# OSCILLATION DYNAMICS OF A $Sn_2P_2S_6$-BASED SEMILINEAR OPTICAL OSCILLATOR

A. SHUMELYUK, A. HRYHORASHCHUK, S. ODOULOV

Institute of Physics, Nat. Acad. of Sci. of Ukraine
*(46, Nauky Ave., Kyiv 03028, Ukraine; e-mail: shumeluk@iop.kiev.ua)*

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The unusual temporal dynamics of a semilinear optical coherent oscillator based on a tin thiohypodiphosphate ($Sn_2P_2S_6$, SPS) crystal has been described. Periodic non-harmonic oscillations of the oscillator output intensity have been studied. The pulse duration was found to depend on characteristic grating formation times and on how much the coupling strength constant exceeds its threshold value. The pulse width changes in a wide range, decreasing closer to the oscillation threshold. It has been proved experimentally that every pulse in a sequence is $\pi$-shifted in phase with respect to the previous one, while the phase of oscillations remains constant within every pulse. Such a behavior of the optical oscillator suggests its analogy with an electronic multivibrator. A competition between two species of movable charge carriers in the course of formation of the space charge grating makes up the origin of the phenomenon concerned.

## 1. Introduction

The advantages of $Sn_2P_2S_6$ photorefractive crystals over others follow from the combination of its fast response (of the order of milliseconds) and a rather high gain in the red and near-infrared spectral ranges [1, 2]. These crystals are successfully applied for the light amplification, phase conjugation, and in coherent optical oscillators [3, 4]. The coexistence of two spatial charge gratings is a rather frequent phenomenon for them. One of the gratings is formed by light-induced holes, and the other appears owing to the redistribution of thermally excited electrons [1, 2, 5]. As a result of the formation of two out-of-phase gratings possessing substantially different decay times, the spectrum of the stationary gain factor reveals two maxima shifted symmetrically with respect to the pump frequency [6]. In coherent oscillators with open cavities, this results in the formation of two components in the oscillation spectrum and in the sinusoidal temporal modulation of the oscillation intensity [7]. In this case, the frequency shift between the modes is proportional to the square root of the pump intensity [8].

In this article, we describe a new type of the optical oscillation dynamics in an open semilinear coherent oscillator with two counter-propagating pump beams. The dynamics of the oscillation beam intensity consists of a sequence of periodic non-sinusoidal pulses, with the phase of every pulse being shifted by $\pi$ with respect to that of the previous pulse. We propose a model, which explains such a new operating mode of the oscillator, and compare the results that were obtained in the framework of this model with experimental data. The gain factors of two out-of-phase gratings, calculated making use of the threshold oscillation quantities, coincide well with the relevant values measured directly in two-beam interaction experiments.

## 2. Experimental Studies

A sample of nominally pure tin thiohypodiphosphate, which had been grown by the gas-transport technique [9] at the Institute for Solid State Physics and Chemistry of the Uzhhorod National University, served as a nonlinear crystal. The $9 \times 9 \times 4.5$-mm$^3$ sample was cut along the crystallographic axes, and its faces normal to the $z$-axis were polished. The sample belonged to crystals of the I type [10] with the pronounced influence of charge carriers of both types on the formation of a spatial charge field. The crystal had preliminary been polarized by applying a voltage of about 1 kV/cm when cooling from 90°C down to room temperature.

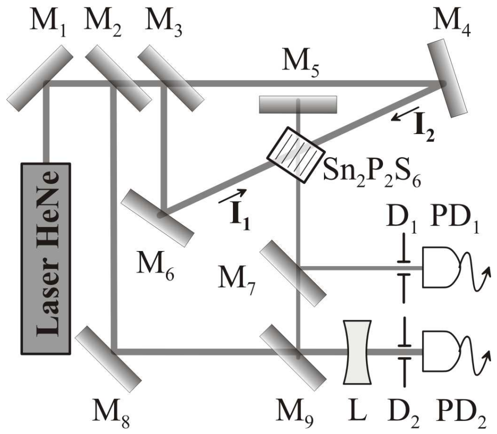


Fig. 1. Experimental set-up: mirrors ($M_1$ to $M_9$), diaphragms ($D_1$ and $D_2$), photo diodes ($PD_1$ and $PD_2$), and lens ($L$)

