Hexagon formation in photorefractive crystals as mirrorless coherent oscillation

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The spontaneous appearance of hexagon patterns in photorefractive crystals is treated as mirrorless coherent four-wave mixing oscillation. It is shown experimentally that the system has a well-defined coupling-strength threshold and behaves as other coherent optical oscillators near the threshold. A simple relation for the angle of the hexagon sideband as a function of the distance between the end mirror and the sample is derived and verified experimentally. © 1998 Optical Society of America [S0740-3224(98)01207-7]

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1. INTRODUCTION

Since the first observation of photorefractive hexagons by Honda,[1] the formation of regular patterns in nonlinear wave mixing in these media has attracted considerable attention, both experimentally[2–10] and theoretically.[11–14] The appearance of other patterns such as rolls,[1] lines,[7] and squares,[8,9] were reported, the conditions necessary for pattern formation were established, and the factors that define the angular structures were found.[4,11–13] It has been shown that formation of hexagons and other regular structures is related to recording of the reflection gratings by the two counterpropagating pump waves and by the sidebands with the pump waves.

In practically all the studies mentioned above the appearance of the regular patterns was considered the result of spatial instability of the counterpropagating pump waves. We emphasize here that the sidebands of hexagons and other regular spatial structures are typical examples of coherent mirrorless oscillation.[15,16] This approach does not contradict the previous one, because mirrorless oscillation is in fact a spatial variation of absolute instability.[17,18] At the same time, it allows us to reveal some new aspects of this phenomenon that underscore the similarity to the usual lasers and other four-wave mixing oscillators. Ideologically our approach is close to that of Refs. 12, 14, and 18 in which hexagon formation was considered a kind of parametric four-wave mixing process.

2. MODELS FOR PATTERN FORMATION

Assume that a hexagon is already excited; then in a certain plane that contains two pump waves we may have, in addition, four sidebands as shown in Fig. 1A. Sideband γ (with wave vector $\mathbf{k}_\gamma$) records with pump wave β (with wave vector $\mathbf{k}_\beta$) a grating (with grating vector $\mathbf{K}_1$). The same grating $\mathbf{K}_1$ is also recorded by the second pump, α (wave vector $\mathbf{k}_\alpha$), with sideband δ' (wave vector $\mathbf{k}_{\delta'}$), as shown in Fig. 1B. In a similar way the grating with grating vector $\mathbf{K}_2$ is recorded by pairs of waves α, δ, and β, γ' (Fig. 1C). Apart from the gratings with grating vectors $\mathbf{K}_1$ and $\mathbf{K}_2$ the principal grating with grating vector $\mathbf{K}_0 = \mathbf{k}_\alpha - \mathbf{k}_\beta$ is recorded by counterpropagating pump waves α and β (Fig. 1D).

By considering wave γ as a signal wave, we can see from Fig. 1B that wave δ' corresponds to the phase-conjugate wave that appears as the result of standard backward wave-four wave mixing.[15] In a similar way waves δ and γ' make a pair, signal wave–conjugate wave, for the four-wave mixing process shown in Fig. 1C.

It is well known that backward-wave four-wave mixing may result in mirrorless oscillation if the coupling strength Γl (Γ is the coupling constant and l is the sample thickness) is larger than a certain threshold value. The necessary condition for this type of oscillation is the presence of local response in photorefractive crystal used, either inherent or that which is due to the frequency shift of the sidebands with respect to the pump wave in the crystal with purely 90° out-of-phase gratings. This conclusion is valid for mixing with predominant transmission gratings as well as for mixing when only reflection gratings are recorded (see, e.g., Refs. 16 and 17). Therefore one could expect the appearance of the frequency-degenerate patterns in photorefractive crystals with drift or photovoltaic charge transport (rings[19] and hexagons[20] were observed in $LiNbO_3$:Fe with the strong photovoltaic effect) or of nondegenerate hexagons in crystals with the diffusion charge transport.

