OPTICAL FORERUNNERS IN CRYSTALS WITH PHOTOREFRACTIVE DYNAMIC GRATINGS

O.M. SHUMELYUK, A.I. HRYHORASHCHUK, S.G. ODOULOV

Institute of Physics, Nat. Acad. of Sci. of Ukraine
(46, Nauky Ave., Kyiv 03650, Ukraine; e-mail: odoulov@iop.kiev.ua)

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The slowing-down of light pulses which is due to the photorefractive wave coupling is accompanied by the fast propagation of the short initial part of a pulse that can be considered as a forerunner (precursor). In accordance with the predictions of Brillouin and Sommerfeld, the forerunners are not affected by dispersion; it is shown, e.g., that they can possess a richer spectral content than the main delayed pulse, and their polarization must not necessarily fit the polarization eigenmode of the crystal with a space-charge grating.

When analyzing the propagation of light pulses in a medium with dispersion, Sommerfeld introduced a notion of forerunner (precursor) [1]. He claimed that the initial front of a truncated light pulse is propagating in a dense medium as in free space, i.e., with the velocity of light in vacuum. The reason for such anomalous propagation is simple: As any other inertial system, the ensemble of atoms-oscillators needs a certain time for the atomic motion to be started. If the incident pulse is shorter or comparable to the build-up time of the atomic motion, the medium does not practically contribute to the output field, its refractive index is equal to unity, $n = 1$, and there is no dispersion, $dn/d\omega = 0$. The phase and group velocities for a so short pulse are therefore equal to the velocity of light in vacuum, $v_{\rm ph} = v_{\rm gr} = c$. The numerical estimates [2] based on parameters adopted by Sommerfeld [1] showed that the duration of the precursor is about 1.7×10^{-22} s making nearly hopeless its experimental observation.

Considering the propagation in a medium consisting of Lorentz oscillators, Brillouin predicted the second precursor [1] which is slower and is running with the velocity c/n even in the vicinity of a resonance where the group velocity decreases.

Both, Sommerfeld's forerunners and Brillouin's forerunners were observed with microwaves [3, 4] and with acoustic waves in fluids [5]. In particular in [5], the forerunners have been studied in a one-dimensional photonic crystal consisting of a periodic sequence of polycarbonate sheets. The forerunners have also been predicted in other dispersive media such as biological [6] or viscoelastic [7] ones. Brillouin's forerunners were successfully detected in the experiment on the propagation of sub-picosecond optical pulses ($\simeq 700$ fs) in GaAs near the exciton resonance [8]. Recently, an anomalous extinction has been reported for Brillouin's forerunners excited with 100-fs light pulses in water [9].

In last two decades, a much attention was paid to the propagation of light pulses in media with artificially created dispersions, mainly in relation to the light pulse slowing-down (the group velocity is much smaller than the phase velocity, $v_{\rm gr} \ll c/n$). An additional dispersion in question may arise in EIT experiments (Electromagnetically Induced Transparency), where two light pulses excite a coherent oscillation of the atomic ensemble at the difference frequency (see, e.g., [10, 11]) or in dynamic grating experiments, where the dispersion results from the coupling of two fields by a self-induced (and therefore Bragg-matched) refraction-index grating [12–15]. In both cases, the coherence state (and dispersion) develops much slower than the electron motion in atoms gets excited. This gives a hope that optical forerunners might be observed on a sufficiently different time scale. It is clear that the conventional dispersion of the medium is always present in these experiments so that Brillouin's forerunner will be observed.

There exists a considerable difference between the light slowing down in EIT and in photorefractive grating experiments, that makes the second technique especially attractive for the experimental detection of forerunners. Initially, when the coherent oscillation of atoms in a gas is not excited, the two light waves are both absorbed very strongly (transmission of the order of $\exp(-20)$ or even less). The absorption is reduced by at least one order of magnitude when the coherent state of atoms is established. This means that the initial part of a light pulse on a time scale comparable with the build-up time of the coherent state (which might form the EIT forerunner) will be strongly absorbed. Therefore, it will hardly be detectable when followed by a much stronger pulse delayed because of fully developed EIT.