A He-Ne laser, which is single-mode by the transverse-mode index and possesses the output beam power of 40 mW, was used as the source of radiation. The beams were polarized in the plane of their convergence (Fig. 1). The crystal was irradiated by the counter-propagating beams reflected from mirrors $M_4$ and $M_6$. In such a geometry with two counter-propagating beams, the $Sn_2P_2S_6$ crystal operates in the mode of a phase conjugate mirror and, together with an ordinary mirror $M_5$ possessing the reflection factor $R$, can form a cavity. The oscillation arises, if losses in the cavity are compensated by the reflection from the phase conjugate mirror with the coefficient $R_{pc}$ that exceeds unity. This means that the condition for the oscillation to emerge can be written down as

$$R_{\rm pc}R = 1 \qquad (1)$$

(see, e.g., work [11]). The sample was oriented in such a manner that its $X$ axis was parallel to the vector of the self-developing photorefractive grating. Such a geometry enables one to take advantage of the largest electrooptical factor $r_{111}$ when recording the grating. Mirrors $M_2$, $M_8$, and $M_9$, as well as photo diode $PD_2$, were used for checking the position of the interference pattern that was created by the oscillation and pump beams. The oscillation intensity was measured by photodetector $PD_1$.

The oscillation dynamics of such a device is shown in Fig. 2,*a*. Every time when the oscillation terminated between two consecutive pulses, the interference pattern underwent the shifting by a half-period; i.e. there occurred the inversion of the contrast of interference stripes which were formed when the generated and pump beams had been converged at a small angle. Photo diode

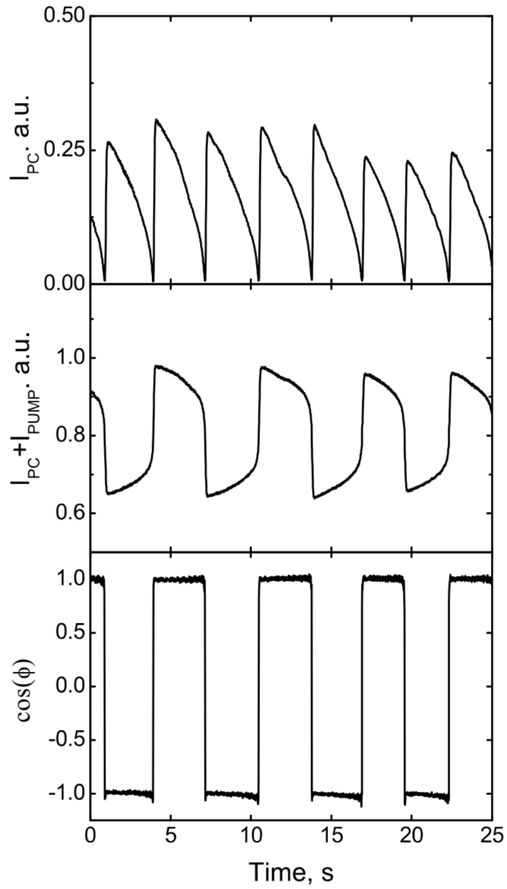


Fig. 2. Time dependences of the oscillation beam intensity recorded by $PD_1$ ($a$), the intensity of the interference pattern recorded by $PD_2$ ($b$), and the calculated phase of the oscillation beam ($c$)

$PD_2$ was used for the registration. The arrangement of interference stripes did not change during a single pulse. In Fig. 2,*b*, the dependence of the oscillation intensity variation on the imposing of the pump beam is depicted. It should be noted that the measurements, presented in Figs. 2,*a* and *b*, were carried out simultaneously. While comparing the experimental dependences shown in those figures, it becomes evident that they are periodic, and one oscillation period consists of two consecutive pulses. This is possible, if the phase of the generated beam varies. Possessing the temporal dependences of the optical oscillation both with and without imposing the pump beam and carrying out elementary calculations, one can plot the temporal dependence of the phase of oscillation beam (Fig. 2,*c*). As one can see, this dependence is not sinusoidal. Fig. 3 demonstrates the dependences of the oscillation beam intensity ($a$) and the pulse duration ($b$) on the ratio between the intensities

