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Pulsed laser deposited barium titanate thin film for tunable optical delay application

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ABSTRACT

The thin film of barium titanate was fabricated by pulsed laser deposition (PLD) technique. Tetragonal phase in the film has been confirmed by XRD and laser Raman spectra. The film thickness estimated from the analysis of the fringes observed in UV–visible spectra was ~3.3 μm. Barium titanate thin film was subjected to Optical delay studies via degenerate two-wave mixing at room temperature, using 200 ns pulse of 2nd harmonic of Nd:YAG laser. Sub as well as superluminal pulse propagation has been observed by tuning the film orientation. The optical delay has been compared for *BTO* crystal and thin film.

1. Introduction

Barium titanate (BTO) is one of the most promising ceramic materials for nonlinear optics (NLO) and electro-optic applications [1–5]. It has very high value of electro-optic coefficients, fast response time and excellent phase conjugate reflectivity [1,2]. Its tetragonal perovskite phase, at room temperature, shows various nonlinear effects such as second harmonic generation, two-wave mixing, four-wave mixing, slow and fast light etc. [6–11]. Slow and fast light has tremendous scope in optical data communication and quantum computation [12]. Slow light enhances the nonlinear effect and hence nonlinearity can be observed at very low power of laser. Thus reducing the cost of high power lasers and prohibits photo bleaching of optical components.

Slow and fast light has been experimentally observed in photorefractive crystals at room temperature via degenerate and non-degenerate two-wave mixing (pump-probe techniques) [8–13]. The effect of pump intensity, pulse width, external electric field, frequency detuning etc. on group delay and corresponding group velocity has been reported recently in these crystals [8–11]. Light speed reduction to 0.025 cm/s is observed in photorefractive barium titanate crystal via degenerate two-wave mixing using Gaussian pulses [8]. Slowing down of light via degenerate two-wave mixing using chopped pulses is also reported in Ce:$BaTiO_3$ crystal [13]. Since both slow as well as fast light is experimentally demonstrated in BTO crystal at room temperature, hence it can be used to make all optical delay line. To implement the technique of optical delay in devices, the medium should be miniaturized in the form of thin film. The deposited thin film should possess the tetragonal perovskite phase in order to observe nonlinear optical effects. The film thickness is also one of the important parameter for slowing down studies because the time delay is directly related to the effective path length traversed by the signal beam in the medium. The time delay T_{del}, experienced by an optical pulse in passing through a material system of length L is given by equation (1) [14].

$$v_g = \frac{L}{T_{del}} \quad (1)$$

where, v_g is the group velocity inside the medium.

In the present work, the delay and advancement in light pulse via degenerate two-wave mixing is reported in PLD thin film of barium titanate using 200 ns (FWHM) pulse of 2nd harmonic of Nd:YAG laser.

2. Experimental details

Barium titanate thin film was grown via PLD using barium titanate pellet as target. The pellet was prepared from BTO

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powder (99% purity), by pressing it into circular shape of diameter 13 mm using a pellet maker and then sintered at 1400 °C for 2 h. The experimental set-up for film growth via PLD is similar to that of reported earlier [15]. A second harmonic of Q-switched Nd:YAG laser (Quanta system model no. INDI-HG), pulse duration of ~8 ns, was focused on to the barium titanate target mounted inside the ablation chamber. The focusing of such high fluence ~10 J/cm^2, results in the formation of plasma of the target material, which expands and was deposited in the form of thin film on a substrate placed at a distance of 4 cm away from the target. The deposition was performed in the presence of oxygen gas at ~0.1 mbar, at a substrate temperature of 750 °C. Thin film sample was deposited on quartz substrate for 6 h and then annealed in a furnace at 1000 °C for 2 h. The annealed film was subjected to X-ray diffraction (XRD), Raman and UV–visible spectroscopy studies for characterizations. The nonlinear coefficients of the BTO thin film were estimated using *Z*-scan technique. The *Z*-scan setup assembled to estimate the nonlinear coefficients of the film is shown in Fig. 1. The laser beam coming from CW He:Ne laser was focused with a 25 cm focal length lens onto the BTO film. The laser wavelength was ~633 nm. The BTO thin film was mounted on the motorized translational stage. The stage was moved from +*z* to −*z* through 0 in a step of 1 mm. The transmitted beam from the sample was detected onto the photodiode and the corresponding power was displayed on the energy meter with and without aperture for every sample position.

The optical delay has been studied in the BTO thin film via degenerate two-wave mixing using the experimental setup shown in Fig. 2. The two coherent degenerate waves were derived from 2nd harmonic of Nd:YAG laser (Quanta system model no. INDI-HG) having FWHM of 200 ns, operated in the long pulse mode. The laser beam was S-polarized. The beam coming out from the laser was steered by 90° prism, and then divided into two beams by a 50/50 non polarizing cube beam splitter (BS1). The direct transmitted beam from BS1 falling onto the thin film sample, acts as a pump beam. The reflected beam from BS1 steered by mirror M2 was further divided into two beams with the help of another 50/50 cube

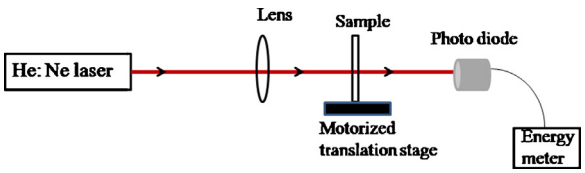

Fig. 1. Schematic of experimental setup for studying *Z*-scan.

