Squeezing of laser pulses in a nonlinear-optical LC cell

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Abstract—Controllable compression of temporal duration of light pulses takes place in a liquid crystal medium via self-action processes of beam mixing, namely, a dynamic Bragg grating formation by incoming light waves with their consequent self-diffraction on the same recorded grating. Wherein the laser pulse duration should be comparable with the time relaxation constant of the medium while the extent of temporal compression is controlled by variation of input pulse durations and the value of nonlocal response.

The interaction of light waves in a nonlinear optical medium is an efficient tool for providing different manipulations of parameters of laser beams [1]. Two- and four-wave mixing in nonlinear media are widely used for amplification of laser beams and optical images, creation of phase delay lines, in beam steering, phase conjugation, shaping of pulses, and in a variety of optical sensors, etc. [2]. Liquid crystal (LC) cells are very promising candidates for realization of these effects, as they combine a very high nonlinear optical gain and non-locality of the response. LC cells with photorefractive properties make an optical element called a liquid crystal light valve (LCLV) that could be used in light-controlled-by-light systems [1]. Processes arisen due to wave-mixing in an LCLV are based on generation of a phase grating, i.e. a modulation of the refractive index due to changing orientations of LC molecules in the cell volume.

In the present work we investigate the two-wave mixing process in a nonlinear LC, which reveals the squeezing of output laser pulse due to incomplete relaxation of the phase grating and further self-conversion processes under the action of a regular series of pulses. These effects of dynamic memory of the medium occur when the duration of laser pulses is comparable with the response time of nonlinearity. We apply a nonlinear problem of waves' self-conversion process in a Kerr-type dynamic medium and develop a theoretical model describing such an effect in an LC-cell. The process of temporal pulse compression strongly depends on the presence of a nonlocal component of nonlinearity. We show that temporal squeezing of pulse duration can be greater than twofold with respect to the input pulse and can be controlled by laser pulse duration.

The simplest dynamic holographic scheme involves the interaction of two input coherent waves I_{10} and I_{20} in a nonlinear dynamic medium. Generally, the process is described with a system of coupled-wave equations for two interacting waves (in the Bragg diffraction conditions) [3-4]:

$$\frac{\partial \mathbf{E}_1}{\partial z} = -\frac{ik}{2n\cos\theta}\varepsilon_1 \mathbf{E}_2 \;, \quad \frac{\mathbf{E}_2^*}{\partial z} = \frac{ipk}{2n\cos\theta}\varepsilon_1 \mathbf{E}_1^* \tag{1}$$

and an evolution equation for the amplitude of the dynamic grating, which comprises two main processes: the gain proportional to the light intensity $E_1 E_2^*$ and relaxation with the time constant T_{relax} :

$$\partial\varepsilon_1 / \partial t = \gamma E_1 E_2^* - \varepsilon_1 / T_{relax} . \tag{2}$$

In Eqs. (1) and (2) the designations are the following: $\mathbf{E}_j(z,t) = E_j(z,t)\exp(i\phi_j(z,t))$, $j = 1,2$ are the light waves, $k = 2\pi/\lambda$ is the wave-vector, θ is the converging angle of waves, *n* is linear refractive index of the medium, "*" denotes the complex conjugation, *p*=1 (or –1) in the transmission (or reflection) geometry, ε_1 is the amplitude of the photoinduced grating, γ determines the maximum modulation depth of the grating.

The dynamic holography involves several simultaneous processes: the two interacting waves form an interference pattern $J_m(t,z) \sim E_1(t,z)E_2^*(t,z)$ (a so-called light lattice) inside a nonlinear dynamic medium; the light lattice induces modulation of the refraction index, i.e. a dynamic grating, $\Delta\varepsilon(t,z)$ the interacting waves diffract on the dynamic grating created by themselves. Two main important effects result from this self-action process, namely, (1) the phase transfer between interacting waves and (2) the energy transfer between them. The most efficient energy transfer takes place when the dynamic grating is nonlocal, i.e. there is some spatial shift between the light lattice and the dynamic grating. In general, the dynamic grating includes both local (ε_L) and nonlocal (ε_N) responses, which is described by the complex value of ε_1 : $\varepsilon_1 = \varepsilon_L + i\varepsilon_N$. It is important to note that in the case of energy transfer, both light lattice and dynamic grating acquire spatially nonuniform distribution of amplitude across the thickness z . Moreover, their amplitude profile gets a soliton-like form: as a bright soliton in transmission

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geometry or a dark soliton in reflection geometry. Furthermore, the whole nonlinear system (1)-(2) is reduced to a nonlinear Schrodinger equation [3], different soliton solutions which are intensively investigated throughout nonlinear physics. An essential condition for these localized states to appear is the finite-time relaxation of the grating. In the following we will investigate shape transformation of laser pulses interacting in the LC medium with the complex response, ε_1, in dependence on the ratio of the time relaxation constant of nonlinear response, T_{relax}, to the laser pulse duration, τ.

