Conference Paper

Electromechanical Measurements of Gd-Doped Ceria Thin Films by Laser Interferometry

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Abstract

Highly sensitive laser interferometer was built to measure electromechanical coupling in Gd-doped ceria $Ce_{0.9}Gd_{0.1}O_{2-x}$ thin films in the frequency range up to 20 kHz. Spurious resonances due to substrate bending were avoided by the special mounting of the film in the center of substrate. Compact design allowed to reach high vertical resolution of about 0.2 pm. Electrostriction coefficient measured in 1 μm thick $Ce_{0.9}Gd_{0.1}O_{2-x}$ film was 4.3×10^{-21}m^2/V^2 and slightly decreased with frequency till the extensional resonance of the substrate at about 20 kHz occurred. As expected, the displacement varied as a square of applied voltage without any sign of saturation. A comparison with ceramics showed much higher electrostriction coefficient in the latter in the same frequency range.

Keywords: gadolinium doped ceria, thin films, laser interferometry, electrostriction

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1. Introduction

Gd-doped ceria $Ce_{0.9}Gd_{0.1}O_{2-x}$ (CGO) is a lead free compound with unusual mechanical and electromechanical properties. Off-center shift of the Ce^{4+} ions in the cubic oxygen environment away from oxygen vacancies under an action of the external electric field in CGO results in appearance of the "giant" electromechanical strain at low frequencies [1]. The effective electrostriction coefficient measured by the cantilever method at low frequencies was M_e = 6.47 ± 0.43 × 10^{-18} m^2·V^{-2} [1, 2]. It is, therefore, attractive for micromechanical applications, if the same effect is pertinent to thin films. Cantilever technique does not have enough sensitivity, and precise laser interferometry method has to be used in order to measure sub-Angstrom electromechanical displacements in thin films. In this work, we built a simple single-beam Michelson interferometer with the high vertical resolution capable of the measurements of electrostriction effect in thin films and report for the first time the electrostriction coefficient value in the $Ce_{0.9}Gd_{0.1}O_{1.95}$ layers of the thickness of about 1 μm.

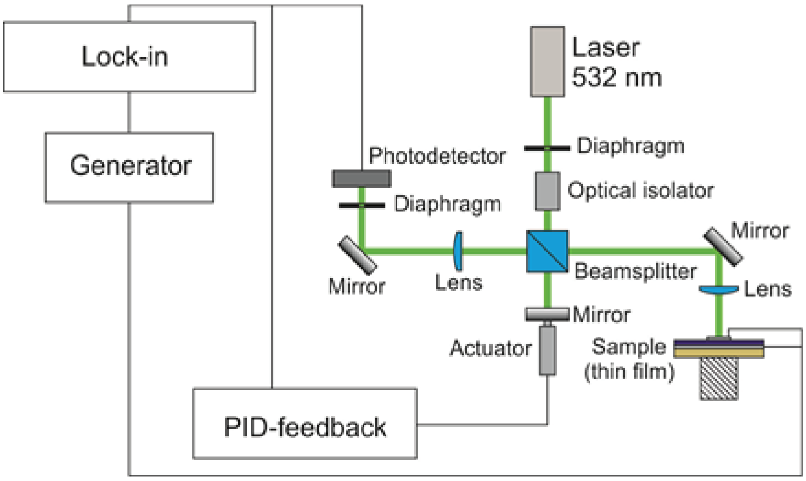

Figure 1: Scheme of the modified Michelson interferometer.

2. Methods

For this study we used CGO thin films samples. Ceramic samples were prepared by the solid synthesis route as described in the earlier work [3, 4]. The surface was polished to optical quality and coated by Al electrodes via magnetron sputtering. Thin films were prepared on the 250 μm thick n-Si^{++} wafers via sequential RF magnetron sputtering of the 150 nm thick of Cr layer, ≈900 nm thick CGO-layer, 300 nm thick of Cr layer. Cr layers served as the top and bottom electrodes. Top electrodes were deposited via shadow masks. The diameters of the top electrodes were 2 mm.

For the measurements of the small electric field induced displacements we used a standard scheme of single-beam Michelson homodyne. interferometer as described in many publications (see e.g. [5, 6]). The schematic of the interferometer setup is shown in Fig. 1. Depending on the path length difference between the interferometer arms, small displacements of the sample surface produce a change in detected light intensity anywhere between the maximum theoretical sensitivity and zero. In order to maintain the working point between the minimum and maximum position of the photodetector, a PID-feedback system was used to stabilize the system against the slow optical path-length drift with the cut-off frequency of 5 Hz. The system compensates for the drift below 5 Hz and assumes that higher frequency is due to electric field induced displacements. The system was as compact as possible with the optical path of each arm about 1 m.