This was the main qualitative result of the theories.[11,12] In fact, the distinction of the mixing process in hexagon formation[11,12] from the usual backward-wave four-wave mixing[16] is in the coupling of sidebands γ, δ and γ', δ' because of the diffraction from the principal grating $\mathbf{K}_0$ recorded by counterpropagating pump waves

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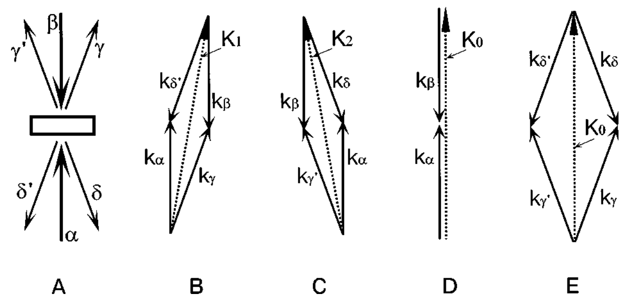

Fig. 1. A, Orientation of the incident and generated light beams in hexagon formation. B, C, Elementary backward-wave four-wave mixing processes, and D, E, diffraction from the principal grating. The wave vectors **k** are shown as the solid arrows in B–E; the grating vectors are shown by dotted lines.

(Fig. 1E). This diffraction can be exactly Bragg matched if the geometrical dephasing is compensated for by the nonlinear dephasing,[11,12] but it may also be nearly Bragg-matched because of rather soft angular selectivity of the reflection grating.[1] This is why a final result of calculations[11,12] is more complicated than that in Ref. 16; in particular, these theories predict a certain angle between the sidebands and the pump waves (whereas the direction of four-wave mixing mirrorless oscillation is not restricted for the equal intensities of the pump waves).

The next important step in theory was reported in Ref. 13, where Honda and Banerjee revealed the additional coupling process for symmetric sidebands, which is related to the transverse intensity modulation inside the sample (owing to the interference of the coherent pump and the copropagating sidebands). As a result, the amplitude of the reflection grating appears to be transversally modulated and the photons from, e.g., pump α are diffracted into both sidebands, δ and δ'. The most important result is that this theory predicts the finite threshold for the excitation of frequency-degenerate sidebands also for a photorefractive crystal with a purely nonlocal response.[13] With the effect of transverse modulation taken into account, the threshold conditions for hexagon generation become softer also for the photorefractive crystals with the local response.[14]

All the theories mentioned above do not predict the total number of sidebands in the three-dimensional case. In the experiment only one pair of sidebands can be excited near each of two pump waves (rolls), four (square) or six (hexagon) sidebands may appear, or the sidebands can form a homogeneous ring. We are aware of no previous reports of the excitation of flowerlike patterns that are known, e.g., from nonlinear mixing of counterpropagating light waves in gases,[21,22] but there is no clear restriction for observation of these more-complicated patterns in photorefractive crystals. We believe that the two-dimensional analysis can be applied for any pattern with an even number of petals (sidebands), as well as for the spatially degenerate case in which the ring is generated.

Summarizing this short overview of the existing models, we conclude that all of them predict the threshold of the sideband excitation. In spite of the fact that different numbers of light beams are coupled together in different models, four-wave mixing is the basic interaction for all of them. Below we consider the manifestation of the threshold in hexagon formation for $BaTiO_3$: Co.

The threshold condition for optical oscillators always includes the amplitude condition and the phase condition of the self-excitation. The amplitude condition imposes a certain restriction on the one-path gain in the amplifying medium (for an oscillator with closed optical cavities, e.g., one-path gain should overcome all losses in the cavity). The phase condition reflects the requirement of positive feedback in the optical oscillator; it defines the frequencies and the spatial structures of the oscillation modes. In Section 3 we present the results of our experiments, which show how these conditions are satisfied in hexagon oscillation.

3. EXPERIMENT

Hexagon formation in a $BaTiO_3$:Co sample was studied experimentally in the geometry proposed and implemented previously in Ref. 4. A 1-mm-thick Z-cut sample of $BaTiO_3$:Co is illuminated by the light beam from a single-mode single-frequency Ar^+ laser ($\lambda = 0.515\ \mu$m; Fig. 2). The incident beam is focused by an objective with focal length $F = 45$ cm onto the plane behind the sample, where a highly reflecting flat mirror is placed (lenses with other focal lengths, e.g., with $F = 15$ cm and $F = 70$ cm, were also used, but below we describe the experiments performed with the 45-cm focal-length objective). The mirror is adjusted to reflect the incident wave exactly in a backward direction; the reflected wave is close to a phase-conjugate wave with respect to the incident one (convergent incident and divergent reflected spherical waves). This mirror is mounted upon a micrometer stage, which allows for its plane-parallel displacement in a range of few centimeters.

