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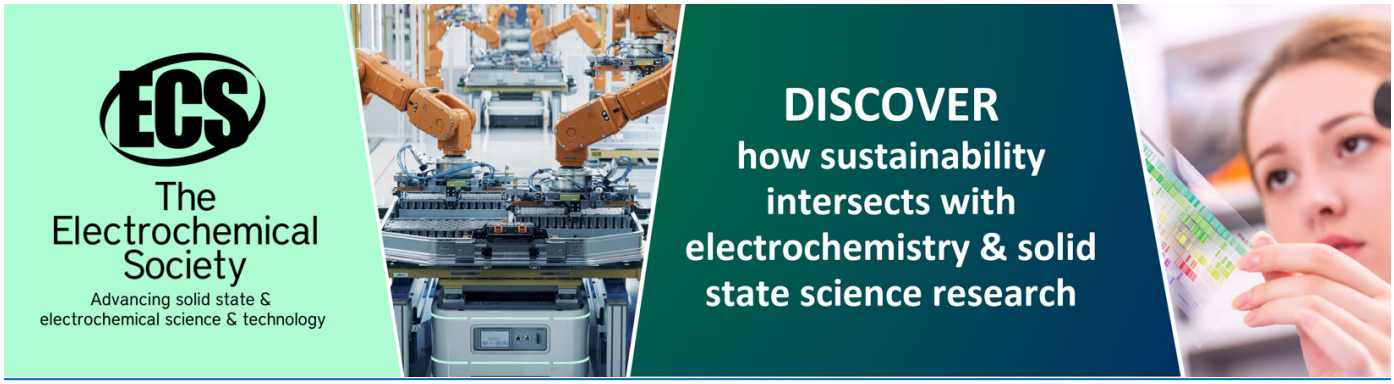

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Optical damage dynamics in reduced nominally pure $LiNbO_3$ crystals

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Abstract. The dynamics of the optical damage in the chemically reduced $LiNbO_3$ has been investigated, when a laser beam is externally focused into a bulk crystal. The temporal evolution of the transmitted power P was recorded, using the closed-aperture pseudo-Z-scan method. We define the three specific levels of the transmitted power that occur at different times during experiment: P_0 and P_1 are the initial and minimal values of the transmitted power during first stage of experiment, when the usual fast rise of optical damage causes decrease of the transmitted power due to laser beam defocusing, while P_2 is the steady-state value after the long-term exposure, when a sequent slow increase of the transmitted power is observed, indicating on the self-compensation of optical damage. This compensation becomes undetectable, when the input light intensity I within focal area is smaller than some specific value ranged from 4 to 90 W/cm^2 (λ=644 nm), depending on the chemical reduction degree. At intensities above such a threshold the self-compensation increases monotonically with I.

1. Introduction

It has been established [1,2], that a steady-state temperature gradient applied in the direction perpendicular to the laser beam propagation is able to decrease the optical damage in lithium niobate crystals. At the same time, the marked temperature gradient may arise due to local crystal heating caused by strong absorption of laser radiation, when light spot size is smaller than a sample dimension [3,4]. Besides, the macroscopic charge separation due to the pyroelectricity represents an important extra contribution to optical damage in ferroelectrics, which can occur whenever the temperature is raised significantly [3]. To examine these supposed light-induced effects, we generated a large temperature gradient by illumination of the chemically reduced crystals, absorbing strongly within visible range, comparing it with as-grown nominally pure and iron-doped crystals, when an extraordinary polarized laser beam is externally focused into a bulk crystal.

