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Microcracking and discontinuous fast switching as acoustic emission sources in 8/65/35 and 9.5/65/35 PLZT relaxor ferroelectrics

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Abstract

Microcracking, discontinuous switching, crystallographic phase transformation, internal discharges or surface friction of the macroscopic sample may be acoustic emission (AE) sources in ferroelectric ceramics. To reduce the number of possible AE sources, AE measurements were performed on hot pressed pore free well polished rhombohedral lead–lanthanum–zirconate–titanate (PLZT). Microcracking is the only AE source in electrostrictive PLZT 9.5/65/35, while in ferroelectric PLZT 8/65/35 fast domain kinetics also contribute to the observed AE pattern. © 2001 Elsevier Science Ltd. All rights reserved.

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1. Introduction

In a previous publication several different possible AE sources in the crystal structure system of lead-zirconate-titanate (PZT) were discussed.[1] Energy based criteria were used to categorise AE sources using certain assumptions for the evaluation of the AE stress amplitudes. For morphotropic phase boundary PZT (2% La) it was possible to separate AE signals associated with crystallographic phase switching between both crystal structures from AE signals associated with discontinuous ferroelastic switching within the tetragonal or rhombohedral phases. Coarse (3 μm) as well as fine (1.5 μm) grained materials showed AE events associated with microcracking. AE also occurred right at E_c for the rhombohedral and morphotropic materials along with the strong macroscopic strain changes at this field. To test the hypothesis of microcracking induced AE, a purely electrostrictive material [PLZT 9.5/65/35, La/(Zr/Ti)] is tested in the present investigation and compared to AE data from ferroelectric 8/65/35.

Generally, the AE method detects all abrupt local stress or strain changes within a material.[2] AE investigations in ferroelectrics have been concerned with phase transitions[3,4] and electrically driven single hysteresis loops.[5,6] In the former case, microcracking[3] in ceramics and the formation of domain wall surface originating at point defects introduced during crystal growth in the case of single crystals[4] have been discussed. Unipolar driving yields significantly less AE than bipolar switching.[7] It was also possible to show, that cyclic fatigue strongly modifies the AE patterns in tetragonal PZT close to the morphotropic phase boundary.[8] The differences in AE patterns were associated with the growth of point defect agglomerates interfering with the free movement of the domain walls. General properties of PLZT are given, e.g. in Refs. 9 and 10.

2. Experimental

The specimens were cut from commercially available optically transparent hot pressed PLZT (Aura Ceramics, now TRS Ceramics) to discs of 1 mm thickness and 10 mm diameter. This particular batch of material has been well characterised electrically and mechanically in several investigations.[11,12] The flat surfaces of the specimens were polished down to a 1 μm finish and fully electroded by sputtering Au/Pd. One additional rectangular sample of 9.5/65/35 of identical thickness and volume was polished on all six surfaces. The experimental set-up consists of a commercial AE-detection device (AMS3, Vallen Systeme, Icking, Germany) described elsewhere.[8]

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The amplification was chosen as 54 dB, the threshold as 20.7 dB (=12 μV). The logarithmic amplitude scale (in dB) is given with respect to 1 μV. The AE-energy scale is 1 e.u. $=10^{-18}$ J at the preamplifier input. In our set-up the decay time of the AE events was identical for all events, therefore energy and amplitude can be equivalently used: $E=\alpha A_{\max}^2$; $\alpha=6.8\times10^{-4}$ (e.u.)/(μV)^2. The strain was monitored using an inductive linear variable displacement transducer (LVDT) in an AC-bridge. The resolution was about 10 nm. The charge was determined from the voltage reading on a 4.5 μF capacitor in series with the sample (electrometer 6127, Keithley Instruments, Cleveland, OH, USA). The HV was applied at 20 mHz triangular bipolar voltage. A high voltage source of very narrow bandwidth near DC was used to reduce spurious electromagnetic noise,[13] the damping was further enhanced by a RC-low pass filter.

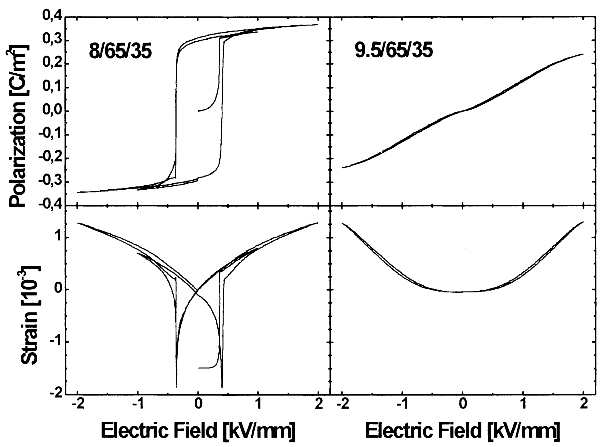

Fig. 1. Polarization and strain hysteresis for ferroelectric 8/65/35 and electrostrictive 9.5/65/35 PLZT. The strain scale is set to zero for the remanent strain of 8/65/35.