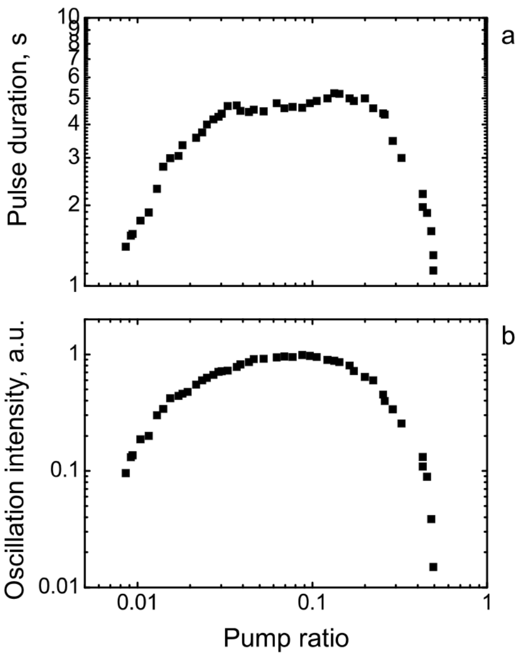


Fig. 3. Dependences of the pulse duration ($a$) and the oscillation beam intensity ($b$) on the ratio between the intensities of pump beams

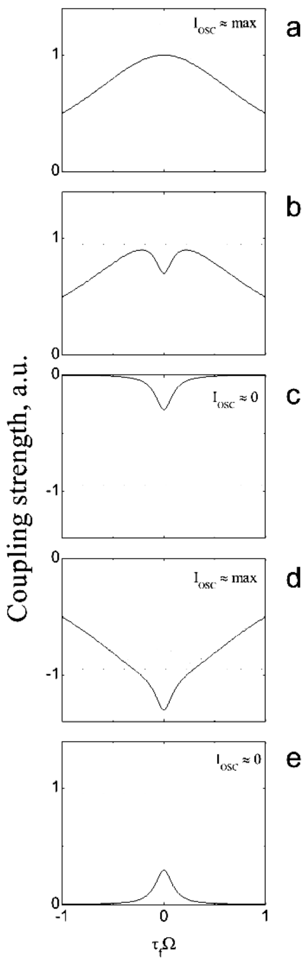


Fig. 4. Gain spectrum at various stages of the oscillation process: ($a$) the oscillation start, (b) the end of the first pulse, ($c$ to $e$) the influence of the slow grating that results in the formation of a new pulse; solid curve $d$ and dotted curve $a$ correspond to maximal gains

of the pump beams. At certain threshold values, the beam intensity and the pulse duration diminish abruptly.

## 3. Description of the Model

The oscillation dynamics can be explained as follows. As the illumination of the sample began at the moment $t = 0$, the fast space-charge grating started to form owing to the redistribution of light-induced holes. Let us assume that the instant value of the coupling strength constant $\Gamma(t)l = 2\gamma(t)l$ is proportional to the induced variation of the refraction index $\Delta n(t)$ at the same moment. Then, the gain spectrum has a Lorentz-like shape, whose halfwidth is reciprocal to the characteristic decay time of the fast grating $\tau_f$. After several intervals with the duration $\tau_f$ each, the gain increases to its maximal value (Fig. 4,$a$) and begins to exceed, to some extent, the threshold value $\Gamma l_{\text{th}}$; the latter is marked by the dotted line. The oscillation starts, and the phase of the oscillation beam may be arbitrary, as it occurs in all photorefractive coherent generators with an open cavity. This phase is determined by the phase of the initial fast grating which is governed by fluctuations.

When the fast grating has already been formed, the gradual development of the slow one is still taking place, because the characteristic development time of the latter $\tau_s$ is much longer. The slow grating is $\pi$-shifted in phase with respect to the fast one, so that its gain factor is negative. The corresponding gain spectrum is also Lorentz-like, but its halfwidth is much narrower and equals $\tau_s^{-1}$ (in Fig. 4, for the sake of illustration, we used $\tau_f$ and $\tau_s$ with a relatively small difference between them). The total gain spectrum has a dip at the zero-difference frequency; the gain maximum decreases and becomes, in due time, lower than the threshold value (Fig. 4,$b$). At this moment, the oscillation stops within the time interval equal to several time constants of the fast grating decay $\tau_f$. During this period, the slow grating remains almost invariable, and the corresponding gain spectrum is shown in Fig. 4,$c$. The pump beam diffracts from the slow grating into the semilinear cavity: the phase of this diffracted beam is $\pi$-shifted with respect to the phase of the previous pulse in the oscillation beam, because the slow grating is out of phase with respect to the fast one.

