Fixed index gratings in $LiNbO_3$:Fe upon long-term exposure to an intense laser beam

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Permanently fixed noisily recorded refractive index gratings are discovered in single crystals of $LiNbO_3$:Fe upon long-term exposure to a single laser beam with high intensities of $I=100$ kW/m^2 and ambient temperature. The fixing process is explained by considering the effect of laser-induced heating of the sample and the well-known simultaneous thermal fixing procedure. © 2007 Optical Society of America

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The illumination of photorefractive crystals with a single laser beam is accompanied by the recording of noisy refractive index gratings. The grating recording is initiated by the interference between the laser wave and waves initially scattered from optical inhomogeneities, which leads to noisy refractive index gratings via the photorefractive effect. Two-wave mixing on these index gratings yields an amplification of scattering waves and then results in the polarization-isotropic wide-angle photoinduced light scattering (PILS) [1]. In Fe-doped $LiNbO_3$, the recording process at ambient temperature and moderate intensity of the laser beam relies on a charge-transport dominated by the photovoltaic current with a minor diffusion contribution. This can explain a nearly symmetric angular distribution of PILS in the $\pm c$ direction [2].

In this Letter we present our investigations on the exposure of Fe-doped $LiNbO_3$ to a single laser beam ($\lambda=514$ nm) of high intensities up to $I=100$ kW/m^2, over a time period of 12 h and at ambient temperature. This procedure yields significant features of the recorded noisy gratings and hence of the scattered light distribution in the $\pm c$ direction: (1) the grating recording, and thereby the buildup of the scattered light distribution, over a time scale of several hours; (2) permanently fixed noisy index gratings, i.e., they cannot be erased by moderate exposure to white light; and (3) a nonsymmetric angular intensity distribution. These features can be explained by considering the effect of laser-induced heating and the *simultaneous* thermal fixing procedure [3] of noisy gratings.

Our studies were performed with a cuts of congruently melting $LiNbO_3$ single crystals with 0.1 mol % Fe_2O_3 in the melt and dimensions of 5 mm $\times$ 4 mm $\times$ 0.5 mm. The unexpanded extraordinarily polarized beam of an Ar-ion laser with a diameter of 3 mm served as recording and probe beam at normal incidence. Initial conditions of the samples were reached by thermal treatment at 350 °C for 60 min and subsequent cooling down to ambient temperature prior to each measurement.

Figure 1 shows the angular intensity distribution of the scattering for exposure to an extraordinarily polarized laser beam with an intensity of 100 kW/m^2 for a duration of 5 min (lower inset) and of 12 h (upper inset), as well as its kinetics during 12 h of exposure (intensity plot). The spectra are determined by measuring the angular distribution of the scattering distribution repeatedly in intervals of 10 min. The legend on the right gives the scale of the scattering intensity on a logarithmic scale normalized to the initially transmitted intensity. The initial PILS profile (lower inset) shows a distribution of scattered light in a wide angular range with slightly different maximum intensities in the $+c$ and $-c$ directions [2]. However, strong changes in the symmetry of the PILS profile, its maximum scattering intensity, and the angle related to the maximum of the side lobes are discovered with ongoing exposure. The steady state is reached not before 20 h and shows a pronounced asymmetry in the scattering profile (such as in the upper inset). We recall that no external heating is supplied to the sample during the measurement. But a local heating of the sample can be well-assumed taking into account the absorption coefficient of

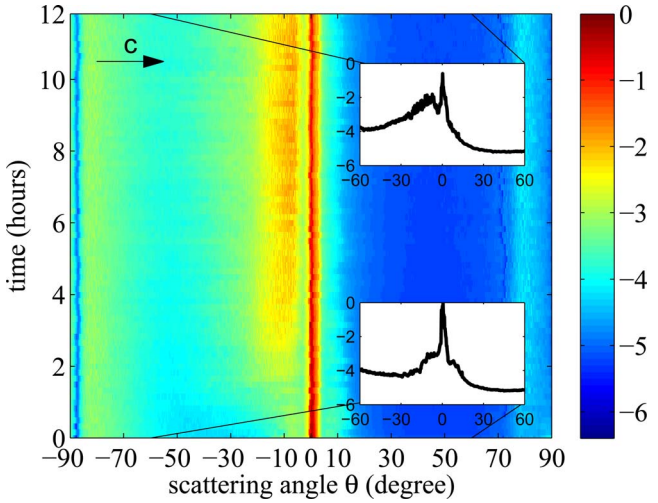

Fig. 1. (Color online) Angular and time dependence of the intensity distribution recorded with a huge intensity of 100 kW/m^2 and without any external heating. Bottom inset: angular distribution after 5 min of recording. Top inset: distribution after 12 h of recording.