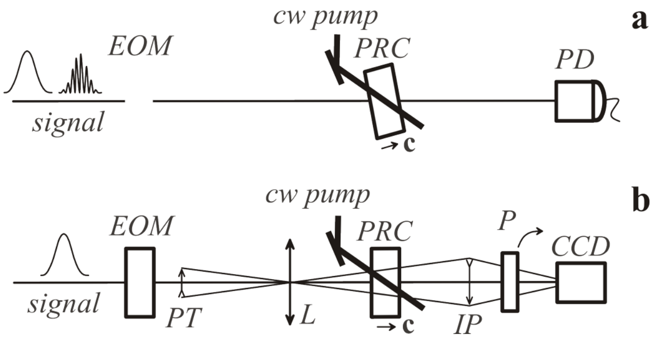

Fig. 1. Schematic representation of the experimental geometry for the analysis of temporal (*a*) and spatial (*b*) contents of the transmitted signal pulse; with the photorefractive crystal (PRC), electrooptic modulator (EOM), photodetector (PD), polarizer (P), polarizing transparency (PT), imaging lens (L), image plane (IP), and charge coupled device (CCD)

In the dynamic grating technique [12–14], the photorefractive crystals $BaTiO_3$ and $Sn_2P_2S_6$ initially are totally transparent (absorptivity on the order of few cm^{-1} or less). The slowing-down is observed for the pulses that are strongly amplified (the intensity gain factor more than 10 cm^{-1}). This allows the easier detection of "dynamic grating forerunners".

Here, we describe, at first, the observation of forerunners in the temporal dynamics of output signal beams in the two-beam coupling in photorefractive $BaTiO_3$. The results of measurements are compared with the calculated one within the model of two-beam coupling for the pulsed radiation [13]. Finally, we describe the study of a fine space-time structure of forerunners and prove, for the first time to our knowledge, the insensitivity of the forerunners to the induced dispersion. As distinct from the previous works, where the truncated pulses with the Heaviside step function have been used to detect forerunners, we work with Gaussian pulses which are short as compared with the decay time of a refractive index grating (which is sufficiently long, of the order of few seconds). This allows us to compare the results with the analytical theory of photorefractive recording by Gaussian pulses developed in [13].

The experiments have been performed with a 6-mm-thick z-cut $BaTiO_3$ sample and a He-Ne laser light of about 40 mW at 632 nm. Figure 1 gives a sketch of the experimental geometry: the output beam of a cw laser is split into two parts, a powerful pump beam and a weak signal beam (beam ratio is about 10,000:1). The signal beam is sent to an electrooptic modulator that tailors a pulse with the Gaussian shape, a variable duration, and, if necessary, harmonic intensity modulation in addition. Inside a $BaTiO_3$ sample, the signal meets the pump beam and gets amplified because of the two-beam coupling. The intensity evolution measured with photodiode PD is analyzed with a PC. To analyze the pictorial information in the transmitted signal, we use the geometry shown in Fig. 1,*b*. Here, the signal beam propagates along the optical axis of a $BaTiO_3$ sample, a polarizing transparency (PT) (a birefringent crystal) is put before the photorefractive sample, and lens L projects the image to the image plane (IP). A CCD camera with PC store the time-resolved images behind a removable polarizer (P).

The propagation of a forerunner is a linear effect [1, 9], so it experiences no gain. The pulse delayed with $BaTiO_3$, on the contrary, is strongly amplified because of the nonlinear wave mixing, up to 3000 times in our previous experiments [13]. The superposition of two pulses so different in amplitudes makes a forerunner practically indistinguishable. To reveal a forerunner, the delayed pulse should be inhibited to a certain extent. This can be done either by reducing the coupling strength of the photorefractive crystal or by choosing short input pulses, with the duration insufficient to develop an effective grating.

The time evolution of the transmitted signal is shown in Fig. 2 for different input pulse durations, while keeping the same coupling strength [16] $\gamma\ell = 3.7$, ℓ being the sample thickness.

The left column in Fig. 2 represents the measured temporal dynamics; the right column shows the results of modeling, as it will be discussed later on. It is obvious that the shorter the input pulse, the more pronounced becomes the narrow peak that propagates almost without pulse delay.

From the data similar to that presented in Fig. 2, we constructed the dependences of the output pulse delay time, pulse duration, and pulse amplification on the input pulse duration (Fig. 3). The data for a forerunner and for a delayed pulse are shown by filled squares and open dots, respectively.

Note that the intensity of a forerunner remains constant, on the same level as the intensity of a transmitted pulse with no pump (Fig. 3,*c*), up to the input pulse duration, for which the intensity of a delayed pulse becomes comparable and larger than that of the input pulse. Within the same range of durations of the input pulse, the duration of the forerunner replicates the duration of the input pulse (Fig. 3,*b*). The pulse delay (measured at the maxima of the first peak on the time scale) increases roughly as t_0^2 but remains much smaller than the overall pulse duration (Fig. 3,*a*).