2. Experimental techniques

To induce the optical damage, the focused extraordinary polarized beam of a laser diode (λ = 644 nm) with a waist (intensity full width) equal to 1.0 mm in air has been used. The laser beam has stable power of 56 mW. To focus the beam, a lens with a focal length of 20 cm was used. The sample is

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placed near the focus point of the lens. By changing the position of the lens, we focus the laser beam with a waist radius w_0 = 70 μm at the entrance surface of the crystal. Thus, thickness of any crystal studied was less than the Rayleigh length, $L_R = \pi w_0^2/\lambda$, of the focused beam. We recorded the laser power transmitted through the crystal with the aid of a small-aperture photodiode located in the far field. The far field observation plane is 45 cm from the crystal. The dimension of the photodiode aperture was chosen to be 1.2 mm that is much smaller than the beam size in the far field zone. Therefore, only ≤ 20 % of the laser radiation transmitted through the crystal reaches the photodiode at the fixed distance of 45 cm from the focal plane. Such a set-up arrangement allows for utilizing the pseudo-Z-scan method in the closed-aperture scheme [1,5], where a lens focuses the laser beam to a certain point, after which the beam naturally defocuses. The aperture causes only the central region of the cone of light to reach the detector. Hence, the detector becomes sensitive to any extra focusing or defocusing that a sample may induce due to the light-induced refractive index change.

The main experimental study was performed with the as-grown, chemically reduced (by annealing in pure hydrogen atmosphere at 500 ^oC, or in vacuum 10^{-3} Torr at 500÷700 ^oC for 20 to 90 minutes), and deeply oxidized (by annealing in pure oxygen atmosphere at 1000 °C for 15 hrs) nominally pure congruent $LiNbO_3$ crystals. Besides, the complimentary measurements were made with the as-grown Fe-doped ([Fe] = 0.001 and 0.07 mol %) congruent $LiNbO_3$ crystals. Thickness of these crystals is ranged from 1 to 7.1 mm.

Table 1. Optical absorption coefficient values evaluated at λ = 644 nm.

Sample #:	1	2	3	4	5	6	7
α, cm^{-1}	~ 0.01	0.033	0.045	0.095	0.34	0.74	1.27
α', cm^{-1}	< 0.003	0.005	0.006	0.055	0.29	0.68	1.21

The optical transmission of the samples was measured using a polarized light at normal incidence on a SF-2000 spectrometer in the wavelength range 0.2÷1.1 μm. The typical absorption spectrum of the chemically reduced $LiNbO_3$ shows a broad absorption band ranging from the absorption edge to the near-infrared region. This absorption growth correlates closely with a significant increase of the so-called bipolaron absorption band [1,6], centered at 470-500 nm. The optical absorption coefficient α was calculated from the transmission data, using the Newton-Ralphson method [1,7]. Mathematical fitting of the spectral dependence of α was used to extract contributions of the long-wavelength tail of the fundamental absorption and the light scattering and to evaluate the additional absorption α' related to the bipolarons, Table 1.

3. Experimental results and discussion

To compare the optical damage at different light intensities, we vary the power of laser beam at a crystal input surface (so-called input power P_{in}) in the wide range from 0.6 to 56 mW with the aid of neutral density filters placed between laser and lens. Thus, the light intensity within a waist area of focused beam is ranged from 4 to 360 W/cm^2. Using the pseudo-*Z*-scan arrangement [1], the temporal evolution of the transmitted power *P* was recorded. We define the three important levels of the transmitted power that occur at different times during experiment: P_0 and P_1 are the initial and minimal values of the transmitted power during first stage of experiment, when the usual fast rise of optical damage causes decrease of the transmitted power due to laser beam defocusing, while P_2 is the steady-state value after long-term exposure, when a sequent slow increase of the transmitted power is observed during the second stage of our experiment, Fig.1.

The most important experimental finding consists of the demonstration of the self-compensation of optical damage for focused laser beams in chemically reduced nominally pure crystals at relatively

small laser powers (several mW), if the optical absorption coefficient at laser wavelength is sufficient to give rise for marked heating within the illuminated area. Figure 1 shows a typical curve of the temporal response of the transmitted power recorded by the closed-aperture photodiode in the far field region for a focused laser beam transmitted through a reduced nominally pure $LiNbO_3$ crystal. At the beginning of the light-induced process, the relative transmission decreases rapidly to a minimum value, then the relative transmittance increases slowly, approaching some steady-state value, which is smaller than the initial value (recorded at t = 5 s). According to the previous investigations [1,5], it can be directly associated with the optical damage kinetics.

