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Measurement of the Pockels Coefficient of PZT Thin Films Using a Two-beam Polarization Interferometer with a Reflection Configuration

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A two-beam polarization (TBP) interferometer with a reflection configuration for measuring the linear electro-optic coefficient is described and investigated experimentally and theoretically. It is shown that a TBP interferometer can be used for measuring the Pockels coefficient of thin film with a strong Fabry-Perot effect. The TBP interferometer technique is used to measure the effective differential linear electro-optic coefficient $r_e = r_{33} - (n_0/n_e)^3 r_{13}$ of lead zirconate titanate (PZT) thin film. The results are in agreement with known data.

Key words: TBP interferometer, Linear electro-optic coefficient, PZT thin film

I. Introduction

Ferroelectric thin films have received recent attention for applications such as microelectromechanical systems (MEMS), actuators, optical switches, and electrooptic modulators (EOM). For applications of thin films determination of their parameters is of great importance.

In the simple reflection technique proposed by Teng and Man[1] as well as independently by Schildkraut,[2] the single-beam polarization interferometer was used for measuring the linear electro-optic coefficient of poled polymer films. This technique was analyzed in detail and applied to measure the r_{33} Pockels coefficient of poled polymer films in.[3-8] Those measurements were done under conditions where the influence of the Fabry-Perot (FP) effect may be disregarded. In reality, however, the FP signal originating from the interference between the beams reflected from two sides of the film is already present in thin films without special antireflection coating.

The purpose of this paper is to present the new two-beam polarization (TBP) interferometer with reflection configuration for thin film testing where the FP effect is clearly in evidence and to apply it to the electrooptic characterization of $Pb(Zr,Ti)O_3$ thin films. Two different ways for p- and s- polarized beams are used in TBP interferometer that provides the possibility to measure Pockels coefficient for thin films with strong FP effect.

II. Analysis

In our analysis we consider the linearly polarized laser beam E_* (Fig. 1) transmitted through an isotropic substrate, a transparent electrode, and thin film, and reflected from the top (1) and the bottom (2) surfaces of the thin film. The phase difference ϕ_{2sp} between s- and p-polarized waves transmitted through film and reflected back from bottom (2) surface of the film is given by[4]

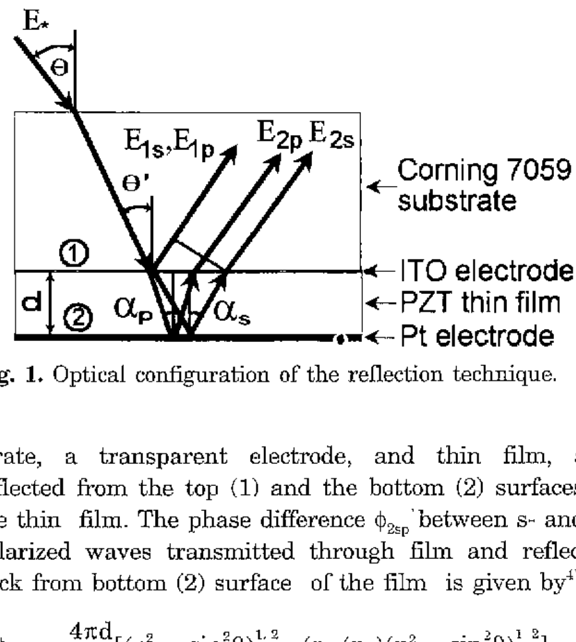

Fig. 1. Optical configuration of the reflection technique.

$$\phi_{2sp} = \frac{4\pi d}{\lambda}[(n_0^2 - \sin^2\theta)^{1/2} - (n_0/n_0)(n_e^2 - \sin^2\theta)^{1/2}] \tag{1}$$

where d is the thickness of the film, θ is the incidence angle of the laser beam, n_0 and ne are the ordinary and extraordinary refractive indices of the thin film. and l is the laser wavelength.