The diffracted wave with a precisely known phase forces to "start" the development of a new fast grating, the latter being $\pi$-shifted with respect to the initial fast one. Thus, at this stage, two gratings, fast and slow, are *in phase* and boost each other (continuous line in Fig. 4,*d*). The total gain reaches its maximal value.

The further development leads to the saturation of the fast grating, and electrons start to move in order to compensate the emerged field of a spatial charge. In other words, the formation of a new out-of-phase slow grating takes place. Step by step, the additional peak in the gain spectrum is transformed into a dip (dotted line in Fig. 4,*d*), and the oscillation stops again. But the slow grating still remains and initiates the development of a next oscillation pulse (Fig. 4,*d*). The only difference between this new pulse and the previous one is a somewhat higher gain factor, as is illustrated by the dotted line in Fig. 4,*a*. Afterwards, the cycles repeat. We note that a single pulsation period includes two pulses of the intensity with two different phases, 0 and $\pi$.

Therefore, the saturated oscillation consists of two modes which differ from each other only by the phase of the oscillation beam. When one of the modes is being oscillated, the other mode is locked, the gain factor for the former reduces, and the gain factor for the next oscillation mode increases with the same rate. Such a behavior of the system is similar to the self-excitation in an Abraham—Bloch multivibrator which is composed of two electronic amplifiers, the inputs and outputs of which are cross-coupled.

In the model described, the pulse duration depends on the characteristic time of the slow grating decay and the over-threshold value of the gain factor. The closer the total gain factor is to the threshold value, the shorter time period is necessary for the out-of-phase grating to develop and to throw the system down below the threshold limit. For this reason, the pulses become shorter if the over-threshold value of the gain factor decreases. Such an operation mode substantially differs from the already known mode in a coherent ring oscillator [7], where the oscillation period tends to infinity when approaching the oscillation threshold.

## 4. Numerical Estimates

The phase conjugate reflectivity $R_{\rm pc}$ depends on the coupling strength $\gamma l$ and the pump intensity ratio $r$ [11]:

$$R_{\rm pc} = \frac{\sinh^2\left(\frac{\Gamma l}{4}\right)}{\cosh^2\left(\frac{\Gamma l}{4} + \frac{\ln r}{2}\right)} . \tag{2}$$

This allows some important parameters of the model to be determined by comparing experimental and calculated data.

Fig. 3,*a* allows the optimum pump intensity ratio to be determined: $r_{\rm opt} = 0.07$. This value may logically be assumed to correspond to the maximum phase conjugate reflectivity (2) at the coupling strength that ensures the sample, i.e. $\Gamma l = 2\ln(r_{\rm opt})$. In this case, the coupling strength of the fast grating $(\Gamma l)_{\rm fast}$ is about $-5.3$.

Making use of the threshold values of $r$, at which the oscillation terminates ($r_1 = 0.008$ and $r_2 = 0.5$), one can calculate the value of $(\Gamma l)_{\rm fast}$ independently. The oscillation thresholds correspond to the same value $R_{pc} = R^{-1}$ (see Eq. (1)). Therefore, we can write down the equality $2\ln(r_1) - \Gamma l = \Gamma l - 2\ln(r_2)$ and obtain the ultimate value of the coupling strength constant $(\Gamma l)_{\rm fast} \approx -5.5$, which differs from the preliminarily calculated value by less than 5%. Moreover, using Eq. (2), we can obtain the value of the effective reflection coefficient for the ordinary mirror in the cavity, which includes all possible losses: $R = 0.78$.

All the aforesaid calculations were carried out for the fast component, because it plays a crucial role in the oscillation and is responsible for the oscillation thresholds. The coupling strength constant for the slow component can be evaluated, if one knows the duration of pulses in the coherent cavity. Neglecting the difference between the Debye screening lengths for charge carriers of different types, it is possible to write down the equation describing the dynamics of a slow grating in the form [6]

$$(\Gamma l)_{\rm slow}^{\rm max} = (\Gamma l)_{\rm slow}[1 - \exp(-t/\tau_{\rm slow})] =$$

$$= (\Gamma l)_{\rm slow}(t/\tau_{\rm slow}), \tag{3}$$

where $(\Gamma l)_{\rm slow}^{\rm max}$ is the coupling strength constant of the slow grating, at which the oscillation terminates; in other words, it is the largest possible value, to which the slow grating develops in experiment.