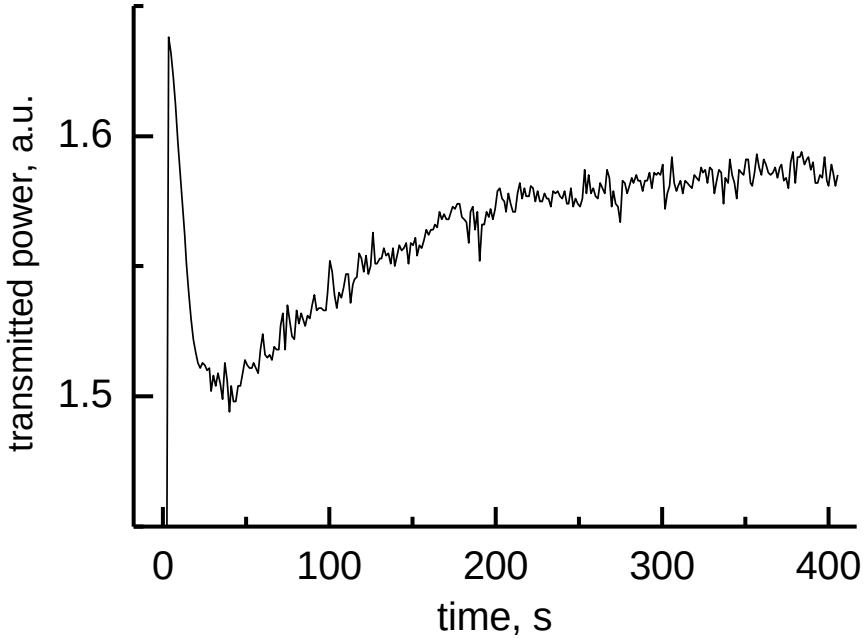

Figure 1. Optical damage kinetics in the chemically reduced sample #7, demonstrating the initial sharp rise of photorefractive defocusing (time period from 5 to 46 s) and sequent self-compensation of optical damage (time period from 46 to 410 s) with the steady-state value of transmitted power at t > 370 s. The input power had a value of 28.

According to previous data on pseudo-Z-scan method [1], the ratio between the transmitted powers P_0 and P_1 has been used as a qualitative measure of the optical damage magnitude, i.e. as an input data for evaluation of maximum refractive index change Δn_{max} caused by photorefractive effect during the first stage of experiment. This optical damage magnitude Δn_{max} observed at any fixed input power level is less significant in chemically reduced samples, than in the as-grown and oxidized $LiNbO_3$ crystals. Note, that the optical damage becomes undetectable with our experimental method in the reduced crystals annealed for more than two hours.

For the evaluation of the steady-state value of the optical damage magnitude, the ratio between P_0 and P_2 has been used by the same approach mentioned above. The degree of partial compensation of the optical damage observed at the second stage of exposure can be characterized by a self-compensation factor (*SCF*) defined as:

$$SCF = 1 - \Delta n_{st} / \Delta n_{max}, \quad (1)$$

where Δn_{st} is the steady-state value of the photo-induced refractive index change reached after long-term exposure.