The basic idea in polarization interferometry with the reflection configuration is to use the electric-field-induced phase-shift difference between the p- and s-polarized beams transmitted through electro-optic film and reflected back from opposite surface of thin film. For poled film, the phase change under the variation of electric field dE is given by

$$\delta\phi_{2sp} = (\partial\phi_{2sp}/\partial_{n0})\delta_{n0} + (\partial\phi_{2sp}/\partial_{n_e})\delta_{n_e} + (\partial\phi_{2sp}/(\partial d))\partial d \quad (2)$$

where the electric-field-induced refractive-index and thickness variation is given by

$$\delta n_0 = -(n_0^3/2) r_{13}\, \delta E$$
$$\delta n_0 = -(n_0^3/2) r_{33}\, \delta E \quad (3)$$
$$\delta d = d\, d_{33}\, \delta E$$

where r_{13} and r_{33} are Pockels coefficients, and d_{33} is a piezoelectric coefficient.

When we apply the dc and small amplitude ac field $E = E_0 + E_m \sin\omega t$ across the thin film, the electric field variation is equal to the ac field amplitude $\delta E = E_m$. Changing the dc field with fixed ac field amplitude E_m, we obtain the dependence of the phase change versus the dc field, E_0. Using Eqs. (1) and (3) to calculate the derivatives in Eq. (2) and taking into account that the birefringence of the usual PZT thin film does not exceed $5*10^{-2}$, we arrive finally at the following expression for the phase change[9]

$$\delta\phi_{2sp} = (2\pi d r_e E_m/\lambda)[n^2 \sin^2\theta/(n^2 - \sin^2\theta)^{1/2}] \quad (4)$$

where $r_e = r_{33} - (n_0/n_e)^3 r_{13}\ {}^a r_{33} - r_{13}$ is the effective electro-optic coefficient[10] and $n = n_0$. The final expression for the phase change depends only on the electro-optical property but not on the piezoelectric property of the thin film. This is so because we can neglect the term related to the thickness variation dd for conventional values of the parameters of a PZT thin film Let us consider now the interference patterns that can be created by beams reflected from thin-film surfaces in the TBP interferometer setup (Fig.2). A laser beam from the standard cw He-Ne laser (λ=633 nm, P_0=2 mW) linearly polarized at an azimuth of ~45°C relative to the plane of incidence is transmitted through a Corning glass 7059 substrate, an indium tin oxide (ITO) transparent electrode, and PZT thin film and is slightly focused onto the Pt electrode deposited upon the bottom surface of the PZT thin film. After it is reflected from the electrode, this beam is separated into p- and s-polarized beams by a polarized beamsplitter (PBS). After reflection from mirrors, servo-mirror and a non-polarized beamsplitter (BS), these beams are projected onto the photodiode where they mix again. A servo mirror with a feedback system is used for movement and stabilization of the operation point of the TBP interferometer. The incident angle of the linearly polarized incident beam are set at 45°C in our experiment. We disregard multiple reflections, and the dependence of the output signal on the incident angle is not analyzed because, although the influence of multiple reflections is important in studies inside the absorption range of materials,[7-8] we use laser radiation with wavelength λ= 633 nm, and thus operate in the transparency range of PZT thin film.

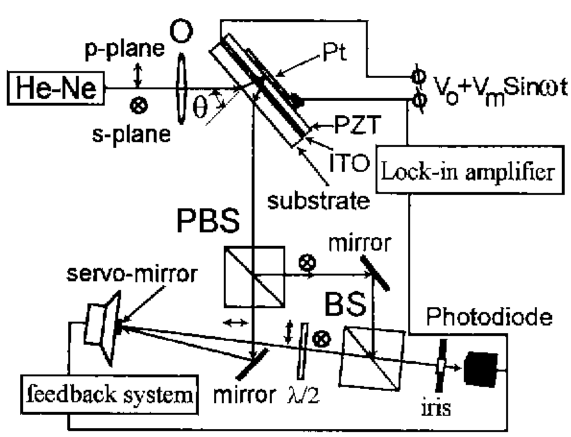

Fig. 2. Experimental setup for PZT thin film testing.

