RAMAN AND PUMP-PROBE STUDY OF E(TO) POLARITONS OF Zn-DOPED $LiNbO_3$

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Abstract: In this report, we present the Raman and pump-probe studies of E(TO) phonons of Zn doped $LiNbO_3$ crystals. We found the the dephasing time of E(TO) polaritons are strongly correlated to the Raman phonon width.

Several techniques have been used to address the propagation and damping of phonon-polaritons[1-3] The pump-probe experiments have been used to detect E-symmetric polariton modes in uniaxial crystals [4-5], and characterized polar and nonpolar phonons in dielectrics, semiconductors, and metals. We investigated the E(TO)-like polaritons of Zn-doped $LiNbO_3$ single crystals measured by the femtosecond pump-probe experiments. The obtained frequencies are compared with the theoretical dielectric dispersion calculations, and the decay time of these polaritons were also found. This result also provides the possible substitution mechanism of Zn atoms incorporated into the $LiNbO_3$ crystals.

Samples were congruent grown single-crystals with various amount of ZnO added. The Zn-doping concentration is from 0 up to 8.3 mol. %. Samples were z-cut (i.e. c-axis is on the sample surface) which is designated to the E(TO)-like polaritons measurement, and both sides are polished and thinned to about 300μm~390μm thick. In the experiment, a Kerr lens mode-locking Ti:Sapphire femtosecond laser is used as the light source. The center wavelength is at 800 nm, and the pulse duration is 58±6 fs according to the autocorrelation measurement. The polarization of pump is along <101> direction, (i.e. 45° off the *c*-axis) to launch E(TO)-like polaritons at zero delay time. Meanwhile, the probe is set to be p-polarized (i.e. along the c-axis). The pump-induced electro-optical (EO) signal in the transmitted probe is collected in the s-polarization with a silicon PIN photodiode. Phase-sensitive lock-in technique is employed to measure the probed EO signal as a function of the delay time between the pump and the probe.

In Fig. 1, the Fourier transform (FT) analysis of the temporal signals as function of frequency is plotted, and three peaks at 3.0±0.1, 4.0±0.1 and 7.1±0.1 THz are clearly observed. These peaks are assigned to the linear combination of three lowest frequencies E(TO)-like phonon-polaritons, ν_1, ν_2 and ν_3. The deduced frequencies of the three lowest E(TO) phonon-polaritons ν_1, ν_2 and ν_3 are compared with the theoretical dielectric dispersion calculation. In Fig. 2, the phonon-polariton dispersion curve of $LiNbO_3$ are expressed in solid lines, and the phase matching angle θ=0° and θ=2.4° of our experimental setup is represented by dot lines. The frequencies found in Fig. 1 are consistent with the experimental prediction of phonon-ploarition dispersion, see the solid dots indicated in Fig. 2. The decay time (τ_1) of the E(TO_1)-like phonon-polariton was determined by the damping of $2\nu_1$ oscillations, i.e. the vibrations around 7.1 THz. In Fig. 3, the decay time of the E(TO_1)-like polaritons as function of Zn-doping concentration is presented, meanwhile, the measured linewidths of E(TO_1) phonons are also plotted for comparison. The dephasing time varies from (2.1±0.03) ps to (1.3±0.01) ps for concentration from 0 up to 8.3 mol. %, while the variations of E(TO_1) phonon widths are from 8 to 16 cm^{-1}. It is generally expected that the phonon width

increase as the phonon lift time decrease. The result found in Fig. 3 is consistent with the expectation.

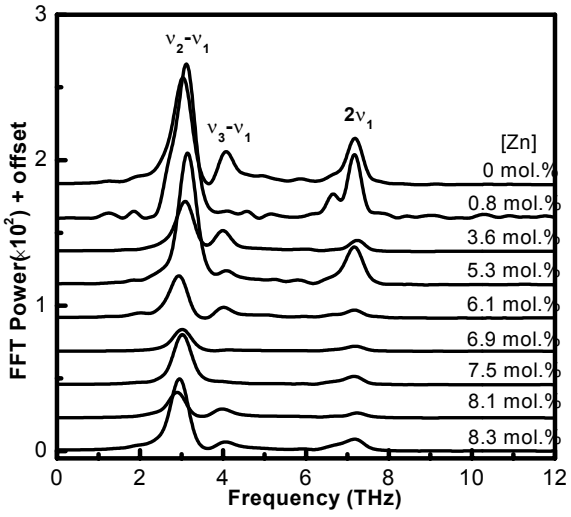

Fig.1 The FT from the temporal response time-resolved pump-probe experiment

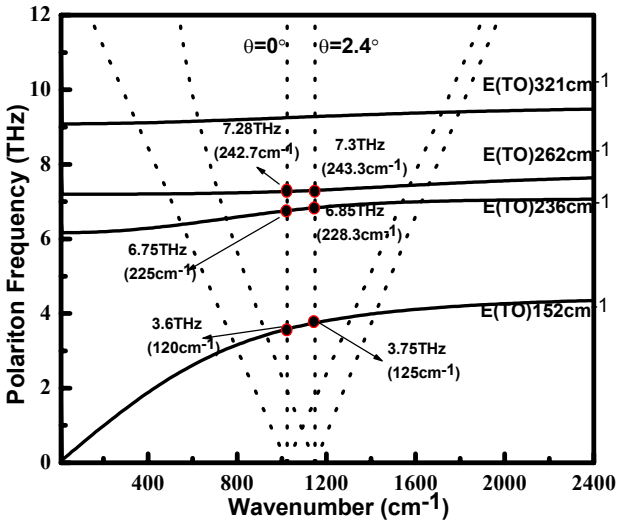

Fig. 2: Phonon-polariton dispersion curves of E(TO)-mode for $LiNbO_3$. The frequencies of phonon-polaritons that can be measured by our setup are indicated as solid dots.

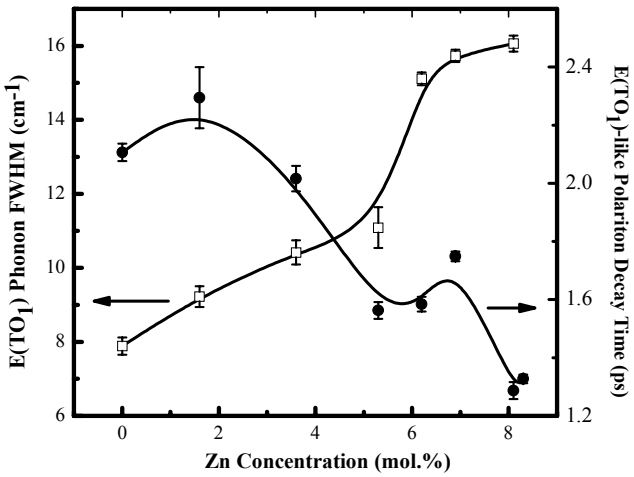

Fig.3 The variations of $E(TO_1)$ FWHM and polariton dephasing time.

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