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$\alpha=(5.9\pm0.5)$ cm^{-1} and the high laser beam intensity [4]. An indication of the local heating is a decreased apex angle of the polarization-anisotropic scattering cone [5] of about 1° compared to its apex angle using a moderate intense laser beam. From this the local temperature in the exposed volume can be roughly determined via the temperature dependence of the birefringence to be more than 100°C [6].

The significant temperature rise influences the particular contribution of the charge-transport mechanisms: At ambient temperature, a dominating contribution of the photovoltaic effect over diffusion is well-known and results in the nearly symmetric angular profile (compared to a one-sided profile for only diffusion charge transport) [2]. With increasing temperature the photovoltaic field $E_{\mathrm{phv}}=j_{\mathrm{phv}}/\sigma$ decreases, because of the increasing conductivity σ [7]. In the regarded temperature range it is decreased by ~70% of its value at ambient temperature. On the other hand the contribution of the diffusion field $E_D=(2\pi k_b T)/(e\Lambda)$ to the space charge field increases linearly with rising temperature T (Λ: grating spacing) [8], which effects the diffusion field by more than 30%. As a result changes of the measured scattered light distribution can be expected, in particular regarding the nonlinear relation between space charge field and scattered light intensity [9].

In the next step it is necessary to clarify the origin for the huge recording time of several hours. Obviously, this cannot be explained simply by considering electrons as free charge carriers, which is valid for many photorefractive crystals. It results in short-time recording and in an acceleration of the recording process at high temperatures due to the increased conductivity. It is rather necessary to take charge carriers with a low mobility, such as ions or ambipolar pairs of electrons and ions, into account. This can even be expected considering the elevated crystal temperature [3,10]. It is therefore also not astonishing that further inspection of the induced noisy gratings turns up features that strongly resemble the processes of *thermal fixing*: a detectable decay of the scattering was not found in the dark at ambient temperature. Furthermore, exposure to spatially homogeneous coherent laser light or to incoherent white light of moderate intensity did not result in a remarkable change of the scattered light distribution. The latter is demonstrated in Fig. 2, which compares the intensity distribution of the scattering on induced noisy gratings at the end of exposure (circles) with the profile determined after a 30 min exposure to incoherent white light (squares). Obviously, the shape of the scattering is not altered by the illumination. Only a decrease of the intensity by a factor of two for negative scattering angles is obvious.

Successful erasure of the noisy gratings was possible only by significant thermal treatment, i.e., via heating the sample to 350°C for a duration of 60 min. This allows to determine the activation energy related to this thermal decay. For this purpose, the increase of the intensity of the transmitted laser beam to its initial value was measured (subsequent to 12 h of exposure and 30 min of white-light illumination) as shown exemplarily in Fig. 3 for a temperature of 100°C. Fitting of the data set with a monoexponential growth function (solid curve) yielded the time constant $\tau(T)$. Then, the activation energy was determined via the Arrhenius relation $\tau(T)=\tau_0\exp(E_a/k_bT)$ [11] (see inset) to $E_a=(0.97\pm0.06)$ eV. This value is found to be in good agreement with the activation energy reported in literature for ionic charge carriers in Fe-doped $LiNbO_3$ (0.8–1.3 eV [11]).

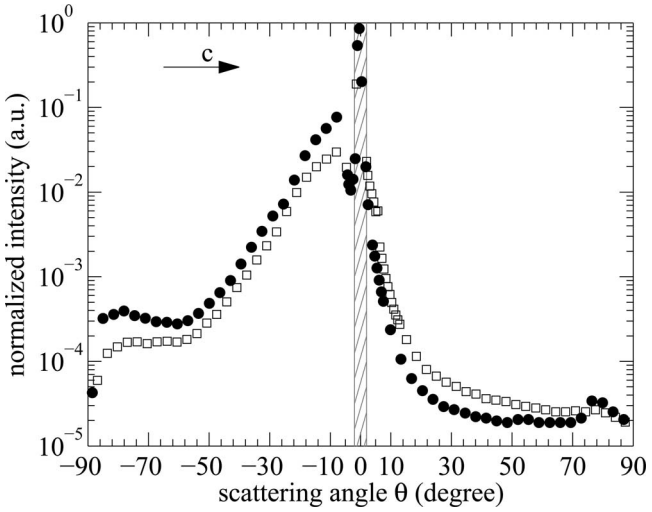

Fig. 2. Angular dependence of the scattered intensity after cooling the crystal to 25°C (circles) and after subsequent developing by white-light exposure with moderate intensity for a duration of 30 min (squares).