The intensity of the incident wave is controlled with a variable beam attenuator, and its polarization can be changed by $\lambda/4$ and a polarizer. Beam splitter BS1 is used to reflect part of the backreflected radiation to the

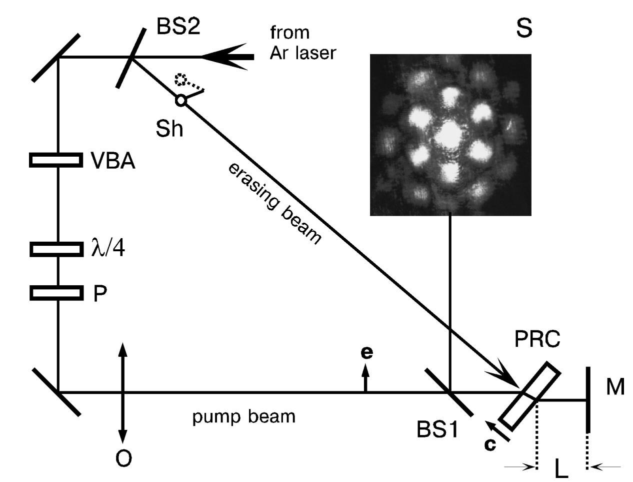

Fig. 2. Schematic of the experimental setup: VBA, variable beam attenuator; λ/4, quarter-wave plate; P, O, objective; BS1, BS2, beam splitters; PRC, $BaTiO_3$:Co sample; M, highly reflecting mirror; S, screen. Shutter Sh is used to open the erasing beam. The vectors **e** and **c** define the polarization of the pump wave and the orientation of the crystal polar axis, respectively. In some experiments, instead of a screen a high-pass spatial filter and a lens focusing the radiation of six hexagon spots to the photodetector (not shown in the figure) are installed.

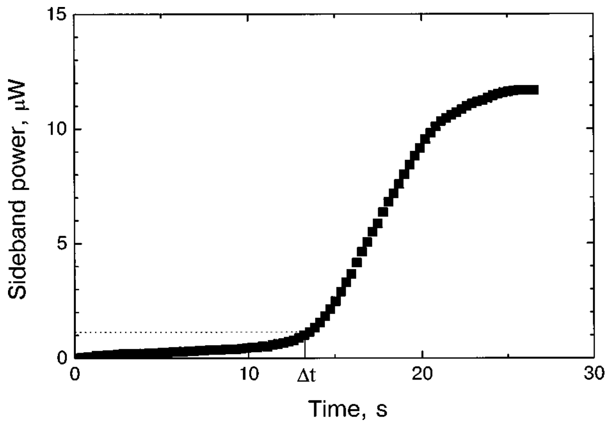

Fig. 3. Temporal development of the sideband intensity. At $t = 0$ the pump waves start to illuminate the sample in which all photorefractive gratings were previously erased by an additional coherent light wave. Dashed line, 10% level from the saturation intensity of the hexagon sideband; Δt, delay time of the oscillation switch-on.

screen for hexagon observation. Beam splitter BS2 reflects an additional light beam from the same Ar^+ laser, at oblique incidence to the sample; this beam serves for optical erasure of the recorded photorefractive gratings. Usually this beam is stopped by a shutter.

With the extraordinary pump wave the self-excitation of a hexagon (like that shown in the inset of Fig. 2) was achieved by careful adjustment of the backreflecting mirror. The intensity of the pump wave was not critical for hexagon generation; the intensity of the sideband increased linearly with the growth of the pump intensity. The ratio of intensity of the sideband to that of counterpropagating pump wave was approximately 3×10^{-4}.

The dynamics of the hexagon buildup are shown in Fig. 3. One can see a typical behavior for optical oscillators[23–25] with two different stages: a relatively long period of linear development when the intensity is increasing slowly and a sharp nonlinear increase of intensity followed by the saturation of the output intensity. We measure the oscillation switch-on time as the time interval between the beginning of the exposure and the moment when the sideband intensity reaches 10% of its steady-state value.

The *amplitude condition of oscillation* consists of the requirement that photorefractive gain overcome the losses of the system. Many possible ways to control the photorefractive gain are known. We choose the one that exploits the dependence of the gain on the polarization of the interacting waves.

The sample of $BaTiO_3$:Co is tilted nearly to $\theta = 35° \ldots 45°$ in the plane of the optical table, as shown in Fig. 2. Therefore the incident wave with the arbitrary polarization excites two waves, ordinary and extraordinary, inside the crystal. Because of the considerable birefringence the grating vectors in the tilted sample are quite different for the reflection gratings recorded by two ordinary and by two extraordinary waves. Thus, because of the severe Bragg-matching condition of the reflection gratings, a grating recorded by the ordinary waves cannot be read out by the extraordinary waves, and vice versa.

