Short-time piezoelectric measurements in ferroelectric thin films using a double-beam laser interferometer

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(Received 2 July 2002; accepted 9 December 2002)

An evolution of the double-beam laser interferometer used for piezoelectric measurements in ferroelectric thin films is reported. Measuring the d_{33} hysteresis of a ferroelectric material using lock-in technique with large time constants requires a varying bias field to be applied to the sample over a long period of time. This long-term application leads to electrical stress during the measurement. We present a measurement technique using a different source for the applied field and a varied method for averaging the interferometric response. The measurement time for a complete d_{33} hysteresis will be shortened down to several seconds. Also, the cycle frequency becomes comparable to electrical hysteresis measurements. Experimental results on quartz and $Pb(Zr_{(X)},Ti_{(1-X)})O_3$ are given to demonstrate the capabilities of the interferometer and the new measurement method. [DOI: 10.1063/1.1544415]

Piezoelectric and ferroelectric thin films are used in a wide spectrum of micro-mechanical applications.[1–3] To integrate these thin films and microdevices into integrated circuitry (IC), complementary metal–oxide–semiconductor-compatible deposition methods must be used.[4] Hence, the films are preferable manufactured by low temperature deposition and micro-machining techniques. The thin films used in this work are therefore fabricated by chemical solution deposition.

A thorough investigation of these materials is necessary in order to use the materials to their full potential. For piezoelectric characterization, the resonance technique,[5] which is widely used for bulk samples, is not applicable for thin films. Since the technique measures the natural frequency of a sample, the characterization of thin films would require frequencies in the GHz range due to the small sample geometries.

Another approach is to measure the piezoelectric properties at subresonance frequencies far below the characteristic frequency. Both the direct piezoelectric and the converse piezoelectric effect are used to evaluate the piezoelectric properties.[6,7] However, measurements using the direct piezoelectric response induce too much mechanical stress in thin films.[8]

Using the converse piezoelectric effect, the electric field induced displacements of the sample are measured. Since the small thickness of thin films limits the voltage applicable to the samples, the displacements are in the angstrom range. The nonlinear piezoelectric response of ferroelectric materials for different applied electric field requires an even higher resolution of about 1–10 pm. Interferometric techniques are one approach to achieve such high resolutions.[6,8,9]

Since single-beam techniques[9–11] do not take sample motion into account, it is prone to errors resulting from sample bending. Therefore, the double-beam technique[8,12–14] is the superior method for measuring ferroelectric thin films.[8]

The conventional technique for measuring the behavior of weak-signal responses at different applied electrical fields (d_{33} hysteresis) uses a bias field with a superimposed, weak-signal ac field. To achieve the high resolution needed for this measurement, a lock-in-amplifier with a large time constant is used. Therefore the measurement time for a complete d_{33} hysteresis is in the magnitude of several minutes, resulting in electrical stress in the sample. This Note presents a technique to shorten the time needed for the measurement down to the magnitude of seconds by using different means of averaging without reducing the accuracy of the interferometer. Several measurements are shown to display the possibilities and limits of the technique.

The double-beam (Mach–Zender) interferometer has been proven to be superior to the single-beam Michelson interferometer[9–11] when used to measure piezoelectric thin films.[8] The disadvantage of this technique is a reduced resolution (10^{-3}–10^{-2} Å).[8] The optical path of the double-beam laser interferometer used in this work is equal to the one used in Ref. 8 and will not be discussed in detail.

Using a photodetector, whose output voltage is proportional to the measured light intensity and using the point of maximum sensitivity at a phase shift of $\pi/2$ as the operating point, the displacement change of the observed sample surface is proportional to the measured voltage change ΔV.[8]

If the sample is driven by a sinusoidal voltage, ΔV can be measured by a lock-in amplifier (Perkin Elmer 7280 wide bandwidth DSP lock-in amplifier), which calculates the amplitude of ΔV and the phase shift between the driving voltage and ΔV. Using high time constants for the lock-in amplifier, resolutions up to 10^{-5} Å can be achieved.[8]

The performance of the system can be evaluated by measuring the piezoelectric displacement of quartz, as its d_{33}

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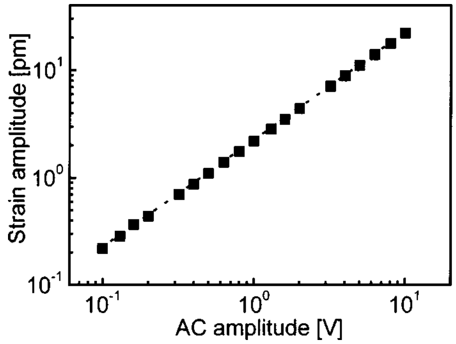

