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Quadratic nonlinear optical parameters of 7% MgO-doped $LiNbO_3$ crystal

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ABSTRACT

Pure and 7% MgO-doped lithium niobate ($LiNbO_3$) single crystals were grown by the Czochralski technique. The shift of optical absorption edge in 7% MgO-doped crystal in direction of shorter wavelength compared to undoped crystal was observed. The second harmonic generation measurements of 7% MgO-doped $LiNbO_3$ crystal were performed at room temperature by means of the rotational Maker fringe technique using Nd:YAG laser generating at 1064 nm in picoseconds regime. Experimentally obtained value of nonlinear optical coefficient d_{33} for 7% MgO-doped $LiNbO_3$ was found to be less than for undoped crystal but higher than for 5% MgO-doped. *I*-type phase-matched second harmonic generation was achieved and the value of phase-matched angle was calculated. High quadratic nonlinearity together with tolerance to intensive laser irradiation makes 7% MgO-doped $LiNbO_3$ crystal interesting for application in optoelectronics.

1. Introduction

Ferroelectric crystal of lithium niobate ($LiNbO_3$) is perhaps the basic material of modern integrated optics and acousto-electronics. It is caused primarily by high values of electric, acoustic and nonlinear optical (NLO) coefficients, and well-tailored industrial production of single crystals. Lithium niobate possesses valuable properties, including non-hygroscopicity and high hardness, which permit high quality surface processing. Not less important is the stability of the crystal to mechanical stress and rapid changes of temperature. The disadvantages include the strong dependence of the refractive indices on the crystal quality and temperature [1].

In recent years, a class of functional and digital integrated-optical schemes, such as spectrum analyzers, phase and amplitude modulators, and sensors were designed on the base of lithium niobate crystal [2]. Thanks to high nonlinear optical parameters the waveguide frequency conversion devices based on second harmonic generation, sum and difference frequencies, as well as parametric light generators, infrared (IR) range lasers were realized [2].

Lithium niobate is characterized by nonstoichiometric composition with a high concentration of internal defects which can be compensated with the addition of foreign impurities such as Mg, Zn, In, Sc that increase the stability of the crystal to laser irradiation [3]. Nonlinear optical properties of pure and doped with MgO till 5% crystals of lithium niobate today are well understood [4–9]. However, lithium niobate crystal with 7% MgO amount is poorly known in terms of nonlinear properties despite the fact that Sen et al. [10] attempted to study optical second harmonic generation (SHG) in this crystal, their investigations are rather qualitative. Meanwhile the study of Kang et al. [11] is concentrated mainly on investigation of electro-optic coefficients. Current paper presents the results of the experimental determination of quadratic nonlinear optical parameters of 7% MgO doped lithium niobate crystal.

2. Experimental details

The samples for investigation were made from congruent $LiNbO_3$ single crystals grown at SRC "Carat" (Lviv, Ukraine) by the Czochralski technique. Magnesium oxide was added to the melt of $LiNbO_3$ to prepare a doped crystal nominally containing 7 mol.% of MgO. The growth technology is described in details elsewhere [12–14]. The *x*-cut plates of $LiNbO_3$ and $LiNbO_3$:MgO (7 mol.%) were prepared with the thickness of 1 mm. The single crystals were polished in order to obtain high optical quality samples. The crystalline orientation was verified using optical conoscopy.

The study of the absorption spectra was carried out in the near ultraviolet region (300–400 nm) using monochromator ZMR-3. The

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optical absorption edge of the crystals was observed in this region of wavelengths. The hydrogen–deuterium lamp was used as a source of light.

SHG measurements were carried out by means of the rotational Maker fringe technique in the transmission scheme for the *s*-polarized fundamental beam (Fig. 1). A *y*-cut crystalline quartz plate has been used as a reference material for SHG measurements. As a fundamental beam, we used the output beam of a Q-switched Nd doped yttrium aluminium garnet laser (model: Quantum Elite) generating at $\lambda = 1064$ nm with 16 ps pulse duration and 10 Hz repetition rate. The incident polarization was selected with a half-wave plate and polarizer in front of the focusing lens. The beam was focused onto the sample with lens of 250 mm focal length. The beam diameter was 0.4 mm at the sample and the applied laser beam intensity was in the range of 2–3 GW/cm^2. A motorized rotation stage with the mounted sample allowed the variation of the incidence angle with a resolution of 0.5°. After passing the sample the KG3 filter was used for cutting of infrared laser beam. The interference filter at 532 nm was used to select the desired wavelength of light. Detector saturation was prevented using linear neutral density filters, whose transmittance value was taken into account during data fitting. The polarization of second harmonic was controlled by polarizer placed before the photomultiplier. The second harmonic signal was detected by the photomultiplier tube (model: Hamamatsu), which was connected to a boxcar and processed by a computer. A portion of the input beam was selected and measured by a fast photodiode (4) to monitor the input energy. Another photodiode (3) was enabled in laser pulse synchronization. Finally, we got the angular dependences of SHG or so called Maker-fringes by rotating the sample towards the normal. Fig. 2 shows the input–output polarization schemes and crystal optical axis orientation in the experimental setup for investigation of second harmonic generation. Such schemes were chosen for determination of d_{33} NLO coefficient and phase matching angle.

