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Simple transmission ellipsometry method for measuring the electric-field-induced birefringence in PLZT thin films

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Ferroelectric materials have played an important role in the area of non-linear optics. Thin film ferroelectrics are promising for electro-optical applications in integrated optics, such as optical shutter, frequency doublers, and waveguide modulator, etc. Lanthanum-doped lead zirconate-titanate (abbreviated as PLZT) thin films have received considerable attentions recently due to their strong electric-field-induced birefringence.

Several authors have reported various methods for the measurement of electro-optic (E-O) properties of ferroelectric thin films. These include the Senarmont method [1], optical polarization microscope [2], and magneto-optic modulation method [3], etc. Recently an ellipsometry method [4] was proposed based on the measurement of phase retardation between the s- and p waves reflected from thin film surface. This method requires lengthy and complicated numerical method to derive the E-O coefficient. It is essential to establish quick and simple methods in the study of electro-optic properties. Here we propose a transmission ellipsometry method for the measurement of electric field induced birefringence in transparent thin films.

Traditionally, an ellipsometry method measures the ellipsomtric angles Ψ and Δ, which are defined, in a reflection ellipsometry, by the expression [5]:

$$\rho = \frac{r_p}{r_s} = \tan\Psi \exp(j\Delta) \tag{1}$$

where r_p and r_s are the complex Fresnel reflection coefficients for light polarized parallel (p wave) and perpendicular (s wave) to the plane of incidence, respectively. Similarly, in a transmission ellipsometry measurement with the light beam traveling in the direction perpendicular to the principal axes of birefringence, the expression for the complex number ρ can be written as [6]:

$$\rho = \frac{t_p}{t_s} = \tan\Psi \exp(j\Delta) \tag{2}$$

where t_p and t_s are the complex transmission coefficients for the p wave and s wave, respectively. The transmission coefficients are complex functions of the refractive index n and extinction coefficient k and their explicit expressions can be found in [6]. Equation 2 can be further expressed as:

$$\rho = \frac{t_p}{t_s} = \tan\Psi \exp(j\Delta) = \exp\left[-\frac{2\pi d}{\lambda}(k_e - k_o)\right] \times \exp\left[-j\frac{2\pi d}{\lambda}(n_e - n_o)\right] \tag{3}$$

where the quantities $(n_e - n_o)$ and $(k_e - k_o)$ represent the birefringence and dichroism of the medium, and d is the total distance traveled by light beam through the medium.

From Equation 3 it is clear that the ellipsometric angle Δ measured in transmission mode corresponds to the phase retardation between the extraordinary and ordinary waves, i.e.:

$$\Delta = \frac{2\pi}{\lambda} d(n_o - n_e) = \frac{2\pi}{\lambda} d\delta n = \frac{2\pi}{\lambda} d\left(\delta n^{(o)} + \delta n^{(E)}\right) \tag{4}$$

where $\delta n^{(o)}$ and $\delta n^{(e)}$ denote the zero-field (natural) birefringence and electric-field induced birefringence, respectively.

The transmission ellipsometry measurements were carried out with a phase modulated ellipsometer (Jobin-Yvon UVISEL) at a fixed wavelength $\lambda = 633$ nm. The photo-elastic modulator (M) consists of a rectangular fused silica block cemented to a piece of piezoelectric quartz crystal oscillating at a resonant frequency of 50 kHz. This generated a periodic phase shift $\delta(t)$ between orthogonal amplitude components of the transmitted beam. This phase shift has the form of $\delta(t) = \delta_m \sin(\omega t)$, where ω is the modulation frequency (50 kHz) and δ_m the modulation amplitude.

The schematic diagram of the optical system is shown in Fig. 1. The incident light beam, perpendicularly directed to the polarizer (P), whose axis was oriented at 45° of the incident plane, was linearly polarized by the polarizer. After traveling through the modulator (M), sample (S) and analyzer (A), it is detected by the photomultiplier tube (PMT). The orientation of the birefringence axes of M and S with respect to the incidence plane are both 0°. The analyzer axis is also at 45° to the incident plane. All the optical components and the sample are aligned horizontally and centered on the

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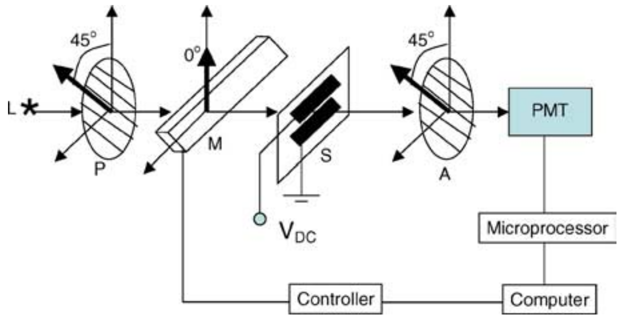

Figure 1 Schematic diagram of the optical setup for electric-field induced birefringence in PLZT thin films using a transmission ellipsometry technique. (L: light source, P: polarizer, M: modulator, S: thin film sample, A: analyzer.)

beam. The measurement parameters are controlled by a computer.