Owing to a very large electro-optic tensor component in $BaTiO_3$, r_{42}, the gain factor for mixing of the extraordinary waves is larger for this orientation than that for mixing of the ordinary waves. The oscillation wave will appear, meeting the requirement of minimum loss and maximum gain; i.e., one can expect oscillation of the extraordinary wave only. This result is in agreement with the published data,[4,6–10] in which both the pump waves and the sidebands were polarized extraordinarily, and in the present experiment we verify this by rotating the polarization of the pump waves. Figure 4 shows that within the experimental error the polarization angle of the sidebands ψ_h remains the same when polarization of the pump wave ψ_p changes more than 10°.

When the pump polarization angle is different from $\psi_p = 0$ the Γ factor diminishes (the ordinarily polarized component of pump wave does not contribute to the amplification process but still contributes to photoconductivity, thus reducing Γ for mixing of the extraordinary waves). By diminishing Γ in such a way one can approach the threshold of oscillation and, going further, completely suppress the oscillation. Figure 5 shows the dependence of the polarization angle on the intensity of the hexagon sideband and the characteristic delay time of oscillation switch-on. Note the sharp decrease of the intensity and its complete disappearance below $\psi_p = -6°$ and above $\psi_p = +6°$. As for other coherent optical oscillators,[23–25] the characteristic time of the oscillation buildup increases considerably in the vicinity of the threshold values.

The *phase condition of oscillation* consists of the requirement that the electric field of the light wave reproduce itself after one round trip of the cavity. There is no closed optical cavity in the case of optical hexagon formation that we are considering here. The phase condition is met here because of certain restrictions imposed on the phase difference of all six interacting waves propagating in the same plane. (Let us recall, e.g., the relation $\varphi_3 = -\varphi_4 + \varphi_1 + \varphi_2 +$ constant for the phases of phase conjugate φ_3, signal φ_4, and pump $\varphi_{1,2}$ waves in standard backward-wave four-wave mixing[16] in a photorefractive crystal with the diffusion nonlinearity or similar relations for the more complicated parametric mixing processes described, e.g., in Ref. 18).

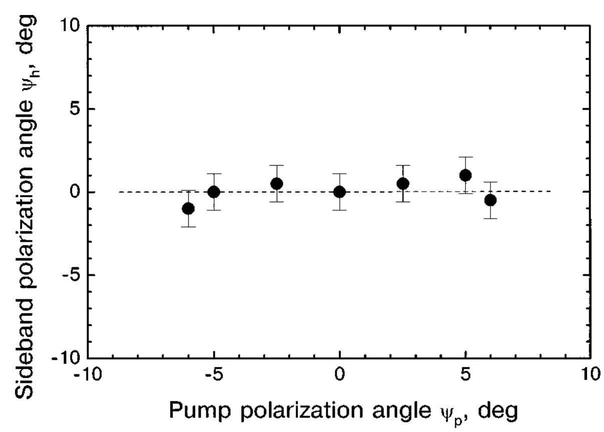

Fig. 4. Polarization angle of hexagon sidebands ψ_h versus polarization angle of incident pump wave ψ_p.

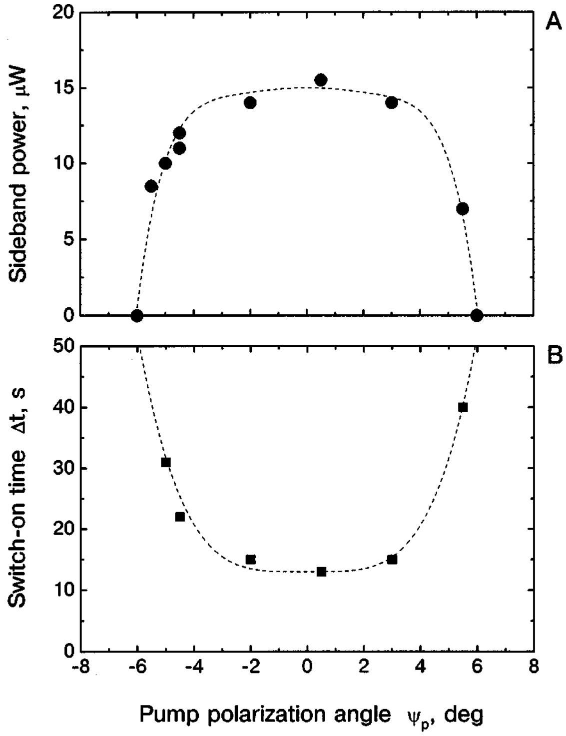

Fig. 5. A, Hexagon sideband intensity, and B, oscillation switch-on time versus polarization angle ψ_p of the incident pump wave. Dashed lines are guides for the eye.