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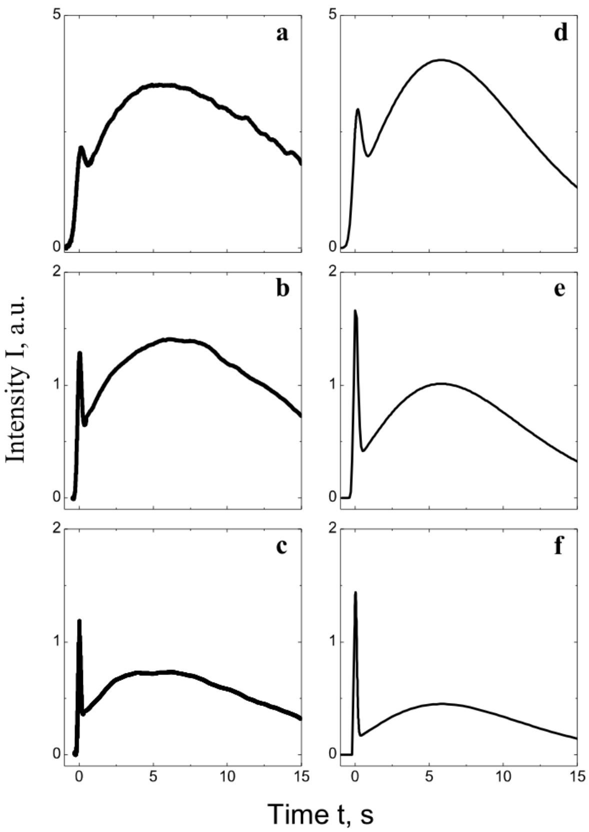

Fig. 2. Temporal envelope of the transmitted weak signal beam with the durations of a Gaussian input pulse equal to 0.48 s (*a, d*), 0.24 s (*b, e*), and 0.16 s (*c, f*), respectively. Left column of frames shows the measured data, while the right one presents the results of calculations

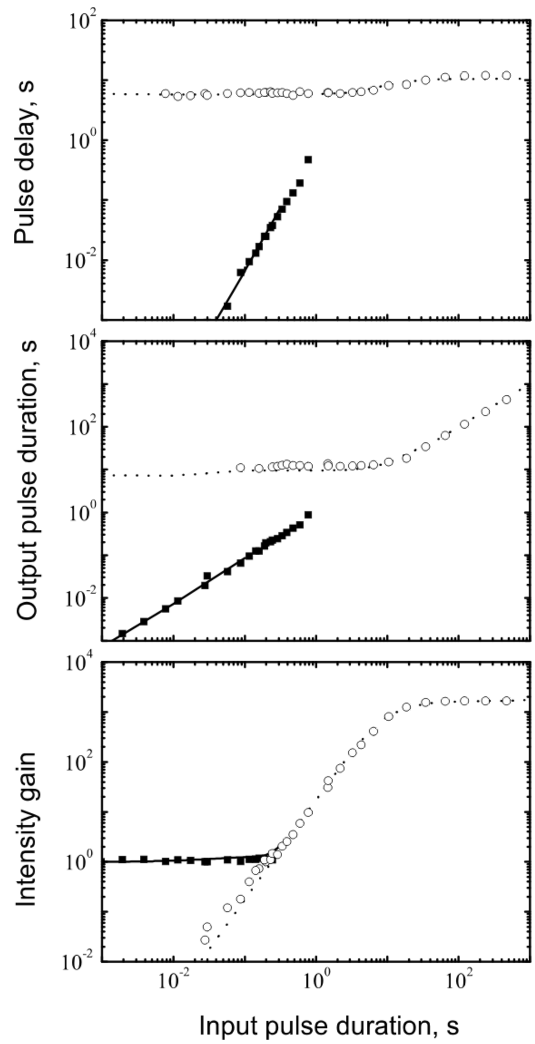

Fig. 3. Output pulse delay (*a*), duration (*b*), and intensity (*c*) versus the input pulse duration. The data for forerunners are shown by filled squares, and the data for a delayed pulse by open dots

For the delayed pulse, the behavior is similar to that described in [12]: the delay time changes not too strongly and saturates for long input pulses (Fig. 3,*a*). For short input pulses, the output pulse is strongly broadened, while the long input pulses do not change its duration (Fig. 3,*b*), and the intensity of the delayed pulse increases as t_0^2 and saturates for long pulses.