The general expression for the detected light intensity has the form [7]:

$$I(t) = I_o + I_s \sin[\delta(t)] + I_c \cos[\delta(t)] \quad (5)$$

The explicit expression for I_o, I_s and I_c can be found in [7]. For the specific orientation of the optical components such as shown in Fig. 1, i.e., $P = 45^\circ$, $M = 0^\circ$ and $A = 45^\circ$, equations for I_o, I_s, and I_c take the forms:

$$I_o = G \quad (6)$$

$$I_s = G \sin(2\Psi) \sin \Delta \quad (7)$$

$$I_c = G \sin(2\Psi) \cos \Delta \quad (8)$$

where G is a constant. Since $\delta(t) = \delta_m \sin(\omega t)$, a Fourier series expansion of Equation 5 yields the following expression:

$$I(t) = I_o + I_c J_0(\delta_m) + 2I_s J_1(\delta_m) \sin(\omega t) + 2I_c J_2(\delta_m) \cos(2\omega t) + \text{higher order terms...} \quad (9)$$

where $J_v(\delta_m)$ is the Bessel identities of argument δ_m and order v. In Equation 9, the first and second terms correspond to the DC components, while the third and fourth terms correspond to the fundamental and second harmonic components, respectively. By adjusting the modulation amplitude $\delta_m = 2.405$ rd., $J_0(\delta_m) = 0$. The electronic system of the ellipsometry demodulates the harmonic components and measures the ratios of the fundamental and second harmonic components to DC component, R_ω and $R_{2\omega}$. Their expressions can be derived from Equations 6, 7, 8 and 9:

$$R_\omega = \frac{2I_s J_1(\delta_m)}{I_o} = 2J_1(\delta_m) \sin(2\Psi) \sin \Delta \quad (10)$$

$$R_{2\omega} = \frac{2I_c J_2(\delta_m)}{I_o} = 2J_2(\delta_m) \sin(2\Psi) \cos \Delta \quad (11)$$

The ellipsometric angles Ψ and Δ can be computed from Equations 10 and 11, and from the value of Δ, both the natural and electric-field induced birefringence can be deduced. Before conducting the measurement on the thin films samples, verification was first carried out by inserting a quarter wave plate in place of the sample in Fig. 1. The fast (or slow) axis of the plate makes an angle of 45° with the polarizer axis. This will convert the linearly polarized light into a circularly polarized light which has a phase retardation of 90° between the two orthogonal components of equal amplitude. Indeed the ellipsomter measures the angle $\Delta = 90^\circ$(or 270°). This check insures the correctness of our subsequent measurements on real samples.

PLZT (9/65/35) thin films were prepared on a transparent fused-quartz substrate using a sol-gel method. Since the fused-quartz is amorphous, it was found necessary to use a buffer layer to obtain crystalline perovskite structure. A thin layer (~50 nm) of $SrTiO_3$ (STO) was deposited on the fused-quartz substrate using radio-frequency (rf) magnetron sputtering as the buffer layer followed by spin-coating of 6 layers of

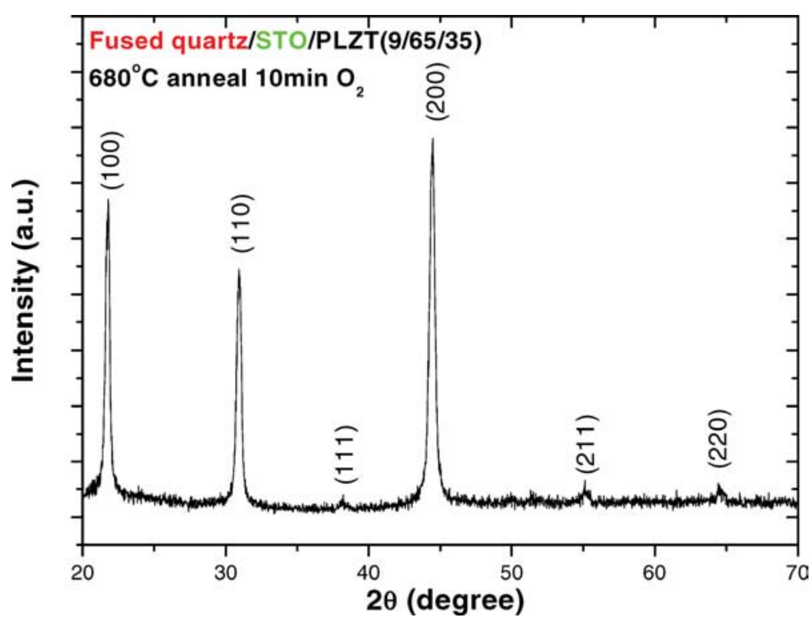

Figure 2 X-ray diffraction pattern of the PLZT (9/65/35) deposited on fused quartz.

