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Dark decay of the optical damage in iron-doped lithium niobate crystals*

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Abstract: In order to research into the optical damage in iron-doped lithium niobate crystals, we investigated the decay behavior of the optical damage in iron-doped lithium niobate crystals. The dark decay time of the optical damage in thin crystal sheets was found to far exceed that in the thick crystal sheets. The OH spectra and Raman spectra were performed in order to clarify the underlying mechanism. In conclusion, the thickness of samples greatly influences the optical damage.

Key words: lithium niobate; optical damage; dark decay; crystal thickness

掺铁铌酸锂晶体中光损伤的暗衰减效应

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摘要: 为了研究铌酸锂晶体中的光损伤, 着重对掺铁铌酸锂晶体中光损伤的暗衰减效应进行研究, 发现在薄样品中光损伤的暗衰减时间远大于在厚样品中的暗衰减时间, 利用红外吸收光谱和喇曼光谱对这个现象的潜在机理进行了初步分析, 综合几个实验结果发现, 晶体厚度对光损伤的暗衰减是一个重要的影响因素。

关键词: 铌酸锂晶体; 光损伤; 暗衰减; 晶体厚度
中图分类号: O472.3; O734 文献标识码: A

Introduction

Optical damage in photorefractive materials has been widely studied for applications such as holographic memories, wave-guide structures, electro-optic modulators and solid-state lasers since its discovery[1~3]. It has been found that the effect of optical damage is dependent on dopants[4], ratios of lithium to niobate ions in crystals[5], incident light intensities, etc[6,7]. However, no systematic studies on dependence of the optical damage on the crystal thickness have been reported so far. In this letter, we investigated systematically the decay behaviors of the optical damage in iron doped lithium niobate crystals of the different thickness and found that the decay behavior of the optical damage is dependent on the thickness of the crystal which was never expected by current photorefractive theory.

1 Experiments

Several x-y-cut $LiNbO_3$:Fe sheets of different thickness (0.64mm, 0.80mm, 0.82mm, 1.12mm and 1.14mm) were used in our experiments. They were cut from a congruent $LiNbO_3$ crystal doped with 0.1wt% Fe_2O_3 that was grown by R & D Center for Photon-Electro Materials of Nankai University. Figure 1 shows the experimental setup to study the dynamic decay behavior of the optical damage. An infrared light of 808nm from a laser diode was used as a pump beam and it was focused into a crystal in order to induce the optical damage. A weak He-Ne laser at 632.8nm was used to image the optical damage regions. The laser-induced optical damage was monitored

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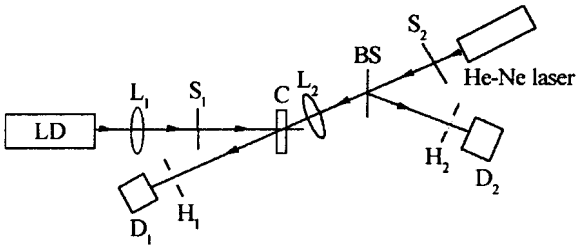

Fig. 1 Experimental setup to study temporal evolution of the transmitted intensity of optical damage with crystals of different thickness
LD—laser diode, L_1, L_2—lens, S_1, S_2—shutters, C—crystal sample, BS—beam splitter, H_1, H_2—pinholes, D_1, D_2—detector

by a detector D_1. A pinhole H_1 was put in front of D_1 in order to choose the measured part of the optical damage image. The stability of the He-Ne laser was monitored by another detector D_2. In order to prevent scattered lights from reaching D_2, another pinhole H_2 was put in front of D_2. We observed bubble-like optical damage image with two bright crescents along the c-axis as reported[8] when the pump light was incident on the sample. The optical damage was found to be stored in the crystal for a period of time even after the removal of the pump beam.

2 Results

Figure 2 shows the dynamic behavior of the optical damage in crystals of different thickness (d_1= 0.8mm, d_2= 1.12mm) with a pump beam intensity of 460W/cm^2. It is seen that the light intensity detected by D_1 (Id_1) decreases at the beginning of the pump beam illumination and then saturates for both crystals.

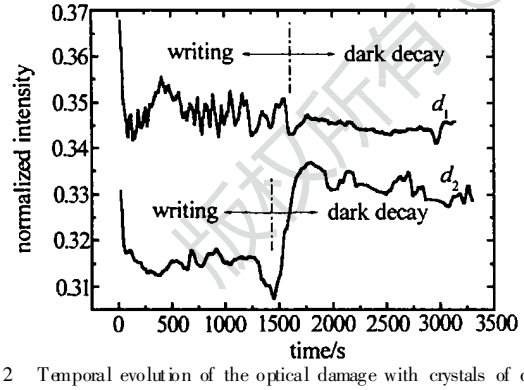

Fig. 2 Temporal evolution of the optical damage with crystals of different thickness

This means the optical damage is induced and finally reaches saturation in the crystals. On the other hand, when the pump light was blocked, no change in I_{d_1} was found for the 0.8mm-thick sample during our measuring period, whereas I_{d_1} increased quickly to its original value for the case of the 1.12mm-thick sample. This definitely shows that the dark decay time of the optical damage in the 0.8mm-thick sample is much larger than that in the 1.12mm-thick sample although the formation time of the optical damage are almost the same for both samples.

