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Kinetics of domain redistribution in $SrTiO_3$ under pulsed electric fields

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ABSTRACT

$SrTiO_3$ is a well-studied perovskite system which undergoes an antiferrodistortive phase transition at 105 K. The low-temperature tetragonal phase exhibits different domains corresponding to the orientation of the symmetry axis. The domain structure can be changed drastically by applying electric fields. Using time-resolved neutron diffraction, the kinetics of the domain redistribution has been investigated on a time scale between microseconds and seconds. It is shown that there are at least two different types of relaxation processes on different time scales which govern the transformation. These can be related to the motion of hard and easy domain walls. Moreover, it is demonstrated that at low temperatures the field induced transition into the ferroelectric phase reveals a significant hysteresis which is reflected by the distribution of tetragonal domains. This finding supports the view of the zero field so-called quantum-paraelectric or coherent-paraelectric phase consisting of polar clusters which are removed only by heating above 40 K.

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Ferroelectrics; domain distribution; kinetics; external electric field; external stress; neutron scattering

Introduction

As a member of the perovskite structure type, $SrTiO_3$ has been investigated extensively during the last decades. As a substrate for superconducting layers, $SrTiO_3$ has received considerable interest due to the structural similarity to high-T_c-materials. Even though, fundamental properties of this compound associated with low-temperature phase transitions and their response to external perturbations like mechanical stress or electric fields are not yet completely understood.

Two competing order parameters have been identified that determine the phase behavior of $SrTiO_3$. Both are related to soft phonon modes. A zone-boundary mode of the cubic room temperature phase condenses on cooling at about 105 K leading to a tetragonal

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superstructure [15]. The corresponding antiferrodistortive transition leads to the formation of three different (twin-) domains with their elongated tetragonal axes along *x*, *y* or *z*. The tetragonal distortion is the order parameter which describes this transition. It is well known that the domain distribution can be controlled by the application of uniaxial stress which favors domains with symmetry axes perpendicular to the stress direction [1, 12]. Unlike the isotypic barium compound $SrTiO_3$ does not become ferroelectric even at the lowest temperatures though a polar phonon mode at the Brillouin-zone center softens continuously on cooling. Only by isotope substitution [8], by the application of mechanical stress [4] or an electric field [6] a ferroelectric phase can be induced below about 40 K which can be described by the spontaneous polarization as the order parameter. Recently, we observed that an applied electric field does not only favor the polar phase but is also able to change the domain structure of the tetragonal phase [14]. This is particularly surprising since the electric field and the tetragonal order parameter exhibit different symmetry properties. Hence, it was concluded that higher order interactions are responsible for this effect. Tetragonal domains with symmetry axes perpendicular to the electric field are energetically favored since the apical oxygen ions of the TiO_6-octahedron are shifted perpendicular to the field induced displacement of the central Ti^{4+}-ion as shown in Fig. 1.

Moreover, it was demonstrated that the polarization of field-induced ferroelectric phase at low temperatures is perpendicular to the tetragonal axis leading to orthorhombic symmetry.

The domain structure is also reflected by polarized Raman spectra. It could be shown that Raman tensors which describe the intensities of modes in different configurations vary from one tetragonal domain to another [5]. Hence, the application of electric fields leads to strong characteristic changes in the Raman spectra.

In recent studies, the properties of twin domain walls in perovskite bulk systems and thin films have been investigated in some detail by Morozovska et al. [9][10][11]. It is claimed that domain boundaries may be sources of ferroelectric polarization even within the temperature regime of the paraelectric phase. Moreover, hard and easy twin boundaries are distinguished which are characterized by the relative orientations of the rotational axes of neighboring octahedra on both sides of the domain wall. This is illustrated in Fig. 2.

Recently, it has been pointed out by Fontcuberta et al. [3] that the domain structure of $SrTiO_3$ and their modification by applied electric fields has, in fact, a strong influence on the