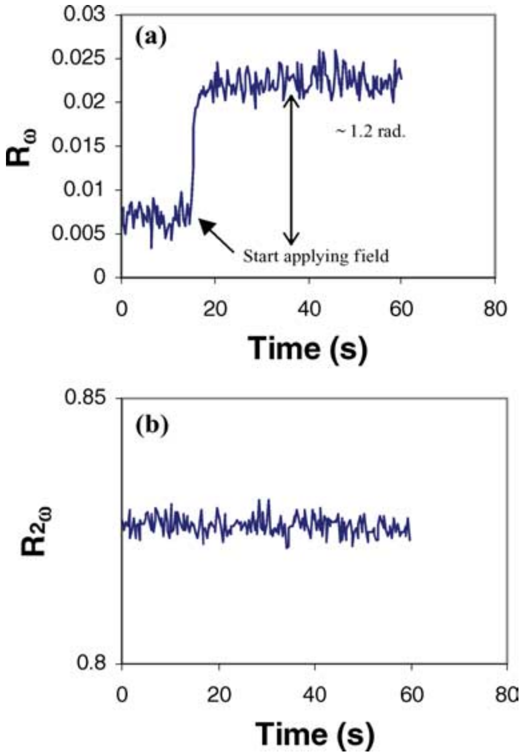

Figure 3 Recorded signals of R_ω and $R_{2\omega}$ during data acquisition. The jump in R_ω is clearly observed after applying an electric field of 100 volts.

PLZT (9/65/35), resulting in a film of ~450 nm thick. Then the PLZT thin film was annealed at 680 °C in O_2 atmosphere for 10 min using a rapid thermal processor (RTP). X-ray diffraction pattern shown in Fig. 2 indicates that the film has a pure perovskite structure with a random orientation. Gold interdigital electrodes (IDE) with a finger spacing of 20 μm were deposited on the PLZT film to form a fuse-quartz/STO/PLZT/Au IDE structure.

The electric field induced birefringence was carried out by measuring the phase retardation as a function of the DC field. Fig. 3 shows a typical measurement result obtained during data acquisition of R_ω and $R_{2\omega}$, with a fixed wavelength of 633 nm of the incident light. From Fig. 3a, a change of R_ω can be clearly seen when a DC field of 100 V was applied to the electrodes. In Fig. 3b it is seen that $R_{2\omega}$ has no change. The corresponding phase retardation Δ can be computed accordingly. For this applied voltage, the induced phase retardation is about 1.2 rd. Thus the electric field induced birefringence δn can be computed according to the relationship $\delta n = (\lambda/2\pi)\Delta$.

Fig. 4 shows the induced birefringence δn as a function of the applied electric field. It is observed that, at zero electric field, the natural birefringence is very small and is almost negligible. The field-induced

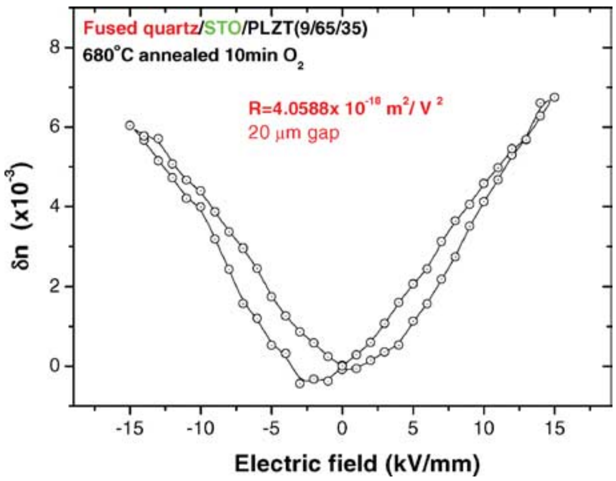

Figure 4 Electric-field induced birefringence in PLZT (9/65/35) transparent thin film with a co-planar electrodes of 20 μm gap.

birefringence obeys a quadratic behavior. The birefringence reaches 6×10^{-3} at the maximum applied field, 15 kV/mm. The value of the quadratic electro-optic coefficient R_{eff} was calculated to be 4.1×10^{-18} m^2/V^2. This result agrees very well with literature report [2] on the birefringence of PLZT (9/65/35) thin film using a transmission confocal optical polarization microscopy.

In conclusion, we have demonstrated a simple method to measure the electric field induced birefringence in ferroelectric thin film by adapting a transmission ellipsometry with a phase modulation technique. The measurement results agree well with the literature report. The merit of the present method lies on its quick set-up and simplicity of measurement.

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