In our analysis we consider three types of interferometers. When an interference pattern is created by all four beams with s- and p-polarization state reflected from the top (1) and bottom (2) surfaces of the thin film (see Fig.1), we define this interferometer as a TBP interferometer. In the TBP interferometer optical setup we use a half-wave retardation plate for rotating the polarization state of p-polarized beams reflected from both surfaces of the thin film to the polarization state of the s- polarized beams. Thus, the output intensity of the TBP interferometer is given by:

$$I_{TBP} = |E_{1s} + E_{1p} + E_{2s} + E_{2p}|^2/2 \quad (5)$$

where E_{1s}, E_{1p}, E_{2s}, E_{2p} are the s- and p-polarized beams reflected from surfaces (1) and (2) respectively, are given by

$$E_{1s} = R_1 E^* \exp(-i\varphi_{1s})$$
$$E_{1p} = R_1 E^* \exp[-i(i\phi_{1p} + \psi)]$$
$$E_{2s} = R_2 E^* (1 - R_1^2) T \exp(-i\phi_{2s}) \quad (6)$$
$$E_{2p} = R_2 E^* (1 - R_1^2) T \exp(-i(\phi_{2s} + \psi))$$

where ϕ_{1s}, ϕ_{2s}, ϕ_{2s}, and ϕ_{2p} are the phase shifts of the s- and p-wave components reflected from the top (1) and bottom (2) surfaces of the film, R1 and R2 are the reflectivity of the top (1) and bottom (2) surfaces of film, respectively. T is transmittance of the thin film for two passes, E^* is the incident light amplitude, and y is the additional phase shift due to different optical paths for p- and s- polarized beams.

Without a half-wave retardation plate the interference pattern is created only by the beams with the same polarization state ("s" or "p") reflected from different surfaces of the film and therefore we define this interferometer as an FP interferometer. The output intensity of the FP interferometer is given by:

$$I_{FP} = |E_{1s} + E_{2s}|^2/2 + |E_{1p} + E_{2p}|^2/2 \quad (7)$$

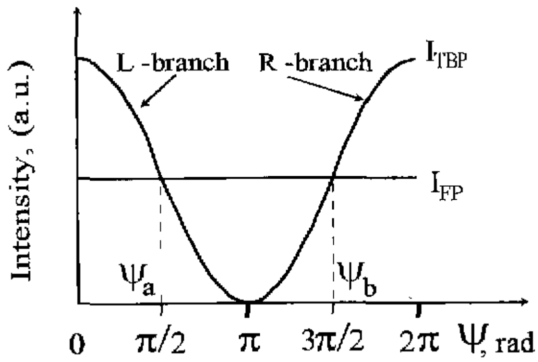

Fig. 3 Output interferometer intensity with a half-wave retardation plate (I_{TBP}) and without it (I_{FP}) versus an additional phase shift y between s- and p-polarized beams. L, left; R, right

If the interference pattern is created only by s- and p-polarized beams reflected from the bottom (2) surface of the thin film, we can define this interferometer as a 2SP interferometer, and the output intensity of the 2SP interferometer can be expressed as:

$$I_{2SP} = |E_{2s} + E_{2p}|^2/2 \quad (8)$$

Note that in contrast to TBP and FP interferometers which can actually be produced the 2SP interferometer is simply a speculative model defined by expression (8).

Figure 3 shows the dependence of output intensity I_{TBP} and I_{FP} on the additional phase shift y between s- and p-polarized beams, calculated from Eqs. (5)-(8) for the following values of parameters: d=0.5 mm, n=2.5, $d_{33}=50*10^{-12}$ m/V,[11] $r_{33}=1*10^{-10}$ m/V, $r_{13}=0.33*10^{10}$ m/V,[12-13] $R_1=0.001 \div 0.1$, $R_2=0.9$, T=0.9. A feedback system with a servo mirror keeps the overage intensity with a characteristic time of ~1/10 s inside the left (L) or the right (R) operation branch. Using a feedback system we can also change the additional phase shift ψ and therefore the operation point of the TBP interferometer.