3. Results

The 8/65/35 PLZT specimens show the expected macroscopic ferroelectric and strain hysteresis (Fig. 1). Like in other rhombohedral PZT compositions[1] no AE occur below the coercive field (Fig. 2). Each dot in Fig. 2 represents one AE event. At E_c, numerous AE events of different amplitudes occur. Right above E_c hardly any events are found. At higher fields and stresses, the AE events are more numerous. The peak AE amplitudes in this field range rise with increasing field level. The high amplitude events occur when polarisation and strain saturate. If the AE events are plotted versus strain, some events occur at all strain levels in ferroelectric 8/65/35, but most events are found close to saturation (right side in Fig. 2). The AE events at the coercive field are smeared out over a broad range of strains. In electrostrictive 9.5/65/35 the first AE events occur at moderate field levels, simultaneously with the first significant strain changes. The maximum event amplitudes rise according to the strain level reached. This type of event is similar to those found in 8/65/35 at high strain and field levels. No events occur for decreasing field levels in either material.[1,13] The energy distribution of the AE

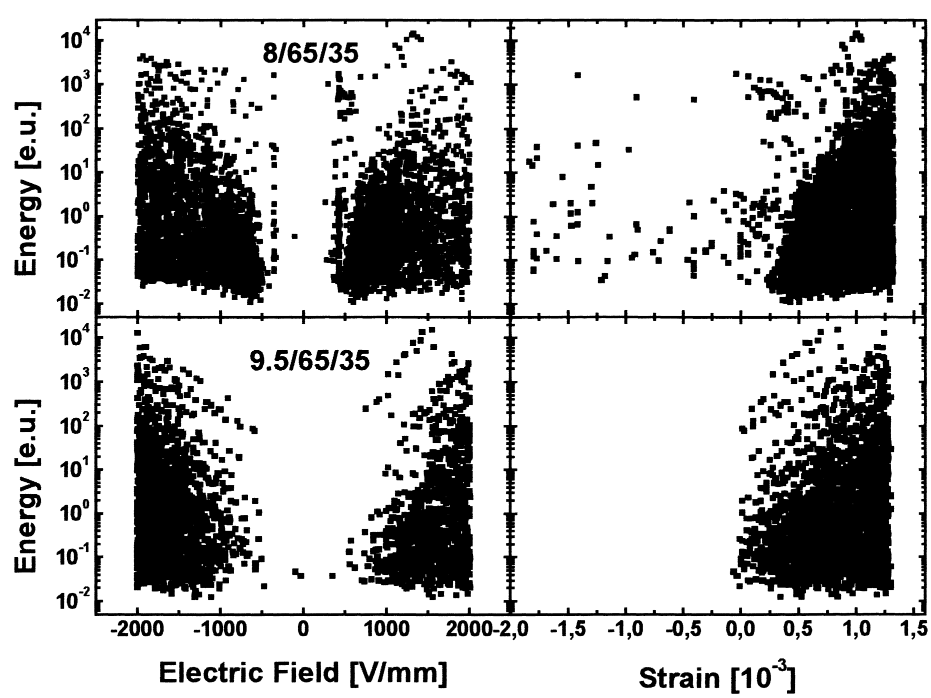

Fig. 2. Field and strain dependent AE for 8/65/35 and 9.5/65/35 PLZT.

shows different slopes on a log–log plot (Fig. 3). In 9.5/65/35 a linear dependence can be found (slope $\beta = -0.24$), while in ferroelectric 8/65/35 a round curve is found and only an approximate slope of ≈ -0.5 can be determined. The rectangular sample of 9.5/65/35 with all six surfaces well polished shows no AE in the same field range.