Taking the pulse duration $\Delta t$ for time $t$, we obtain $(\Gamma l)_{\rm slow}^{\rm max} = (\Gamma l)_{\rm fast} \times (t/\tau_{\rm slow})$. Provided $\Delta t = 5$ s, $(\Gamma l)_{\rm fast} = 5.4$ and $\tau_{\rm slow} = 12$ s, the coupling strength constant can reach the value $(\Gamma l)_{\rm slow}^{\rm max} = 2.25$. In general,

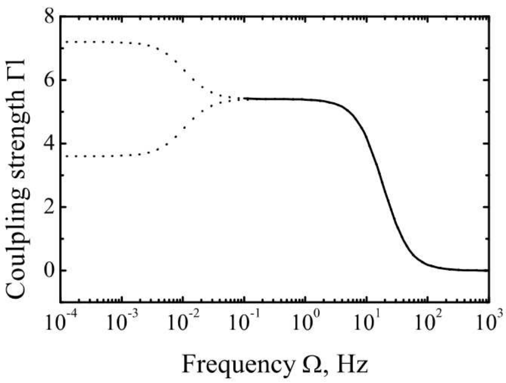


Fig. 5. Ultimate value of the coupling strength constant at the competitive interaction of gratings

the value of $(\Gamma l)_{\rm slow}$ depends on a specific experiment, and, in the framework of our approximations, $(\Gamma l)_{\rm slow} \equiv (\Gamma l)_{\rm fast}$.

In Fig. 5, the gain spectrum

$$\Gamma l\,(\Omega) = \frac{(\Gamma l)_{\rm fast}}{1+(\Omega\tau_{\rm fast})^2} \pm \frac{(\Gamma l)_{\rm slow}}{1+(\Omega\tau_{\rm slow})^2}, \qquad (4)$$

is depicted, where $\Omega$ is the frequency shift between the oscillation and pump waves. It is evident that, in the degenerate case $\Omega \to 0$, the total gain is switched between two values which differ from each other by the sign $\pm$ in expression (4).

In the same figure, the horizontal dotted line marks the threshold value of the coupling strength constant which exceeds, to some extent, the total gain in the case of the out-of-phase summation of two gratings. Thus, the model proposed is confirmed by calculations. Every time, when the formation of the out-of-phase grating reduces $\Gamma l$ to its threshold value, the oscillation terminates, and the fast grating becomes erased within a time interval of about 40 ms. At the same time, the much more inertial slow grating still exists and leads to the development of a new fast grating, which is in phase with the slow one. In this case, the gain effects of both gratings boost each other, and this situation corresponds to the top branch of the plot in Fig. 5. After the fast grating having saturated, a new slow grating, which is $\pi$-shifted with respect to the previous one, again reduces the gain to the bottom branch, which is below the oscillation threshold. The oscillation stops, and a new pulse starts.

The described periodic pulsations obviously belong to interactions which are not degenerate by frequency. In addition, it is important to emphasize that this operating mode is qualitatively new and cannot be reduced to the case of the already known sinusoidal modulation of the intensity, when only two symmetric oscillation frequencies arise in the spectrum [6]. It can compete with the known mode [7], because the corresponding coupling strength constant is larger than that in the stationary mode of the two-frequency oscillation, at least during the halfperiod of the pulse.

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ДИНАМІКА ГЕНЕРАЦІЇ НАПІВЛІНІЙНОГО ОПТИЧНОГО ГЕНЕРАТОРА НА КРИСТАЛІ $Sn_2P_2S_6$

*О. Шумелюк, А. Григоращук, С. Одулов*

Р е з ю м е

Описано незвичну часову динаміку напівлінійного оптичного когерентного генератора, що базується на виродженій за частотою чотирихвильовій нелінійній взаємодії в кристалі тіогіподифосфату олова $Sn_2P_2S_6$, SPS. Досліджено періодичні негармонічні коливання вихідної інтенсивності в генераторі. Встановлено, що тривалість імпульсу залежить від характеристичних часів стирання ґраток та від перевищення константою взаємодії її порогового значення. Тривалість імпульсів лежить в широких межах та зменшується з наближенням до порога генерації. Експериментально встановлено, що кожний наступний імпульс за фазою зміщений на $\pi$ відносно попереднього, в той час як в межах одного імпульсу фаза є незмінною. Така поведінка оптичного генератора вказує на аналогію з електронним мультивібратором. В основі цього явища лежить конкуренція носіїв заряду різного знака (електронів та дірок) при формуванні ґраток просторового заряду.