We used a single-mode stabilized solid state diode laser LCM-S-111-20-NP25 (Laser-compact, Russia) with a wavelength 532 nm and a power 20 mW, lock-in amplifier SR830 (Stanford Research, USA), multifunction data acquisition board USB-6251 (National Instruments, USA), and signal generator Agilent 33210A (Keysight Technologies, USA). For the PID-feedback loop piezo actuator P-841.01 and piezo controller E-709.SRG (Physik Instrumente (PI), Germany) were used.

The minimal displacements (interferometer resolution) are determined by the noise in the laser and in photodetector by the minimal voltage measured by the lock-in amplifier. On the top of this, the system experiences also a vibrational noise from the

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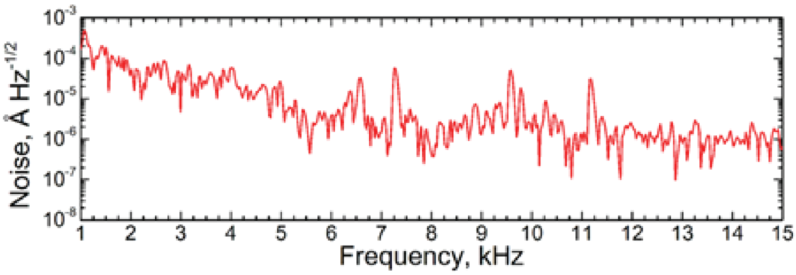

Figure 2: Spectral density of the noise registered by the interferometric setup.

environment amplified by the mechanical resonances in the sample holder. In order to reduce this noise, we used a special fixture for the sample holder (Fig. 1). The thin film sample was rigidly glued to the small steel rod with the contact area of 5 mm^2 in the central part. Thus, the effect of bending resonances on the vibrational noise was avoided. Also, the part of the interferometer containing both arms were isolated from the environment using a special box made of foam boards, foam rubbers, and aluminum frame. Figure 2 shows the equivalent displacement noise of the system in the range 2-15 kHz measured for the time constant of lock-in amplifier 100 ms. It is clearly seen that the vibrational noise still dominates the response and the resolution varies in the range $10^{-2} \div 10^{-4}$ Å depending on frequency. Thus, the frequency dependence of the electrostrictive displacements can be measured with high enough accuracy.

The performance of the interferometer was validated by measuring the displacements in a standard ceramic PZT5A pellet [7]. The frequency response within the range of 1 to 10 kHz (Fig. 3a, red curve) was almost flat with a small number of spurious resonances that prevented accurate measurements in a broad frequency range. In comparison, strong resonance behavior was observed in the film in a standard configuration, i.e. when it was glued by entire surface (black curve). It demonstrates effective suppression of the bending effect in bulk ceramics. Measured piezoelectric coefficient of PZT5A was 374 ± 5 pm/V. This value is in a good agreement with the table value for the piezoelectric coefficient d_{33} of PZT5A [7]. To verify the linearity of the setup, we measured the AC-voltage dependence of displacements in congruent lithium niobate (CLN) sample (Z-cut) that has negligible own non-linearity. Figure 3b demonstrates that the amplitude of the displacement is linear with the amplitude of applied voltage up to 5 V. The resulting piezoelectric coefficient of CLN sample d_{33} was 6 pm/V, which also corresponds to the table value [8]. The minimum displacement measured in this case was $2 \cdot 10^{-2}$ Å (signal to noise ratio 1).

3. Results

The results of the electrostriction measurements in CGO thin film samples are shown in Figure 4. Measurements were done in the frequency range 5-30 kHz (second harmonic) and voltage amplitudes up to 6 V. The frequency dependence was almost flat with a tendency to decrease up to a clear resonance at 20.1 kHz. This peak is attributed to the extensional resonance of substrate that cannot be avoided in the present setup. The measured effective electrostriction coefficient was $M_{33} = 4.3 \cdot 10^{-21}$ m^2/V^2 at frequency

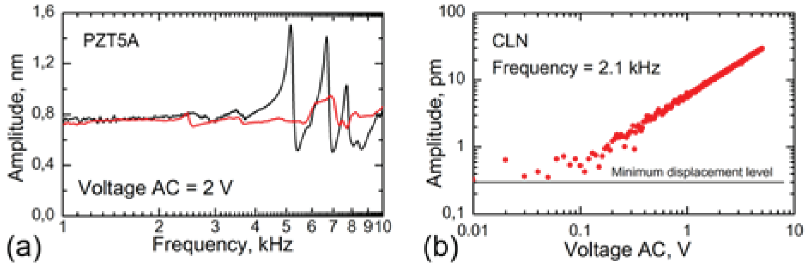

Figure 3: (a) Frequency dependency of piezoelectric response of the PZT5A sample measured by interferometer before modifications (black) and after (red). (b) Voltage dependency of piezoelectric response of the CLN sample.

