should be aware of the dependence of the characteristic time on the coupling strength given by the empirically established Curie–Weiss law.

ACKNOWLEDGEMENTS Two of us (S.O. and O.S.) acknowledge financial support of the Ministry for Education and Science, Ukraine via Grant No. F4/356-97-20. We are grateful to M. Segev for helpful discussions and to our reviewers for their constructive criticism.

REFERENCES

1 P.L. Ramazza, S. Residori, G. Giacomelli, F.T. Arecchi: Europhys. Lett. **19**, 475 (1992)
2 D. Engin, S. Orlov, M. Segev, G. Valley, A. Yariv: Phys. Rev. Lett. **19**, 1743 (1995)
3 M. Goul'kov, O. Shinkarenko, S. Odoulov, E. Krätzig, R. Pankrath: Appl. Phys. B **72**, 187 (2001)
4 S. Odoulov, M. Goul'kov, O. Shinkarenko: Phys. Rev. Lett. **83**, 3637 (1999)
5 H. Haken: Synergetics (Springer, Berlin, Heidelberg 1978)

6 M. Harris, R. Loudon, G. Mander, J.M. Vaughan: Phys. Rev. Lett. **67**, 1743 (1991)
7 A. Yariv, D. Pepper: Opt. Lett. **1**, 16 (1977)
8 J. Feinberg, R. Hellwarth: Opt. Lett. **5**, 519 (1980)
9 D.A. Fish, T.J. Hall, A.K. Powell: Opt. Commun. **84**, 85 (1991)
10 H.J. Eichler, P. Guenter, D. Pohl: Laser-induced dynamic gratings (Springer, Berlin, Heidelberg 1986)
11 S.R. Liu, G. Indebetouw: J. Opt. Soc. Am. B **9**, 1507 (1992)
12 M. Cronin-Golomb, B. Fischer, J.O. White, A. Yariv: IEEE J. Quantum Electron. **QE-20**, 12 (1984)
13 P. Mathey, P. Jullien, S. Odoulov, O. Shinkarenko: J. Opt. Soc. B, accepted
14 D. Engin, M. Segev, S. Orlov, A. Yariv: J. Opt. Soc. Am. **11**, 1708 (1994)
15 A.A. Zozulya: IEEE J. Quantum Electron. **QE-29**, 538 (1993)
16 R. Kubo: *Statistical Mechanics* (North-Holland, Amsterdam 1965)
17 F. Iona, G. Shirane: *Ferroelectric Crystals* (Pergamon, Oxford 1962)
18 E. Garmire, J.H. Marburger, S.D. Allen, H.G. Winful: Appl. Phys. Lett. **34**, 374 (1979)
19 H.A. Al-Attar, H.A. McKenzie, W.J. Firth: J. Opt. Soc. Am. B **3**, 1157 (1986)
20 S. Odoulov, M. Soskin, A. Khizhiak: *Optical Oscillators with Degenerate Four-Wave Mixing (Dynamic Grating Lasers)* (Harwood Academic, Chur, London 1991) pp. 37–39
21 A.A. Bagan, V.B. Gerasimov, A.V. Golyanov, V.E. Ogluzdin, V.A. Sugrobov, I.L. Rubtsova, A.I. Khyzhnjak: Sov. J. Quantum Electron. **17**, 49 (1990)