The main nonlinear optical mechanism usually employed in LC systems is the reorientation of an LC director in the bulk of a cell under the action of light field. A well-resolved one-to-one correspondence between the modulation of light intensity and that of the refractive index is required as far as different holographic applications are concerned. Owing to elastic forces of an LC medium, however, the LC director reorientation spreads far outside the illuminated region concealing the optical information recorded, making orientational nonlinearity inapplicable for dynamic holography.

Modulation of the LC order parameter does not require reorientation of the optical axis (director) while affecting the refractivity of the medium. A dye-doped LC is used as the nonlinear optical medium with employment of the LIOM-type nonlinearity (light-induced order modification), which accounts for the changes of the refractive indices resulted from the light-stimulated modulation of the LC order parameter: $\delta n_{o,e} \sim \Delta n_0 C_{ph}$ ($\delta n_{o,e}$ is the change of ordinary or extraordinary refractive indices (o, e - respectively), Δn_0 is the initial LC birefringence, C_{ph} is the concentration of light-excited molecules) [5]. The LIOM-mechanism explains experimentally observed large optical nonlinearities with extremely fast, as for an LC system, recording times [6]. Due to efficient local response, a very fine resolution is achieved (<1μm) accompanied with quite high diffraction efficiency within a very wide intensity range of pumping light [7].

A model nonlinear optical LC cell supporting the LIOM-mechanism, has been fabricated on the base of the nematic LC E7 dye-doped with Methyl Red (concentration of the dye was around 0.3 wt.%). A typical sandwich-like cell including two glass substrates covered with transparent ITO electrodes providing homeotropic anchoring was filled with an LC mixture. The cell gap was given by calibrated stripes of 30μm thickness. The application of the AC voltage (~100V, 1÷10kHz) ensured stabilization of the LC director orientation perpendicularly to the substrates deterring its reorientation.

In standard two-wave mixing (TWM) experiments [8], pumping beams came from an Nd:YAG continuous wave laser (single-mode operation, λ=532 nm) (see Fig. 1).

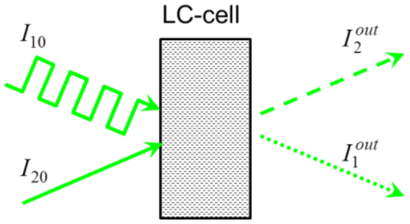

Fig. 1. Sketch of TWM arrangement. Waves designations: I_{10} (square pulsed) and I_{20} (continuous) – input; $I_{1,2}^{out}$ – output waves.

The input I_{10} wave is square-pulsed with pulse periodicity, T, and pulse duration, τ, the input beam I_{20} is of constant intensity.

Light-induced *trans-cis* isomerization of dye molecules resulted in the changes of LC ordering and, thus, of LC refractive indices [5]. A weak probe beam from an He-Ne laser (633nm) has been used to determine the time constants characteristic of the grating recording and relaxation (Fig. 2). The polarization of all beams was linear, parallel to each other and perpendicular to the plane of incidence. Typical time relaxation constants depended on pumping light intensity and appeared to be within the range of ~1÷50ms.

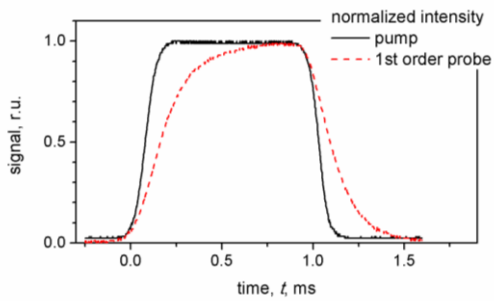

Fig. 2. Typical dynamic curves of pumping pulse (solid line) and probe beam diffraction (dashed line).

Two effects are predicted to take place under such experimental conditions, in accordance with the above-mentioned reasoning when the interacting waves form a dynamic grating with the non-uniform envelope of its amplitude along the z-direction. Firstly, there should be a significant amplification of one beam due to energy transfer from another beam. Secondly, the narrowing of the signal pulse is to be observed under the condition $\tau < T_{relax}$ (T_{relax} is the grating relaxation time). In this case, the dynamic grating is not erased completely, and the next recording/self-diffraction cycle starts on a partially existed grating. Thus, the effect of "dynamic memory of the medium" is manifested, which leads to many new interesting results.