It has been established, that *SCF* increases monotonically with P_{in} at low and moderate values of input power and approaches to a maximum value SCF_m at the largest input power used. The self-compensation becomes undetectable, i.e. $SCF < 0.02$, when input power is smaller than some specific value ranged from 0.6 to 14 mW, depending on the chemical reduction degree. The self-compensation factor depends on crystal parameters, e.g., SCF_m has different magnitudes for the crystals studied, and varies within the range from 0.09 to 0.92 in the chemically reduced nominally pure crystals. For example, comparison of the partial self-compensation effect in the different crystals demonstrates clearly that SCF_m correlates with the optical absorption coefficient α' in the reduced and as-grown crystals, Fig.2. It is important to note, that the self-compensation effect is not observed in deeply extra oxidized and as-grown nominally pure $LiNbO_3$ crystals subjected to proper oxidation annealing during the fabrication process. Also, any self-compensation of the optical damage was not observed in as-grown iron-doped congruent lithium niobate crystals even at the largest light-intensities of red laser emission used in our studies, Fig.3. At the same time, the optical absorption coefficient in the studied iron-doped crystals was the same or larger, comparing to the reduced crystals demonstrating the self-compensation effect.

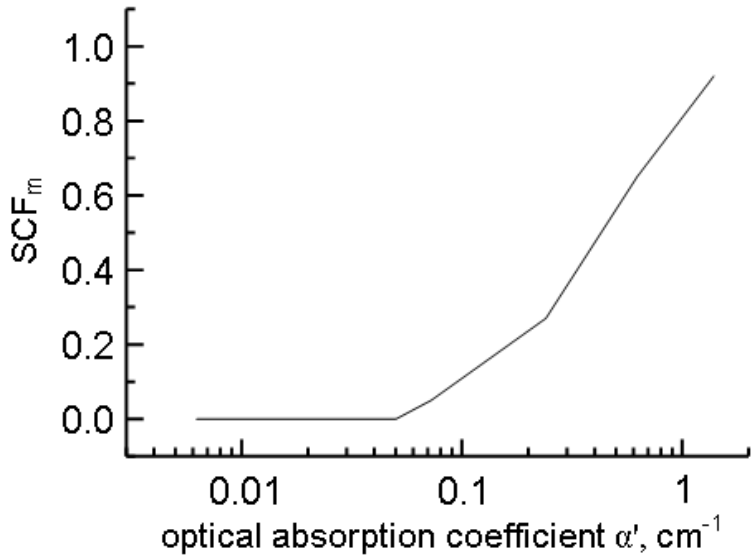

Figure 2. Maximum self-compensation factor SCF_m versus α' (at 644 nm) in various crystals.

The specific time τ_2 of self-compensation effect, i.e. the process corresponding to the partial transmittance recovery, correlates roughly with the optical absorption coefficient α': if value of α' is larger, then the self-compensation effect is faster. The weak quasi-linear dependence of τ_2 on P_{in} has been observed with the red emission at low and moderate powers used in our experiments, demonstrating the following intensity dependence:

$$\tau_2 \sim A/[\alpha'(B+CI)], \quad (2)$$

where *A, B, C* are proportionality constants, *I* – light intensity within focal area of the laser beam, and, according to our experimental data $B >> CI$, even at largest *I* reached in our experiments.

The main experimentally observed feature, which gives key information about dominating mechanism of the self-compensation effect, is the specific range of time constant τ_2 for the transients

observed during the second stage of the optical damage developing (see Figs.1). In any reduced crystal studied, the values of τ_2 (hundreds of seconds) is much longer than expected time constants for the thermo-optic [8] and pyroelectric [3,4] contributions in the optical damage. At the same time, the kinetics of the self-compensation effect is much faster than a speed of the dark relaxation of the optical damage.