FIG. 1. Linearity of the interferometer response ($f = 3$ kHz) measured with an *X*-cut quartz sample in comparison to literature value (dotted line).

value is well known. The measured driving field dependence of the piezoelectric response of an *X*-cut quartz sample manufactured by Crystec (10 mm×10 mm×1 mm) is given in Fig. 1. The measurement shows a linear behavior even for displacements near the resolution of the interferometer. Defining resolution as the minimum displacement measurable due to noise, 2×10^{-1} pm can be achieved using a time constant of several seconds. The calculated piezoelectric coefficient corresponds well to the literature value of 2.3 pm/V. Measuring the dependence from the driving frequency, only minor deviations are found until the beginning of sample resonance at 40 kHz.

The conventional technique for studying the piezoelectric weak-signal behavior uses a *LCR* bridge to generate a stepwise changed electric field (bias) which is superimposed by a small ac field. The resulting deformation caused by the ac field is determined via a lock-in amplifier and measured at different defined states.[8,10–12,14] The lock-in amplifier requires several seconds of measurement time to achieve a high-resolution measurement of the electric field induced displacement per defined state of the sample. Depending on the number of states measured, the complete measurement requires a minimum time of several minutes. During this long period, a relatively high unipolar electric field is applied to the sample, resulting in stress and imprint effects. Another disadvantage is the slow frequency of the bias change. Therefore, direct comparison and calculations using electrical and mechanical measurements is difficult.[15,16]

In order to decrease the measurement time per defined state or voltage stepping, new methods for averaging and

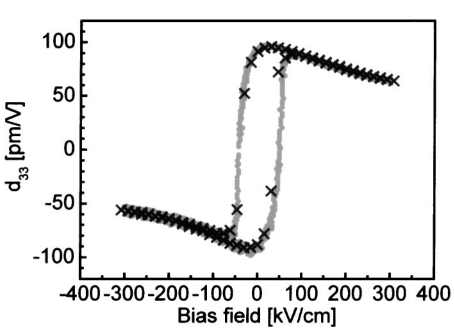

FIG. 2. Piezoelectric weak signal response of a 130 nm PZT (45/55) thin film sample measured with both the old (80 points acquired in 15 min, black stars) and new (5000 points acquired in 100 s, $f_{bias} = 10$ mHz, gray dots) technique ($V_{ac} = 100$ mV$_{rms}$, $f_{ac} = 8$ kHz).

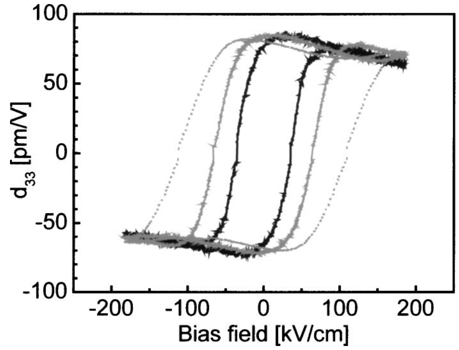

FIG. 3. Piezoelectric weak signal response of a 130 nm PZT (45/55) thin film sample measured with continuously changed bias field using 0.5 ms (black line), 20 ms (gray line), and 50 ms (gray dotted line) time constants for the lock-in ($f_{bias} = 1$ Hz, $V_{ac} = 100$ mV$_{rms}$, $f_{ac} = 8$ kHz).

filtering of the interferometric response signal should be used. Combining the internal oscillator of the lock-in amplifier with a bias voltage generation by a wave form generator (Wavetek 395), unlocking of the lock-in from the signal is prevented. Thus, the measurement time per defined state is reduced below the millisecond range and the bias voltage can be changed continuously. In order to achieve a better resolution, several cycles are measured and averaged. This idea corresponds to the technique proposed for measuring strain curves in Ref. 8, but is refined using the lock-in to reject most of the noise.

An example measurement for Lead Zirconate Titanate (PZT) (45/55) is shown in Fig. 2 (gray dots). The sample with a film thickness of 130 nm was prepared on a double side polished silicon substrate (1 in.×1 in.×0.5 mm) using a chemical solution deposition. Its backside is vapor deposited with Au to achieve better reflectivity. For direct comparison the measurement was also done using the old technique (black stars). As can be seen, the results from both measurements fit very well and are comparable to measurements published in Refs. 8 and 14.

Another advantage of the new technique is the higher accuracy due to the high number of measurement points, which is 5000 points for the measurement using the new technique in comparison to 80 points measured by the classic technique. In particular the vicinity of the coercive field is measured accurately, which is very difficult using the old technique due to the high steepness of the curve. The short-time measurement was done with a bias frequency of 10 mHz in order to achieve comparable results. Therefore, a time constant of 50 ms was chosen and one bias cycle was sufficient for the measurement. Hence, the measurement time was decreased from several minutes to 100 s.