Based on the set of nonlinear equations (1)-(2), the dynamics of the I_2^{out} signal and the output intensities for the transmission TWM arrangement have been calculated. The results of numerical simulation for some particular parameters are shown in Fig. 3.

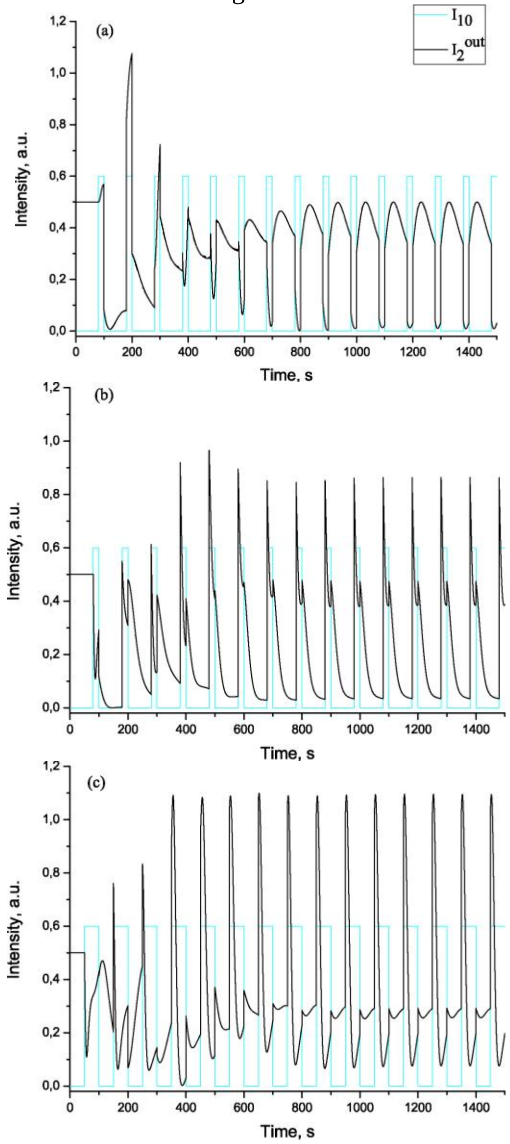

Fig. 3. Calculations of two-wave mixing in a LIOM-type LC-cell with relaxation time T_{relax}=100 ms. System parameters are: I_{10} is periodically pulsed with maximum intensity I_{10}=0.6 (n.u.), I_{20}=0.5 (n.u.) is continuous illumination, (a) T=100ms, τ=20ms, m_N=0, m_L=0.2; (b) T=100ms, τ=20ms, m_N=0.17, m_L=0.1; (c) T=150ms, τ=100ms, m_N=0.17, m_L=0.1.

The main parameters in the current model are the pulse periodicity, T, the time relaxation constant of the nonlinearity in the medium T_{relax}, and the values of the nonlocal (local) response, which are determined by the constant $\gamma_{N,L}$: $m_{N,L} = \gamma_{N,L} kd$ (d=30μm is the cell thickness used in simulations, $k = 2\pi/\lambda$). Our calculations show that despite continuous input of wave 2, the pulsed output beam I_2^{out} appears due to wave-mixing in the LC-cell. Designations and arrangement of waves are presented in Fig. 1.

In Fig. 3(a) the nonlinear response is purely local and no signal squeezing occurs. In the case of the nonlocal response, the duration of output pulses of wave 2 grows shorter than that of the input pulse $\tau_2^{out} < \tau$ and depends on the duration of the input pulse τ (see Fig. 3(b) and Fig. 3(c)). These calculations prove that the dynamics of wave-mixing in a medium with a complex nonlinear response and relaxation may be very complicated, and the time characteristic of both input laser pulses and medium relaxation should be considered. The pulse duration compression depends on mutual ratios of T_{relax}, T and τ, and the value of m_N. The exact way to pulse compression control will be addressed in further studies.

In conclusion, we show by theoretical and numerical modeling the existence of the effect of "dynamic grating memory" at mixing laser pulses in a dynamic nonlinear optical medium with relaxation. This effect may lead to the narrowing of laser pulses combined with their amplification. This effect occurs when the duration of laser pulses is comparable with the time relaxation constant of the medium. In our further work we will take into consideration the nonzero time constant of the grating recording after switching the light on, and also we will carry out experimental researches with the proposed nonlinear LC-cells supported LIOM-mechanism. Our studies show new possibilities for manipulation of laser pulses in photonic systems.

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