What one could expect for a forerunner is its nearly complete independence on the developing grating: it should keep the same intensity and the same duration and must be not delayed in time. This is what we observed (Figs. 3,*b*,*c*) except a very small but well measurable time delay of the first peak. The reason for this delay is quite clear, if we take into account that a forerunner is superposed to the up-growing "wing" of the amplified pulse with Gaussian time profile. The longer the input pulse, the stronger is the slope of the pedestal, to which the forerunner is superposed, and the larger becomes the apparent delay of a forerunner.

It should be noted that the appearance of forerunners follows also from direct calculations of the temporal envelopes of output pulses (see, e.g., Fig. 5 in our previous paper [13]). We fit the analytical expression for the pulse shape derived in [13] (τ being the space charge decay time) to the experimental data (Fig. 3). The solid lines in this figure are the result of such a fit that gives the fitting parameters $\tau = 2.85$ s and $\gamma\ell = 3.7$, close to those measured directly from the cw two-beam coupling experiments ($\tau = 3$ s and $\gamma\ell = 3.8$).

Now, with the known t_0, τ and $\gamma\ell$, it is possible to reconstruct the temporal profiles of output pulses for

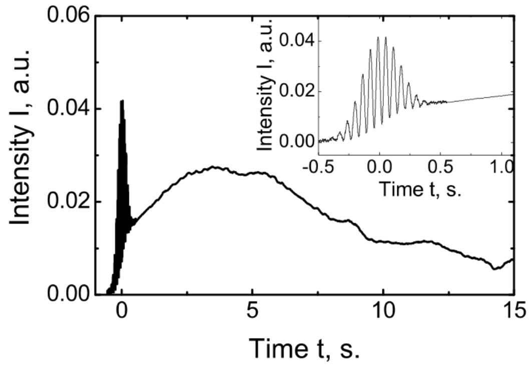

Fig. 4. Temporal variation of the transmitted signal beam. The inset shows the fine temporal structure of a forerunner

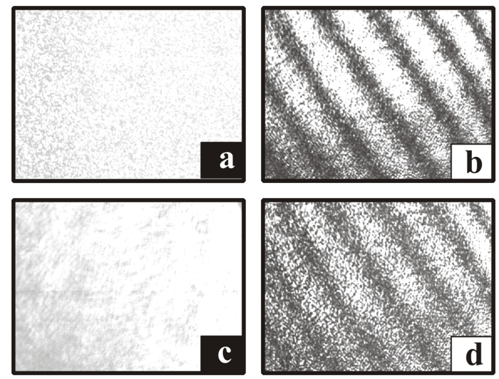

Fig. 5. Transverse intensity distribution in a signal beam propagating along the optical axis of the photorefractive crystal. (*a*) no pump and no polarizer are in front of CCD; (*b*) no pump, the polarizer is inserted in front of CCD; (*c*) no polarizer, the pump wave is switched on, the picture is taken within the forerunner duration; and (*d*) no polarizer, the pump wave is switched on, the picture is taken within duration of the delayed pulse

the experimental parameters of Fig. 2. The results are shown in the right column of this figure. A quite good qualitative correspondence in the measured and calculated dynamics can be considered as an independent verification of the validity for a theory given in [13]. The only free parameter that was used in this procedure is an amplitude scaling factor (which accounts for different losses that may occur because of the crystal absorption and scattering, Fresnel reflections, *etc.*)

When talking about forerunners, Brillouin underlined that, by definition, they must not feel the dispersion, because they leave the medium before the dispersion is established [1]. Moreover, Brillouin suggested that a forerunner should be insensitive also to the birefringence of anisotropic media [1], so that the polarization of the forerunner may be different from the polarization of crystal eigenmodes. A relatively long relaxation time of the photorefractive nonlinearity and the possibility of the gain control over a propagating signal beam allows for proving these statements experimentally.

In the first experiment, we introduce the sinusoidal intensity modulation with the frequency $\Omega \approx 100\ \mathrm{s}^{-1}$ to the input signal pulse, which is out of the bandwidth of the photorefractive grating, $\Omega > 1/\tau = 0.35\ \mathrm{s}^{-1}$ (the other parameters being $t_0 \simeq 0.3$ s and $\gamma\ell = 3.7$). The temporal evolution of the output pulse is shown in Fig. 4, with a magnified forerunner in the inset. One can clearly see a high-contrast intensity modulation within the forerunner which disappears completely in the delayed pulse.