When we apply alternating voltage to the thin film we modulate the output intensity of the TBP, FP and 2SP interferometers. When the modulating amplitude is small, the output acmodulated intensity signal at the first harmonic of excitation is proportional to the first derivative of the output intensity of the respective interferometer

$$S_{TBP} = \partial I_{TBP}/(\partial E)$$
$$S_{FP} = \partial I_{FP}/(\partial E) \quad (9)$$
$$S_{2sp} = \partial I_{2sp}/(\partial E)$$

Figure 4 shows the relation of the TBP, FP and 2SP interferometers' signals to the additional phase shift ψ between s- and p- polarized beams, calculated from Eqs. (9). At a low reflectivity $R_1=0.001$, the TBP interferometer signal is approximately equal to the 2SP interferometer signal, and we can disregard the influence of the FP

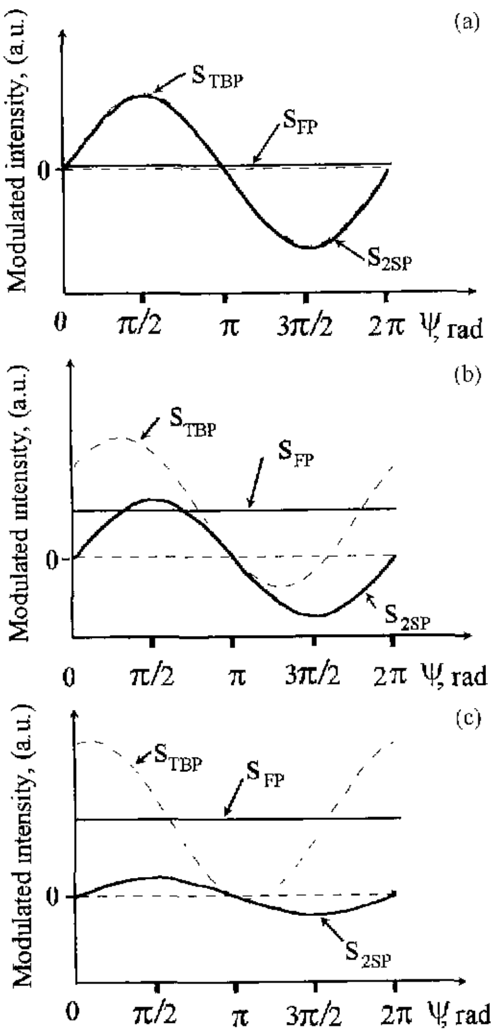

Fig. 4. Modulated output intensity of TBP, FP and 2SP interferometers versus an additional phase shift ψ between s- and p-polarized beams for different values of the top surface of the PZT thin-film reflectivity: 4a) $R_1=0.001$, 4b) $R_1=0.025$ and 4c) $R_1= 0.1$

interferometer (Fig 4a). But at higher reflectivity $R_1=0.025$ (Fig 4b) and R1=0.1 (Fig 4c) the 2SP interferometer signal S2SP must be calculated as the difference between TBP- and FP- interferometer-signals for center points of the right or left operation branch

$$S_{2SP}(\psi_a)=S_{TBP}(\psi_a)-S_{FP}(\psi_a)=-[S_{TBP}(\psi_a)-S_{FP}(\psi_b)] \quad (10)$$

where $\psi_a=\pi/2$ and $\psi_b=3\pi/2$ are the additional phase shifts that correspond to the center points of the left and right operation branches, respectively, of the TBP interferometer.

For estimating errors we introduce the characteristic number F equal to the ratio of the value of the TBP interferometer-signal at yb to its value at ψ_a

$$F=S_{TBP}(\psi_b)/S_{TBP}(\psi_a) \quad (11)$$

When the FP signal influence is weak, the value of F=-1 and the 2SP interferometer signal is equal to the TBP interferometer signal, but when the FP signal is too large, the value of F is F=+1 and the TBP interferometer signal is approximately equal to the FP signal.