From these properties we conclude that the noisy gratings are *thermally fixed* upon the long-term exposure with the single laser beam at high intensities. Obviously, this procedure is best comparable with the *simultaneous thermal fixing* procedure [3], where the sample is illuminated during the heating process. Hence, we performed equivalent recording of noisy gratings by a single laser beam, but at moderate laser beam intensities and with external heating of the sample to 180°C. The results are shown in Fig. 4. The lower inset is the angular scan with a moderate intensity of the laser beam ($I=10$ kW/m^2) and at am-

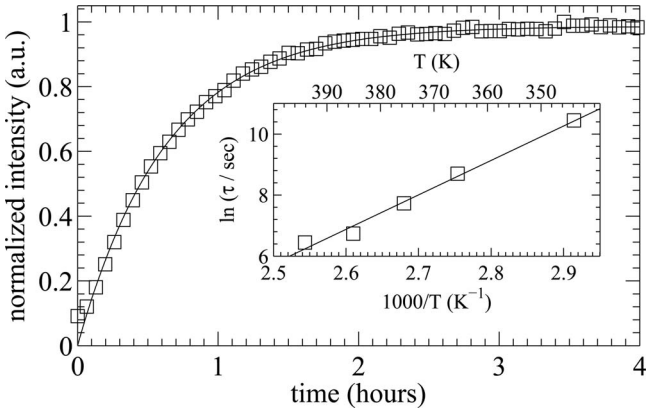

Fig. 3. Intensity of the transmitted light during thermal decay of the light scattering at 100°C. Solid curve: fit with a monoexponential growth function. Inset: Arrhenius plot to determine the activation energy via the linear fit.

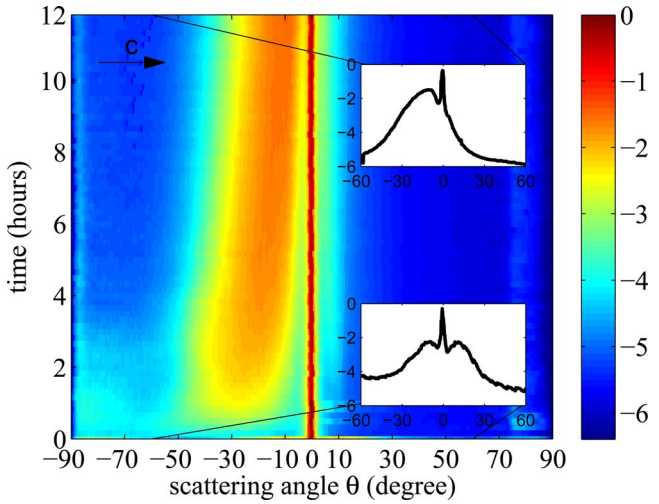

Fig. 4. (Color online) Angular and time dependence of the scattered intensity distribution according to the simultaneous fixing procedure ($I=10$ kW/m^2, $T=180$°C). Bottom inset: PILS prior to heating. Top inset: angular distribution after 12 h of exposure.

bient temperature. Two pronounced scattering lobes in the $+c$ and $-c$ directions appeared (PILS). Heating the crystal to 180°C in the presence of the laser beam at the beginning results in a disappearance of the scattering. According to the simultaneous fixing process, it results from field-compensation processes of the charge-carrier distribution with thermally activated ionic motion. However, even in this procedure a scattering distribution reappears during 12 h of exposure (upper inset) that—in agreement with our findings in Fig. 1—appears only in the $-c$ direction. To investigate the stability of the recorded noisy gratings, the sample has been cooled down to ambient temperature and was illuminated homogeneously by white light. According to the results shown in Fig. 2 slight difference between the scattering intensity distribution before and after white-light illumination is observed. In addition, the activation energy for ions involved in the thermal fixing process of holographic scattering was determined to be 1.05 ± 0.02 eV (see, e.g., Ellabban *et al.* [12]). It should be added that changes in the angular intensity distribution of the scattering as well as fixing of noisy gratings do not appear on the long-term for the case of exposure with a laser beam of moderate intensity at ambient temperature.

Inspecting these results we can conclude that long-term exposure of $LiNbO_3$ with an intense laser beam at ambient temperature results in a local heating of the sample and thus allows for thermal fixing of noisy gratings according to the simultaneous fixing procedure. The results therefore are in full accordance with the reports obtained with two-beam interferometers [13]. It has to be stressed that the local heating is induced by the recording beam itself, instead of using a second (intense) beam for local heating besides a (moderate) recording beam, like Liu *et al.* [14]. Additionally we like to point out that investigation of the angular distribution of the light scattering turns out to be an additional valuable tool for the understanding of charge-transport mechanisms at elevated temperatures in photorefractive crystals [15]. In particular, it impressively shows the interplay between photovoltaic and diffusion charge transport mechanisms. Besides, the results are of interest for several applications of photorefractive crystals including archival holographic data storage and fixed, volume Bragg gratings for light control. We suggest that the results in particular might also be considered in optical damage mechanisms occurring during second-harmonic generation, where the samples are exposed to intense light in the long term.

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