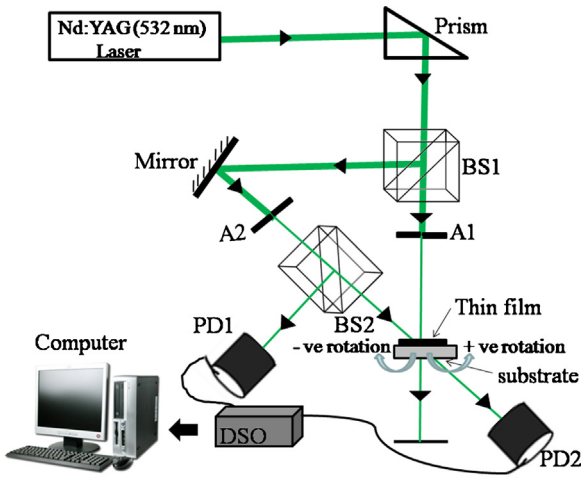

Fig. 2. Schematic of experimental setup for studying optical delay of light via degenerate two-wave mixing.

beam splitter BS2. The reflected beam from BS2 served as a reference beam and was detected by the photodiode PD1 (13DSI001), fed onto one of the channel of digital storage oscilloscope (DSO) (model no Tektronix DPO 4104B). Transmitted beam through BS2 was used as a signal beam and detected by another photodiode PD2 (13DSI001), displayed on another channel of DSO. PD1 and PD2 were placed at equal distances from BS2. The cable lengths for both the photodiodes were equal and the optical path lengths traveled by the pump and signal beam before entering the thin film were kept same. The response time of photodiode was 0.35 ns. The pump and signal beams were made incident on the PLD BTO thin film at a beam crossing angle of 31°. Time delay Δt experienced by the transmitted signal pulse recorded from PD2 was measured w.r.t. the reference pulse. The group velocity of the signal beam was measured by $v_g = L/\Delta t$, where L is the thickness of the film (~3.3 μm) and it is the delay in signal pulse as compared to reference pulse. Initially, the film was placed normal to the pump beam. This was defined as the initial zero position of the film. The film was mounted on a rotational stage and rotated in a step of 5° from its initial position in both the directions, to study the effect of angle of incidence on the propagation of light. The measured average pump intensity was ~63.69 W/cm^2 and that of the average probe intensity was ~25.4 W/cm^2.

3. Result and discussion

3.1. Characterization of PLD deposited BTO thin film

In order to get information about phase, uniformity and structure the PLD BTO thin film has been subjected to optical characterization using XRD, RAMAN and UV–visible studies. Fig. 3 shows the XRD patterns of PLD thin film of BTO. The XRD pattern shows peaks corresponding to (1 0 0), (1 1 0), (1 1 1), (2 0 0) and (2 1 1) planes of BTO [16,17]. Splitting of the (2 0 0) peak shown in the inset of Fig. 3, confirms the formation of tetragonal phase of BTO [18] in the thin film sample.

Fig. 4 shows the Raman bands of PLD deposited BTO thin films. The 308 cm^{-1} E (TO + LO) band called B1 band, corresponding to the tetragonal phase [19] is distinctly observed in the film. The other Raman bands were observed at 183, 272, 520 and 717 cm^{-1}. The 185 cm^{-1} band corresponds to E (TO)/E (LO)/A1 (LO) mode of BTO. The 520 cm^{-1} corresponds to the onset of asymmetry in the E (TO), A1 (TO) mode [19–21]. The 717 cm^{-1} band also corresponds to tetragonal phase in the PLD BTO thin film supporting the XRD data.

The UV–visible spectrum of PLD BTO film shows clear interference fringes as shown in Fig. 5 confirming the uniform growth of BTO film on substrate. The thickness of the PLD deposited BTO film was estimated using envelope approximation [22]. The typical envelope approximation is shown for BTO film in Fig. 5. The inset of Fig. 5 shows the plot of $(\alpha h\upsilon)^2$ vs. $h\upsilon$, where α is the

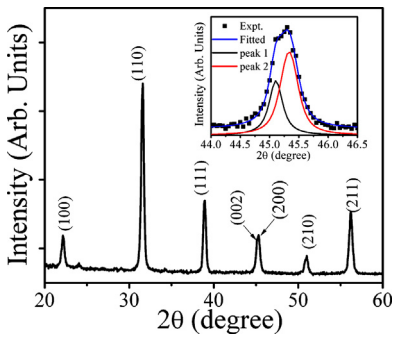

Fig. 3. XRD pattern of PLD BTO thin film. Inset shows deconvolution of (0 0 2) peak.