Numerical simulations based on relations (5)-(10) show that the difference between the value of the S2SP signal calculated from Eqs. (9) and that found from relation (10) is less than 1.5% for a negative characteristic number F. Finally, the Pockels coefficient can be found from relation (4).

III. Experimental Results

A PZT thin film was deposited five times upon ITO Corning 7059 glass by a spin coating method (2000 rpm, 30 s) with a Pb 1.1(Zr0.52Ti0.48)O3(10% excess Pb) solution made by a metal-organic decomposition (MOD) method. The initial thickness of the ITO was ~0.2 mm, and the substrate size was 20 mm × 20 mm × 1.2 mm. A drying procedure (drying temperature, 300°C ; time duration, 5 min) was applied to the film to remove the retained organics. Afterward, heat-treatment (temperature, 650°C; time duration, 30 min) was applied for the crystallization of the PZT after the third and fifth steps of the deposition process. Finally, the thickness of the PZT thin film was ~0.5 mm. Poling of the PZT thin film was done at 150°C for 10 min by application of 19 v. of electricity.

The experimental setup for the determination of the electrooptic coefficients of PZT thin films is shown in Fig. 2. The sum of the dc and small-ac voltage $V=V_0+V_m\sin wt$ at frequency $\omega/2\pi=1$kHz was applied to the ITO and Pt electrodes of the thin film from a standard signal generator. The output electrical signal was measured by a photodiode with an SR-510 lock-in amplifier (load resistance $R_L=100$ MΩ) at the first harmonic of the excitation frequency. The sensitivity of the system obtained experimentally was about10^{-5} rad, corresponding to 0.001 nm for displacement measurement for the 1 Hz bandwidth .

The measurement of the Pockels coefficient is done in the following way. Only the FP interferometer modulated intensity signal S_{FP} was measured when the half-wave retardation plate was removed. When a half-wave retardation plate was installed, the s- and p-polarized beams reflected from the Pt and ITO surfaces began to create an interference pattern. Moving the operation point of the TBP interferometer by means servo mirror displacement, we installed the operation point of the TBP interferometer in the center of the left or right operation branch. The difference of the TBP and FP interferometer signals gives the value of the 2SP interferometer signal.

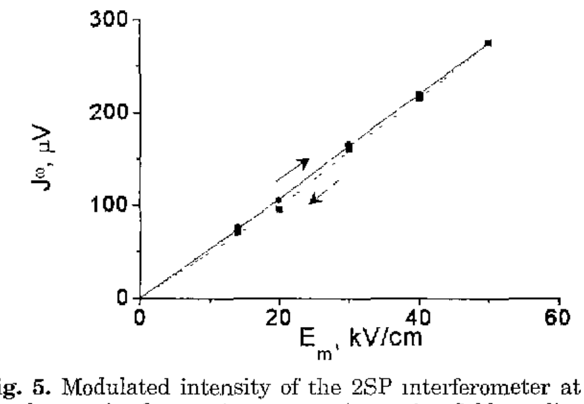

Fig. 5. Modulated intensity of the 2SP interferometer at the first harmonic of excitation versus alternating field amplitude.

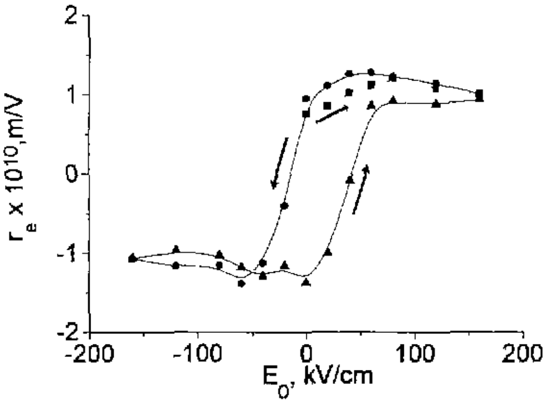

Fig. 6. Dependence of the effective differential Pockels coefficient re on the dc electric field (E_m=20 kV/cm)

First we checked that the measured modulated intensity signal was from a linear electro-optic effect. Figure. 5 shows the 2SP interferometers signal versus the amplitude of the ac field, Em. The linear dependence without strong hysteresis demonstrates that this electro-optic signal is from a linear electro-optic effect.