3. Results and discussions

3.1. Optical absorption edge in pure and 7% MgO-doped lithium niobate crystals

Lithium niobate crystal is optically transparent in the visible spectrum with a distinct absorption edge in the ultraviolet region. The optical absorption spectra of undoped and 7% MgO-doped lithium niobate crystals obtained in polarized light at room temperature are shown in Fig. 3.

The optical absorption edge of undoped $LiNbO_3$ crystal is observed at 325 nm while absorption edge of MgO-doped crystal is shifted in direction of shorter wavelength and for 7% MgO-doped $LiNbO_3$ crystal is located at 315 nm. The shift of absorption edge in direction of shorter wavelengths in $LiNbO_3$:MgO is caused by decrease of defects density – interstitial centers (Nb_{Li}), in this way crystal structure approaches towards stoichiometric composition [15]. MgO doping of lithium niobate can extend the working range of wavelengths for SHG in the short-wavelength region.

3.2. Second harmonic generation in pure and 7% MgO-doped lithium niobate crystals

Lithium niobate crystal possesses trigonal symmetry (point group 3 m) with lattice parameters: $a = 5.15052$ Å and $c = 13.86496$ Å and belongs to negative uniaxial crystals ($n_o > n_e$) with a range of optical transparency: 0.4–5 μm [2]. Dispersion dependence of refractive indices n_o and n_e, obtained at 293 K for a single crystal of lithium niobate grown from congruent melt (Li/Nb ~ 1.0) is presented in [16]. Lithium niobate possesses four non-zero nonlinear coefficients: d_{22}, d_{15}, d_{31} and d_{33}, and when the Kleinman symmetry condition is fulfilled the coefficients d_{15} and d_{31} are equal between themselves. Lithium niobate crystal is used for SHG of YAG:Nd laser beams, however, possesses disadvantages such as photorefractive effect (changing of refractive index under the action of light radiation) and necessity of thermal stabilization of the crystal. To substantially reduce the impact of photorefractive effect in SHG the MgO doped (4.5 mol.% or more) $LiNbO_3$ crystals are applied. This can increase the conversion efficiency up to 50% [17]. Therefore, the investigation of $LiNbO_3$ crystal doped with a higher concentration of MgO is of great importance.

The angular dependences of SHG intensity observed in $LiNbO_3$ and $LiNbO_3$:MgO (7%) are shown in Fig. 4. The *x*-cut plate of these crystals was rotated around *c*-axis in *s*-polarized laser beam (as shown in Fig. 2a). The rotation of the crystal around the optical axis does not permit to achieve phase matching, there are only peaks in the intensity of the second harmonic which have an interference nature and appear when the optical path of the laser beam in the crystal is multiple to coherence length in a given direction. In asynchronous interaction the periodic transfer of energy from the incident wave to generated one and vice versa takes place.

According to the theoretical model which describes the process of SHG [18,19] the experimental SHG data were fitted and the quadratic NLO coefficients were calculated. The applied scheme of experiment allowed to determinate tensor component d_{33} of NLO coefficient. The values of refractive index used in calculations were taken from the data of $LiNbO_3$:MgO crystal investigation [20]. The obtained values of d_{33} of $LiNbO_3$ and $LiNbO_3$:MgO (7 mol.%) crystals together with other author's results are given in Table 1. Nonlinear optical coefficient d_{33} of pure lithium niobate crystal is

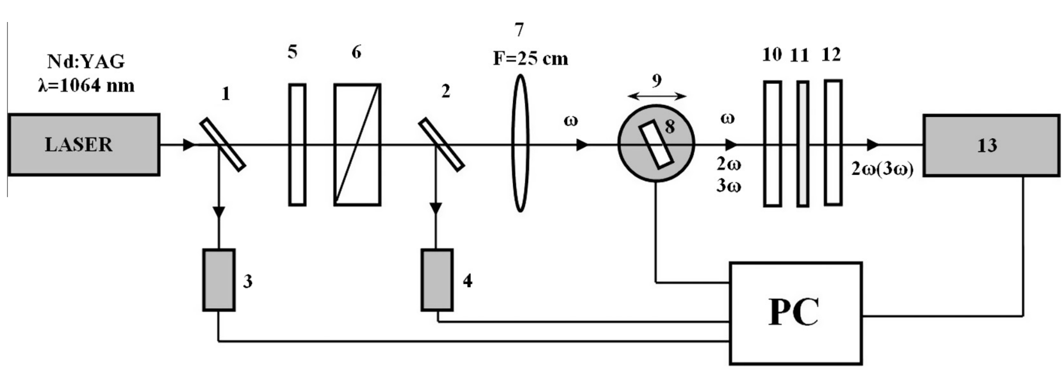

Fig. 1. Experimental setup for the SHG measurements: (1, 2) the beam splitters, (3, 4) the photodiodes, (5) the half wave plate, (6, 12) the polarizers, (7) the lens, (8) the sample, (9) the rotation stage, (10) the filter/s, (11) the interference filter, and (13) the photomultiplier tube and (PC) personal computer.