

Photorefractive properties of Ru doped lithium niobate crystal

Chang-Hung Chiang^a, Jyh-Chen Chen^{a,*}, Yeeu-Chang Lee^b, Chao-Hung Lin^c, Jenq-Yang Chang^c

^a Department of Mechanical Engineering, National Central University, No. 300, Jhongda Road, Jhongli City, Taoyuan County 32001, Taiwan

^b Department of Mechanical Engineering, Chung Yuan Christian University, No. 200, Chung Pei Road, Jhongli City, Taoyuan County 32023, Taiwan

^c Department of Optics and Photonics, National Central University, No. 300, Jhongda Road, Jhongli City, Taoyuan County 32001, Taiwan

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ABSTRACT

We investigated the photorefractive properties of ruthenium (Ru) doped LiNbO₃ crystal, grown by the Czochralski method using a 532 nm solid-state laser. An examination of the blue light-induced absorption change showed a peak around 530 nm which corresponds to that of the commonly used nonlinear 532 nm solid-state laser. The dark decay time constant of the Ru:LiNbO₃ decreased as the temperature increased, from which it can be deduced that in lithium niobate, the energy level is shallower at the Ru center than that at the Fe center. The erasing time constant of the Ru:LiNbO₃ crystal was short and decreased with the erasing beam intensity. Furthermore, given in these experimental conditions, the sensitivity and dynamic range of the 0.01 mol% Ru:LiNbO₃ crystal were 0.0234 cm/J and 0.75.

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1. Introduction

The use of photorefractive materials for holographic data storage applications has been widely investigated in the past few years. Of all the photorefractive crystals, lithium niobate (LiNbO₃) is the most promising medium for holographic data storage. It has good electro-optical and nonlinear optical coefficients, and can be easily grown to a large size using the Czochralski method, while still retaining excellent optical qualities [1]. However, the photorefractive properties of un-doped congruent LiNbO₃ crystals are often relatively poor. One of the most effective methods to enhance performance is to add a small amount of transition metal ions, such as Fe [2,3], Mn [4], Rh [5], Cu [6], Ce [7], and so on, to the LiNbO₃. Of these transition metal ions of dopants, Fe:LiNbO₃ has become one of the most important for holographic storage media, because of its higher diffraction efficiency and photorefractive sensitivity. Nevertheless, more research is still being devoted to investigating which other dopants could improve the photorefractive properties of lithium niobate crystal.

Ruthenium (Ru) and iron (Fe) are both transition elements. Since they are close to each other in the periodic table it is therefore plausible that the effects of Ru ion doping in nonlinear crystals would be similar to those of Fe doping. In recent years, the optical

properties in the visible spectral region produced by the doping of Ru ion into various photorefractive crystals have become a subject of intense study. For example, Ru doping has been found to improve the photorefractive sensitivity of KNbO₃ in the red spectral region [8] and to increase the conductivity of BaTiO₃ [9]. The photorefractive and photochromic properties of Ru doped Bi₁₂TiO₂₀ [10], SBN [11], Bi₄Ge₃O₁₂ [12], and Bi₁₂SiO₂₀ [13] have all been studied, and indeed it has been ascertained that Ru ions could function as photorefractive centers for the regulation of photorefractive properties. However, few researchers have studied the optical and photorefractive properties of Ru:LiNbO₃. Fujimura et al. [14] found some preliminary optical results for 0.18 wt% Ru:LiNbO₃. They indicated that there are deep and shallow energy levels in a single doping Ru:LiNbO₃ crystal, and they succeeded in achieving nonvolatile holographic recording at a wavelength of 632.8 nm using 457.9 nm pumping light illumination.

Chiang and Chen [15] used the Czochralski method to grow Ru:LiNbO₃ crystals from congruent composition melts with different Ru doping concentrations. An examination of the absorption spectra of their Ru:LiNbO₃ crystals showed two absorption peaks around 370 and 530 nm, that is, within the ultraviolet–visible (UV/VIS) region. They found that as the absorption coefficients increased, with the increase in the Ru concentration, the absorption edges shifted towards longer wavelengths.

It should be noted that one of the key considerations for holographic storage systems is the selection of the recording

* Corresponding author.

E-mail address: jcchen@ncu.edu.tw (J.-C. Chen).

wavelength. Typically, the optimal recording wavelength is the center wavelength of the absorption band corresponding to the photorefractive excitation of the carriers from the donors. The absorption peak of Ru:LiNbO₃ crystals is located just at the 530 nm wavelength, which corresponds to the commonly used 532 nm solid-state laser. This makes Ru:LiNbO₃ crystal very suitable for compact green laser holographic applications and is another reason why the determination of the photorefractive properties of Ru:LiNbO₃ utilizing a 532 nm solid-state laser is very important.

To shed light on this situation, we measured the photorefractive properties of Ru:LiNbO₃ with a two-beam coupling experiment conducted at 532 nm. The dark decay time, diffraction efficiency, sensitivity, and the dynamic range of Ru doped LiNbO₃ crystal were all investigated.

2. Experimental procedure

The Ru doped lithium niobate crystal was grown by the Czochralski method. The Ru doped lithium niobate crystal sample was transparent, crack-free, and pale orange in color. The sample was cut into approximately $15 \times 10 \times 3$ mm³ pieces, with the *c*-axis along the long 15 mm direction. Each side of the crystal was polished prior to the optical experiments. According to inductively coupled plasma mass spectrometer measurements (ICP-MS) (Perkin Elmer, SCIEX ELAN 5000) the Ru concentration in our sample was about 0.01 mol%. The UV/VIS absorption spectra of the sample were measured with a UV/VIS spectrometer (HITACHI U-4100) at room temperature, operated in the 300–800 nm range; the wavelength precision was ± 1 nm.