This result is easy to understand in terms of the recording of dynamic gratings: within a short time t_0, we see, at the output, only the light transmitted from the input. The efficiency of the dynamic grating is negligibly small, and no coupling between the signal wave and the pump wave occurs. So, the temporal spectrum of an output signal pulse is the same as that at the input, and a deep intensity modulation is observed. With increase of the exposure time, a photorefractive grating develops and leads to the unidirectional intensity transfer from the pump to the signal wave. This grating should have a strong immobile component (written by a pump wave with frequency ω and the signal wave with principal frequency ω, too). The moving gratings, written by the pump wave with side-frequencies $\omega \pm \Omega$ of the signal wave are strongly inhibited for $\Omega > 1/\tau$. Thus, the contribution to the signal wave which is due to the diffraction of the pump wave from the immobile grating possesses only one main frequency ω. This is why there is no intensity modulation in the delayed pulse (Fig. 4).

The second experiment deals with the 2D polarization image imprinted in the signal wave (Fig. 5). To produce a spatial variation of the light polarization, the wedge of a birefringent crystal (quartz) is used. It is aligned at 45° with respect to the polarization plane of the incident wave (PT in Fig. 1,*b*). In this particular experiment, the signal beam is sent to the sample exactly along its optical axis. This is the only one direction in birefringent $BaTiO_3$, where the small-divergent beam with inhomogeneous polarization can propagate with no polarization distortion.

When a photorefractive grating develops, with the space charge field aligned roughly at 90° to the crystal

c-axis, the initial $4mm$ symmetry of $BaTiO_3$ is broken, and the azimuthal polarization degeneracy in a plane normal to the c-axis does not exist no more. New polarization eigenmodes are those linearly polarized along the space charge field and orthogonal to it. This should impose certain constraints to the further recording of the dynamic grating and to the diffraction by it.

The first frame (a) in Fig. 5 shows the homogeneous intensity distribution in a signal wave stored with a CCD camera, when the polarizer P in front of CCD is removed. The second frame (b) shows intensity fringes, when the polarizer is put in front of CCD, proving in such a way that the polarization is spatially nonuniform. Two last frames, Figs. 5,c,d, show the images recorded by CCD with no polarizer in 0.2 and 5 s after the exposure beginning, i.e., images transmitted by the forerunner (c) and the delayed pulse (d), respectively.

The result of this experiment confirms that the inhomogeneous spatial polarization of the signal is preserved in the forerunner but not in the delayed pulse. In terms of the dynamic grating, it can be explained in the following way: in $BaTiO_3$, the photorefractive grating is recorded via the diffusion-driven charge transport [16]. To ensure the efficient diffusion, one needs to have large gradients of the photoexcited carrier density and, therefore, the strong intensity modulation in light fringes. In turn, this means that the polarization of the signal wave should be identical to that of the pump wave. This requirement imposes a selection rule for the polarization of the delayed pulse that does not apply for forerunners.

To conclude, when studying the slow light propagation in photorefractive crystals we have found the experimental conditions for the observation of a “photorefractive” Brillouin forerunner: a part of the output signal represents a pulse with nearly the same intensity and duration as the input pulse, and it is not delayed with respect to the incident pulse. The spatial, temporal, and polarization contents of this pulse can be more rich as compared to those of the delayed pulse, because a forerunner is insensitive to the filtering properties of the arising dynamic grating. One obvious conclusion of this study concerns the possible slowing down of pulse sequences: to get a delayed pulse train with no distortion, one should use a photorefractive crystal with the response time that is comparable or slightly shorter than the duration of every pulse in a sequence.

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ОПТИЧНІ ПРОВІСНИКИ В КРИСТАЛАХ З ДИНАМІЧНИМИ ФОТОРЕФРАКТИВНИМИ ҐРАТКАМИ

О.М. Шумелюк, А.І. Григоращук, С.Г. Одулов

Р е з ю м е

Експериментально та теоретично показано що уповільнення світлових імпульсів внаслідок фоторефрактивної взаємодії супроводжується швидким поширенням короткої початкової частини імпульсу, що можна розглядати як існування провісника. Відповідно до передбачень Бріллюена і Зоммерфельда дисперсія не впливає на провісників; показано, наприклад, що в них може бути більш багатий спектральний склад порівняно із основним уповільненим імпульсом, а також, що поляризація провісників не обов’язково повинна відповідати поляризації власної хвилі кристала із ґраткою просторового заряду.