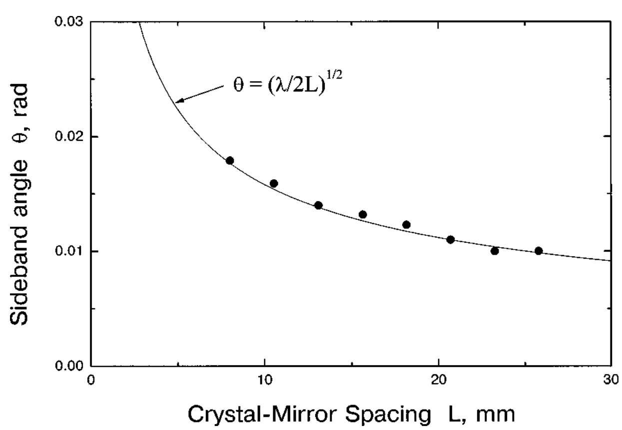

Fig. 6. Angle θ between the hexagon sideband and the pump beam versus the distance L between the sample and the backreflecting mirror. The solid curve is the best fit to relation (2) with $N = 1$.

One particular manifestation of the conditions for phase oscillation for hexagons consists in the requirement of self-reproduction of the transverse intensity distribution near the output face of the photorefractive sample. As has been already mentioned, the periodic transverse intensity distribution is of primary importance in hexagon formation[13] because it modulates the diffraction efficiency of the reflection grating. To ensure the best conditions for reflection-grating recording the requirement is imposed that the transverse intensity distribution in the light wave that is reflected by the mirror inside the sample be exactly the same as that in the wave traveling in the direction of the mirror.

It is well known that periodic amplitude or phase transparencies reproduce themselves at a certain distances when they are illuminated by a coherent light wave (Talbot self-reproduction). For an amplitude transparency (we ignore a possible minor contribution of the phase transmission grating recording) the distance between a transparency and its Nth sharp identical image is

$$L_N = \Lambda^2 N/\lambda, \tag{1}$$

where λ is the wavelength of light, Λ is the spacing of a periodic structure, and N is an integer.

Taking into account that the angle of diffraction θ from the periodic structure is $\theta = \lambda/\Lambda$, one can easily get the relation for the angular position θ of the sideband:

$$\theta \approx (\lambda N/2L)^{1/2}, \tag{2}$$

where L is now the distance between the sample and the mirror; it is equal to half of the reproduction distance, $L = L_N/2$.

Both θ and Λ are the self-adjustable parameters for the process of pattern formation considered here; they are not imposed by any external factors (such as cavity mirror position and mirror alignment for the oscillators with the external cavity). This is why for the arbitrary spacing L between the sample and backreflecting mirror one should expect the appearance of the sidebands at an angle that meets the condition of relation (2).

Figure 6 represents the measured dependence of the angle between the pump wave and the hexagon sideband as a function of the spacing between the end mirror and the sample. The solid curve shows the best fit to the dependence calculated for the first self-reproduction plane ($N = 1$). The only fitting parameter in this comparison is a small constant correction factor Δ for the distance L, because L cannot be measured exactly for the tilted sample. It should be kept in mind that the absolute value of Δ for several measurements was always much smaller than the self-reproduction distance $2L$ ($\Delta \approx 1 \ldots 2$ mm, whereas $2L \approx 20 \ldots 40$ mm). Therefore the agreement between the measured and the calculated data is quite reasonable.

Relations similar to relation (2) were also derived in Refs. 11 and 13 from other considerations; however, to our knowledge, they were not verified experimentally.

4. CONCLUSIONS

The appearance of the symmetrical sidebands (forming the rolls, squares, hexagons, or concentric rings) when two counterpropagating waves are recording the reflection grating in a photorefractive crystal can be considered mirrorless coherent oscillation. The threshold properties were studied experimentally. It was shown that the intensity of the sidebands gradually decreases to zero while the switch-on time of the oscillation is increases drastically as the coupling strength diminishes to the threshold value. As for other optical oscillators, the output radiation always corresponds to modes with the highest gain: The polarization of the hexagon sidebands, e.g., is purely extraordinary, whatever polarization of the pump waves and angular separation of the sidebands from the pump wave are imposed by the condition of self-reproduction of the optical field in the sample after reflection from the end mirror.

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