An examination of the light-induced absorption change shows us the energy levels of the Ru:LiNbO₃ crystals. Lithium niobate crystal usually takes a long time to reach stability during the light-induced absorption process. An effective method of increasing the crystal's response time is to elevate the sample temperature, so we heated the samples to 170 °C in an oven. Shutter controlled Kimmon He–Cd and NEC He–Ne lasers, with wavelengths of 442 and 633 nm, respectively, were used to pump the sample. When the shutter was switched on, the pumping beam would pass through the sample, exciting and redistributing the carriers in the energy levels throughout the conduction band, so that we could probe for the existence of deep and shallow levels. The absorption coefficient of the spectra at a particular wavelength was first measured, after which the pumping beam was turned on continuously for 10 min. The absorption coefficient of the crystal was then measured again, and the original values (i.e., without the pumping beam) were subtracted, to obtain the light-induced absorption change. After the pumping beam was turned off for at least 10 min to let the carriers return to their original state, the experiment for next wavelength was performed. The light-induced absorption change spectrum operated in the range from 350 to 900 nm, with an interval of 10 nm. This procedure was repeated 10 times and the results averaged to obtain the data in order to decrease the tolerance of the light-induced absorption change.

In order to characterize the photorefractive properties of Ru:LiNbO₃, a two-beam coupling experiment was conducted at a wavelength of 532 nm. Fig. 1 shows the experimental two-beam coupling setup. We used a solid-state laser with a wavelength of 532 nm as the recording light source. The total intensity of the laser beam was modulated by being passed through a half-wave plate and a polarizer. It was expanded by a beam-expander in order to illuminate the crystal homogeneously. The laser beam was then split into two beams by a beam splitter. One of these two beams acted as the recording beam, and the other as the erasing beam. The recording beam was also split into two coherent beams

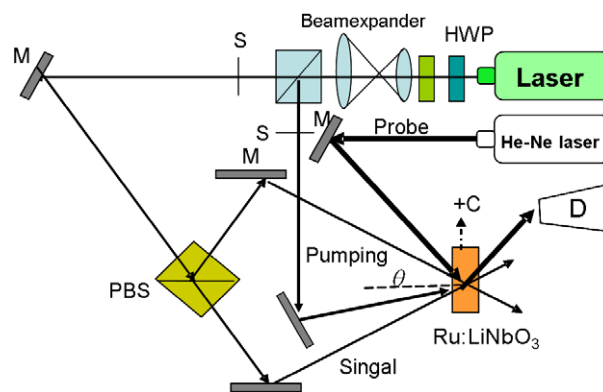


Fig. 1. Apparatus for the two-beam coupling experiment where HWP: half wave plate; M: plane mirror; PBS: polarizing beam splitter; S: shutter; D: silicon detector.

(i.e., the pumping and signal beams) by another beam splitter. Each beam was polarized as an extraordinary ray (E-ray) in order to utilize the large electro-optic component r_{33} . The intensity ratio of the pumping to signal beam was one, and the angle between the two beams was 60° ($\theta = 30^\circ$). The grating period in the crystal was about $0.5 \mu\text{m}$ and the grating vector was aligned along the *c*-axis. A He–Ne laser, with a 633 nm wavelength, was used as the probe beam to monitor the formation and erasure of the gratings in the sample. The intensity of the He–Ne laser was modulated to be below 30 mW/cm^2 . The power variation in the diffractive beam behind the crystal was measured from time to time with a silicon detector. To measure the dark decay, the crystals were placed at the center of a $10 \times 10 \text{ cm}^2$ hotplate. The temperature at the center of the hotplate surface was monitored by a thermocouple and was controlled from room temperature to 125 °C within 1 °C accuracy. The recording and dark decay were controlled by a shutter mounted in front of the 532 nm laser. When the shutter was switched on, the recording process was initiated. When the shutter was switched off, only the He–Ne laser functioned to probe the grating decay in the sample.

We also measured the performance (such as the diffraction efficiency, sensitivity, the dynamic range, etc.) of the Ru:LiNbO₃ crystal for holographic storage at room temperature. Shutters were used to control the recording and erasing status. The recording process was initiated by allowing both the pumping and signal beams to pass through the crystal to form a grating. After the diffraction efficiency reached saturation, only the erasing beam (which was of equal intensity to the recording beam) illuminated the crystal, and the grating in the crystal was gradually erased. Through this two-beam coupling examination we were able to study the dark decay time, diffraction efficiency, erasing time constant, sensitivity, and the dynamic range of the Ru:LiNbO₃ crystal.

3. Results and discussion

A preliminary examination of the absorption spectra helped to determine the optical properties of the crystals. Fig. 2 shows the absorption spectrum in the UV/VIS region of the Ru:LiNbO₃ crystal. According to other researchers [14,15], there are two absorption peaks around 370 nm and 530 nm within the UV/VIS region of Ru:LiNbO₃ crystals. However, in our Ru doped sample, the Ru concentration was lower, and there was no clear absorption peak in the measurement region. In addition, we observed that the absorption coefficients of Ru:LiNbO₃ were relatively small when the wavelength was longer than 600 nm. This indicates that the photoconductivity may be greater for short wavelengths, such as 532 nm, than for long wavelengths, such as 633 nm. The absorption coefficient at 530 nm was about 0.84 cm^{-1} .

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