In calculating the thin film linear electro-optic coefficient we used relation (10) for S2SP estimation and then obtain re from relation (4). Because in our experiment the value of the F coefficient was F=−0.12, the error was less than 1% according to numerical estimation.

We determined the absolute value of the PZT thin film linear electro-optic coefficient by placing an additional servo mirror with a well-known relation between displacement and applied voltage in one arm of the interferometer.

A strong hysteresis effect with a slightly asymmetric form of the hysteresis loop was observed at the dependence of the re versus dc electric field amplitude (Fig. 6). The curve was measured for ac field amplitude E_m=20 kV/cm. There are some reasons for the slight asymmetry, such as the different nature of the electrodes used and a partial clamping of the electromechanical deformation owing to residual stress at the film-substrate interface.[14] We must emphasize that in our technique we measure the differential value of the Pockels coefficient. The differential Pockels coefficient is proportional to the slope of the dependence of birefringence on dc field, and therefore the maximum value of the differential Pockels coefficient may exceed the average value of the Pockels coefficient usually used in the literature. The field-induced birefringence shift is expressed as an integral of the differential

Pockels coefficient:

$$\Delta n(E) = -\frac{n^3}{2}\int_{-E_0}^{E_0} r_e(E)dE \quad (12)$$

where $\pm E_0$ is the range of dc field variation.

The field-induced birefringence, calculated from Eq. (12) is approximately equal to $8*10^{-3}$ for the external field change from 0 to 100 kV/cm. In general, the value of the field-induced birefringence of PZT and lead lanthanum zirconated titanate (PLZT) thin films depends on the film composition, the substrate used, and the deposition conditions. As reported in Ref.[16] the field-induced birefringence of the PZT 0/65/35 thin film deposited onto glass and sapphire substrate is $2*10^{-3}$ and $15*10^{-3}$, respectively, for the field change from 0 to 100 kV/cm. The value of the birefringence obtained in our experiment is located in the center of this interval. The sufficiently high value of the field-induced birefringence in our experiment could be related to relatively high perovskite phase contents and high domain alignment in the direction perpendicular to the film surface for our PZT 52/48 thin film deposited onto ITO-glass substrate by the solgel method.[17]

The measured value of the differential effective Pockels coefficient r_e lies inside the interval between $-1.38*10^{-10}$ m/v and $+1.28*10^{-10}$ m/V for an external dc field changed from −160 kV/cm to 160 kV/cm (see Fig. 7). These values correlate well enough with the average values of the Pockels coefficient of PZT and PLZT ceramics that lie in the interval $(0.4\pm0.2)*10^{-10}$ m/V for different ceramics compositions, and were obtained by use of different techniques. [12,13,15] In this regard, the measured value of the re is in agreement with the known value obtained by other techniques.

IV. Conclusion

We have developed a two-beam polarization interferometer for measuring the linear electro-optic coefficient of a poled PZT thin film. We showed that a two beam polarization technique facilitates measurement of the Pockels coefficients of thin films with the strong Fabry-Perot effect that is usually present in ferroelectric thin films. Based on a theoretical analysis, a simple Pockels coefficient measurement method was proposed. The measured differential effective Pockels coefficient $r_e \approx r_{13} - r_{33}$ of a spin coated PZT thin film demonstrated a strong hysteresis shape with a slightly asymmetric form of the hysteresis loop. The measured value of the differential effective Pockels coefficient re lies inside the interval between $-1.38*10^{-10}$m/v and $+1.28*10^{-10}$ m/V for an external dc field that varied from −160 kV/cm to 160 kV/cm and is in agreement with the known values.

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