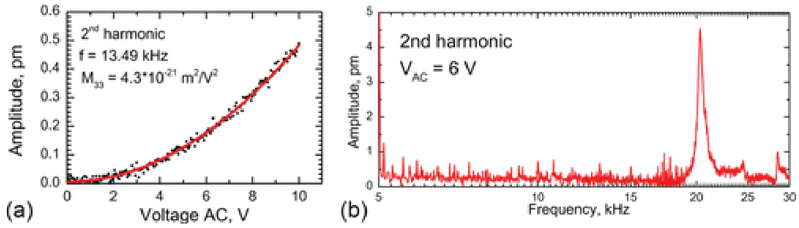

Figure 4: (a) Voltage and (b) frequency dependencies of amplitude of CGO thin films.

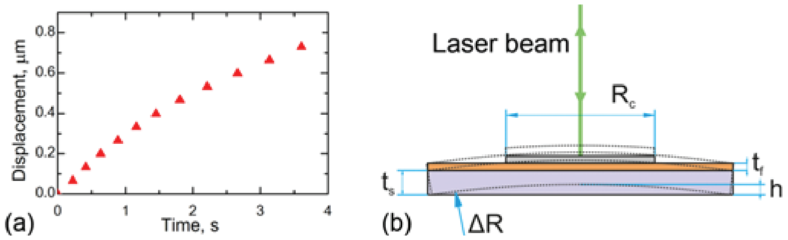

Figure 5: (a) Time dependence of the displacement of CGO thin film sample's top surface and (b) scheme for Stoney formula.

13.49 kHz (Fig. 4a, b). This value is much lower than that reported in Ref. [1], measured in ceramics at low frequency. Measurements below 1 kHz are difficult in our setup because of the increase of vibrational noise from the nearby machines and natural limit due to the cut-off frequency of the feedback system. Low value of M_{33} can be also attested to the clamping effect of the substrate and smaller grain size as compared to ceramics.

For the interferometric studies, the surfaces of the ceramic samples were polished and the reflecting Cr-electrodes were sputtered using Edwards Auto 500 electron-beam evaporation system. Measurements of the electrostrictive coefficient M_{33} were performed by applying AC voltage with frequency *f*, and the surface displacement was measured at *2f* using the corresponding feature of the lock-in. The time constant of the lock-in was always 300 ms.

In our setup, we could also measure a transverse electrostriction coefficient M_{13}. In this case, 700 nm CGO thin film was allowed to bend during DC voltage application of 4.9 V. In this case, bending moment develops, which translates into the displacement of the top surface of the film (see schematic in Fig. 5a). The effective electrostriction coefficient M_{13} can be calculated from this data using the Stoney formula [9] (Fig. 5b) that is generally used to calculate the residual stress value:

$$\sigma = \frac{Y_s}{6(1-\nu_s)} \frac{t_s^2}{t_f} \Delta R \tag{1}$$

where Y_s is the Young's modulus of substrate (179 GPa), ν_s is the Poisson's ratio (27), t_s is the thickness of the substrate (250 um), t_f is the thickness of the film (700 nm), and ΔR is the change of the curvature radius of the bent sample (1.25 m). Then, the stress value can be used in calculation of M_{13} coefficient:

$$M_{13} = \frac{\gamma_{13} \cdot (1-\nu_f)}{Y_f} \tag{2}$$

where γ_{13} is the stress coefficient ($\sigma \cdot E^{-2}$, ≈600 $kPa \cdot kV^{-2} \cdot cm^{-2}$), Y_f is the Young's modulus of film (190 GPa), and ν_f is the Poisson ration of the film (43). The estimated electrostrictive coefficient was M_{13} = $1.7 \cdot 10^{-16}$ m^2/V^2, which is very close to the value reported in Refs. [1, 2]. We should take into account that this value is measured at DC (i.e., at zero frequency) in the partially clamped conditions. That effective electrostriction coefficient is determined not only by the intrinsic mechanism due to ionic displacements and bond stretching, but can also depend on the competition between intragrain conductivity and Schottky behavior of the electrodes; large difference between the DC and high frequency measurements is not a surprise. Further measurements need to clear out the electrostriction mechanism in the CGO films.

4. Conclusions

Highly sensitive single beam interferometer to measure electrostrictive displacements was developed in this work. Resolution of the interferometer varied in the range $10^{-2} \div 10^{-4}$ Å depending on frequency and the minimum displacement measured was $2 \cdot 10^{-2}$ Å. Gd-doped ceria $Ce_{0.9}Gd_{0.1}O_{1.95}$ (CGO) thin films have been investigated to get both M_{33} and M_{31} coefficients. Interferometric measurements showed relatively low electrostrictive coefficients at high frequency (M_{33} = $4.3 \cdot 10^{-21}$ m^2/V^2) possibly due to elevated impedance of electrodes at high frequencies. Applying the DC voltage to the film provided giant displacement in thin film and the calculated transverse electrostrictive coefficient was M_{13}= $1.7 \cdot 10^{-16}$ m^2/V^2.

Acknowledgements

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