We performed the same experiments as described above in several other crystal sheets. All results definitely show that, although the formation time of the optical damage is almost the same in these crystal sheets of different thickness, the dark decay time of the optical damage in a thin crystal sheet is longer than that in a thicker crystal sheet. For example, with a writing intensity of 396W/cm^2 the relationship between the dark decay time and the thickness is shown in Table 1. The writing time is 17s in a 0.8mm $LiNbO_3$:Fe crystal, while it is 12s in a 1.14mm $LiNbO_3$:Fe crystal. On the other hand, the dark decay time is 240h in the 0.8mm sample, while it is 1.9min in the 1.14mm sample. In addition, we also repeated the same experiment in several z-cut $LiNbO_3$:Fe crystal sheets and no optical damage was observed in these z-cut crystal sheets. This result indicates that the observed optical damage in the x (or y)-cut samples has no relation with the thermal effect as we expected.

Table 1 Thickness dependence of writing time and dark decay time

thickness / mm	writing time/ s	dark decay time/ s
0.80	16.98	864000
1.12	14.46	238.14
1.14	11.97	114.18

It is difficult to explain such a thickness-dependence of the dark decay time of the optical damage in $LiNbO_3$:Fe crystal on the basis of the current photorefractive theory. In order to find out the underlying mechanism, we measured the infrared absorption spectra of the optical-damage region and the non-optical-damage region, respectively. In this way, we can distinguish whether or not other types of charge carriers (for example H^+ ion) other than electrons take part in the charge transport process and contribute to the observed optical damage. Figure 3 shows the measured infrared absorption spectrum for the 0.8mm $LiNbO_3$:Fe crystal sheet. It is seen that the OH^- absorption bands of both the optical-damage region and the non-optical-damage region consist of three

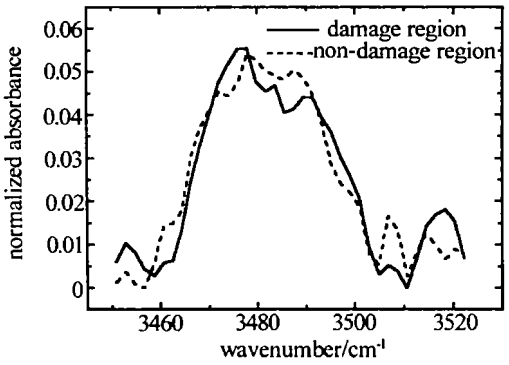

Fig. 3 OH^- absorption spectra for the optical damage region and the non-optical-damage region

absorption peaks as those observed before[9]. However, the main OH^- absorption peak for the optical-damage region shifts to red as compared to that for the non-optical-damage region. The red-shift observed in optical-damage region with respect to the non-optical-damage region indicates that H^+ ions tends to move to the Li vacancies as well as anti-site Nb ions and drive the Nb^{5+} on the lithium sites to its normal sites under the pump light illumination. But no change in the infrared absorption spectra between the optical-damage region and non-optical-damage region was observed in thick samples. Our results clearly show that the movement of H^+ ions during light pumping plays an important role in the long dark decay time of the optical damage observed in the thin lithium niobate crystals.

We also measured the Raman spectra for the optical-damage region and the non-optical-damage region in order to see whether there is destruction of crystal structure. We studied the E(TO) mode of the Raman scattering under the $y(z-x)y$ configuration and found that the half widths of the scattering band at Raman shift wavenumber of 579.2 cm^{-1}, 320.3 cm^{-1}, 236.3 cm^{-1} and 152 cm^{-1} in optical-damage region are a little narrower than those in non-optical-damage region as shown in Table 2. Since the difference is very small, there is no destruction of crystal structure in optical damage region. And no change in Raman spectra between the optical-damage region and non-optical-damage region was observed in thick samples.

Table 2 The half widths of the Raman scattering band

Raman shift wavenumber/ cm^{-1}		579.2	320.3	236.3	152
half widths (measurement error $\pm 0.5 cm^{-1}$)	optical-damage region	22.7	15.2	10.1	9.9
	non-optical-damage region	22.9	15.5	10.2	9.9

3 Conclusions

In conclusion, our experimental results obviously demonstrate a big different dynamic dark decay of the optical damage between the thin and the thick samples, the optical damage observed here probably resulted from the light-induced movement of H^+ ions. It is different from the traditionally so-called "optical damage" in which electrons rather than H^+ ions are considered to be the dominant carriers which is investigated further. However, the exact mechanism for our observed phenomenon remains unclear now. We guess that the surface charge wave, internal electric charge field and coercive electric force may play roles in it, and one possibility is that the volume leakage of the photovoltaic current in thick crystals is larger than that in thin crystals, consequently the saturated photovoltaic field in thin sample will be much higher than that in thick crystals. In this way, H^+ ions are easier to move back to lower energy level sites for example Li^+ sites in thin crystals due to the higher space charge electric field in thin sample when light pumping, which causes the difference in the dark decay behavior of optical damage in thin and thick LiNbO$_3$:Fe crystals. Even though its mechanism remains uncertain, our observed results definitely show that the thickness of samples greatly influences the optical damage, which makes us to reconsider the mechanism of the optical damage for $LiNbO_3$ and understand the optical damage deeply and correctly. The result will be helpful to microphotonic device applications.

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