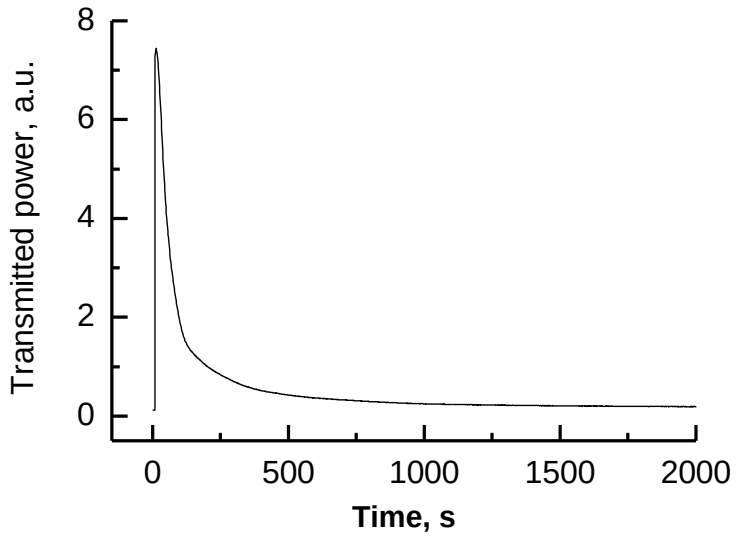

Figure 3. Optical damage kinetics in the iron-doped ([Fe] = 0.001 mol %) sample at input power of 56 mW, demonstrating the usual developing of the optical damage.

The dark relaxation of light-induced changes of the relative transmittance has been studied for the different stages of the optical damage developing in the various lithium niobate crystals. Only the very slow dark recovery (specific time is ranged from few hours to several tens of hours) of the transmitted power to an initial value P_0, that can be related to the slow dark relaxation of the optical damage, was observed, when illumination was paused at the optical damage developing in any lithium niobate crystal studied. It's the common well-known feature of the optical damage reported previously for the as-grown nominally pure and iron-doped lithium niobate crystals at ambient conditions [5,9].

According to the previous studies of the holographic recording in $Sn_2P_2S_6$ crystals [10] and thermal fixing of photorefractive hologram in Fe:$LiNbO_3$ [11], such a dynamics of photorefractive response is an evidence of the presence of two types of movable charge species of opposite sign. It has been regarded [10] as the fingerprint of strong charge-coupling effects between the active (photo-generated) and passive (thermally generated or permanent) charge carriers. The specific kind of electron-hole competition in photorefractive crystals, with considerably different (several orders of magnitude) dielectric relaxation times for electron and hole subsystems was already defined [10]. However, in our case of chemically reduced nominally undoped $LiNbO_3$ crystals the both types of movable charge carriers has been established undoubtly to be optically active, as no compensation effect with time constant about τ_2 is observed in the dark. These photo-induced charge carriers have been assigned to the small electron and hole polarons, appearing due to the photoexcitation of the intrinsic defects appearing at reduction treatment.

It is important to note, that the specific time τ_2 of the self-compensation effect is very close to the specific time of the temperature gradient-induced suppression of optical damage [1], when both effects are measured in the same crystal. Note, that the sample noted as #4 (Table 1) along this study was used in our previous study (sample #7 in ref.[1]). It allows for conclusion on same dominating mechanism of both effects.

Accordingly, the pyroelectric and thermo-optic effects is expected to give a significant contribution in the refractive index modulation only at much higher light intensity [12], which can be obtained by a further increase of the laser power or/and a much sharper focusing of the laser beam. In fact, such a situation was observed experimentally with a focused powerful radiation (100 ÷ 1850 mW) of an Ar^+-laser [3,4,13-16].

4. Summary

The optical damage developing process has been found to be substantially different in the chemically reduced nominally pure $LiNbO_3$ crystals, depending on intensity of bipolaron absorption band. A pronounced nonlinearity of transmittance followed by a quasi-steady-state behavior occurs, when some moderately reduced $LiNbO_3$ are exposed by a focused 644-nm-laser beam. Under such conditions, these crystals develop the optical damage quite rapidly, and then recover (partially compensate) more slowly, establishing that appears to be a less damaged steady-state. Such a self-compensation of the optical damage is observed at relatively small laser powers (5÷56 mW), when the optical absorption is sufficient to give rise for a marked temperature gradient within the illuminated area in chemically reduced $LiNbO_3$ crystals.

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