Using lower time constants the frequency of the bias voltage cycle can be raised up to 1 Hz, which was done in Fig. 3 (black line). Above that frequency the lock-in amplifier cannot follow the displacement change fast enough and produces errors, as seen in Fig. 3 (gray lines). Even when averaging several bias voltage cycles becomes necessary due to low time constants, the measurement time is reduced to several seconds. Therefore, stress to the sample is reduced while the resolution is still adequate for studying ferroelectric thin films.

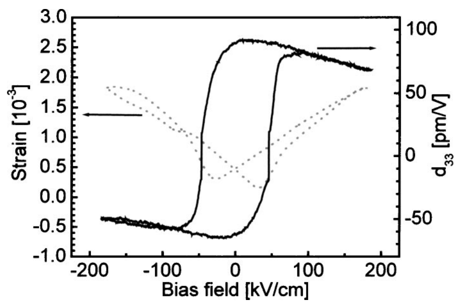

FIG. 4. Piezoelectric weak (solid line) signal response and piezoelectric strain (dotted line) of a 130 nm PZT (45/55) thin film sample measured synchronous ($f_{\text{bias}}=2$ Hz, $V_{\text{ac}}=100$ mV_{rms}, $f_{\text{ac}}=8$ kHz).

To enable direct comparison between weak and large signal measurements, as done by Bolten in Ref. 16, a four channel digital oscilloscope, like the Tektronix TDS 684A or TDS 744, is used. With this setup, it is possible to measure the weak signal behavior of the piezoelectric displacement synchronous or at similar conditions to the displacement induced by the large bias signal, as shown in Fig. 4.

In conclusion, this measurement method to investigate the piezoelectric properties of ferroelectric thin films reduces the dc field stress during weak signal measurements due to lower measurement times of several seconds instead of minutes. Thus, imprint effects can be reduced significantly. The resolution of the double-beam laser interferometer used was demonstrated to be better than 0.2 pm. Furthermore, the number of measurement points was enlarged by a factor of 60, which corresponds to an increased resolution. Also, the large and weak signal response can be recorded synchronously, due to the faster method for weak signal measurements. Therefore, better comparison of the results is now possible.

The authors wish to thank D. Damjanovic of the EPFL Lausanne for helpful suggestions.

[1] N. Setter, *Piezoelectric Materials for the End User*, Conference notes (Interlaken, Switzerland, 2002).
[2] H. Schaumburg, *Keramik* (B. G. Teubner, Germany, 1995).
[3] KFA Juelich, 26. IFF-Ferienkurs (1995).
[4] O. Lohse, Ph.D. thesis, Institute of Materials in Electrical Engineering and Information Technology, RWTH Aachen, 2001.
[5] I. Kannno, S. Fuji, T. Kamada, and R. Takayama, Appl. Phys. Lett. **70**, 1378 (1997).
[6] K. Lefki and Dormans, J. Appl. Phys. **76**, 1764 (1994).
[7] F. Xu, F. Chu, J. F. Shepard, Jr., and S. Troiler-McKinstry, Mater. Res. Soc. Symp. Proc. **493**, 427 (1998).
[8] A. L. Kholkin, Ch. Wütchrich, D. V. Taylor, and N. Setter, Rev. Sci. Instrum. **67**, 1941 (1996).
[9] Q. M. Zhang, W. Y. Pan, and L. E. Cross, J. Appl. Phys. **63**, 2492 (1987).
[10] J.-F. Li, P. Moses, and D. Viehland, Rev. Sci. Instrum. **66**, 215 (1995).
[11] L. Lian and N. R. Sottos, J. Appl. Phys. **87**, 3941 (2000).
[12] Q. M. Zhang, S. J. Jang, and L. E. Cross, J. Appl. Phys. **65**, 2807 (1989).
[13] W. Y. Pan and L. E. Cross, Rev. Sci. Instrum. **60**, 2701 (1989).
[14] H. Maiwa, J. A. Christman, S.-H. Kim, D.-J. Kim, J. P. Maria, B. Chen, S. K. Streiffer, and A. I. Kingon, Jpn. J. Appl. Phys., Part 1 **38**, 5402 (1999).
[15] A. L. Kholkin, E. K. Akdogan, A. Safari, P. F. Chauvy, and N. Setter, J. Appl. Phys. **89**, 8066 (2001).
[16] D. Bolten, Ph.D. thesis, Institute of Materials in Electrical Engineering and Information Technology 2, RWTH, Aachen (2002).