4. Discussion

Different AE types were previously deduced from different observations. AE events occurring at E_c were associated with fast kinetics.[1] Directly above E_c, ferroelastic switching was assigned as the AE source, particularly in tetragonal PZT.[1] At high field levels close to saturation strain as well as polarisation changes become increasingly difficult as the domain system is clamped and no longer mobile enough to accommodate high local fields or stresses. Strong jumps in polarisation and microcracking occur. It was concluded from energy estimations that microcracking is the dominant AE source in this regime, but both mechanisms may actually occur.[1] Furthermore, very large AE events found for all field values above E_c were assigned to the crystallographic phase change of some grains in a coarse grained specimen right at the morphotropic phase boundary. This correlation was determined from the disappearance of these events after short cycling.[1]

The AE amplitudes found in the present study for PLZT 8/65/35 and the range of their occurrence are similar to other coarse grained rhombohedral compositions.[1] Large strain changes occur at E_c accompanied by AE events. As no AE occur for the strong strain changes from negative maximum field to beneath positive E_c, sample friction on the microphone is not a cause of these AE. Thus fast domain switching (possibly across some type of obstacles) generates the AE events right at E_c in the ferroelectric PLZT. Similar to other rhombohedral compositions, no AE events are observed at field levels right above E_c, where events previously assigned to pure but slower discontinuous switching are expected and were observed in tetragonal PZT. This type of event does not occur in PLZT 8/65/35, thus moderate switching speeds do not induce AE in rhombohedral PLZT. At high strain levels (values above the remanent strain s_r, which is set to zero here for better comparison to the electrostrictive composition) the domain switching saturates. Strains elastically convert to stresses as the domain system is no longer mobile and able to accommodate the strain changes (right side in Fig. 2). The highest number of AE arises. All energy values in between the threshold and the maximum value occur without gap. In this range the discontinuous fast switching of domains (or domain systems) clamped by some obstacles and driven by high field levels as well as microcracking due to high local stresses are possible AE sources.

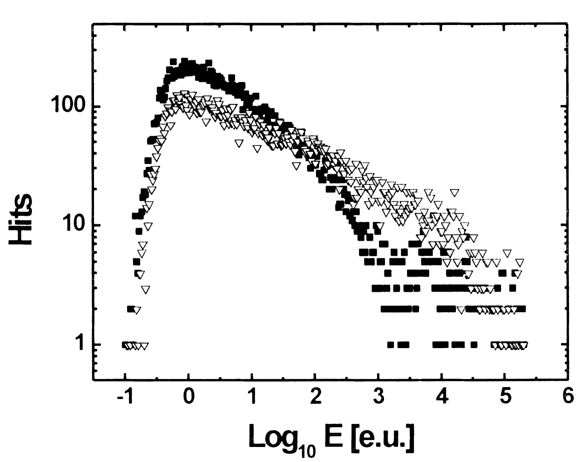

Fig. 3. AE energy distribution (8/65/35, ▽ 9.5/65/35).

The saturation range of strain in 8/65/35 above s_r coincides with the total strain range found in electrostrictive 9.5/65/35. It was discussed in Ref. 14 that microcracking on the order of single grain facets becomes optically visible in 9.5/65/35 PLZT. Calculations also showed that this range of AE energies and fields is best described by microcrack formation of single grains.[1] PLZT 9.5/65/35 does not have a domain system to accommodate local stress enhancement. All strain changes will directly yield high stress levels due to field induced strain anisotropy. These seem sufficient for microcrack formation at surface defects, which shows as AE. PLZT 9.5/65/35 does not change its optical properties for the entire field range covered. A switching from the nanodomain system present in the relaxor compositions to a microdomain system does not occur. It would render the sample opaque.[11,12] To be completely sure that surface flaws were responsible for the microcracking, a second rectangular sample was used with all six surfaces neatly polished. Complete electrode coverage was ensured and the identical sample volume and thickness were chosen. No AE occur in this case. Thus, the AE observed in the cylindrical samples of 9.5/65/35 are due to microcracking initiating at surface flaws on the circular perimeter of the sample. This correlates well with the optical observations for microcrack clouds forming under cyclic electric loading.[14]

The energy distribution indicates that there are differences in the underlying mechanism of AE generation between both compositions. It is known in magnetic systems that sufficient disorder is needed in the microstructure, to yield fractal exponents in energy distributions of Barkhausen-pulses[15] and correlation between AE and Barkhausen-pulses has been given earlier.[8] In all small grained compositions of an earlier study, these sufficiently disordered AE were assigned to microcracking, but no proof could be given.[1] In this study, all other sources can be excluded for 9.5/65/35 and microcracking seems to yield a non-integer exponent of the energy distribution. In 8/65/35, the energy distribution differs from that in 9/65/35 and is particularly not linear on a log–log plot. Thus other types of AE sources are also present, which we associate with discontinuous switching. As the samples in the present study are 100% dense, internal discharges cannot be a source of AE in either material.

Altogether, microcracking on the order of single grain facets originating at surface flaws seems to be the dominant AE source in the electrostrictive PLZT 9.5/65/35. Besides microcracking, some contribution from switching is encountered in ferroelectric PLZT 8/65/35.

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