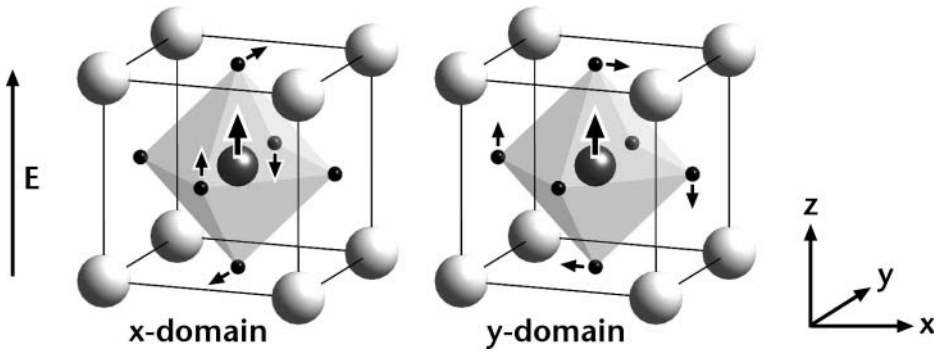

Figure 1. The displacement of the Ti^{4+}-ion in the center of the octahedron in the direction of the electric field (*z*) leads to a preferred formation of *x*- and *y*-domains associated with a shift of the apical oxygen ions in transverse direction (after [14]).

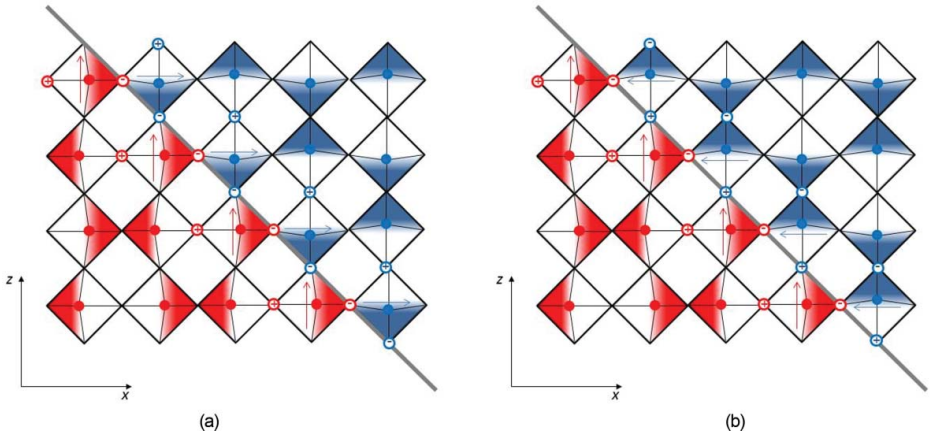

Figure 2. Illustration of a boundary between *z*- and *x*-domain.
Left: easy twin wall, right: hard twin wall (after [11]).

magnetic behaviour of manganite layers deposited on this perovskite substrate. Hence, the properties of the layer seem to be intimately related to those of the substrate which are not yet completely known.

More information about the interactions between the different order parameters can be obtained by studying the real-time variation of the domain structure after the application of an electric field which is associated with the motion of domain walls. Using time-resolved neutron scattering, we have therefore performed kinetic experiments and determined the time evolution of the domain distribution during a pulsed electric field.

Experimental

The observation of a series of superlattice reflections allows the determination of the volume fraction of tetragonal domains which are formed on cooling a single crystal of $SrTiO_3$ below the transition temperature of 105 K. Details of this method are described elsewhere [14]. The present neutron scattering experiments were performed using the three axes spectrometer PUMA at the Heinz-Maier Leibnitz center in Garching and the four-circle diffractometer D10 at the Institut Laue-Langevin in Grenoble. Both instruments are equipped with Eulerian cradles which guarantee the access to Bragg reflections in three dimensions. The integrated intensities of the individual superlattice reflections of the (1/2 1/2 7/2)- and (1/2 3/2 5/2)-type[1] were obtained from rocking-scans.

Samples of high quality were cut from a commercial $SrTiO_3$-boule provided by MaTecK GmbH (Jülich) and Crystal GmbH (Berlin). In order to prevent formation of additional dislocations we refrained from polishing the sample faces. Typical sizes of the transparent and colorless specimen were about $4 \times 6 \times 8$ mm^3 oriented along the cubic directions [001], [110] and [1–10], respectively. The mosaicity was determined with high resolution γ–ray diffraction using 316 keV radiation from an Ir-192 source and was below 0.03°. Electrodes

[1] Note, that we will use the cubic directions *x*, *y* and *z* from the paraelastic phase to describe the tetragonal crystal below 105 K.

were applied to the [001]-face of the sample with silver paint. The sample was mounted in a uniaxial pressure cell to allow the simultaneous application of stresses along [110] perpendicular to the electric field. This device was installed within a closed cycle cryostat. Details of this equipment may be found in [14]. The temperature was monitored with a Pt100 resistor attached directly to the anvil in the vicinity of the sample. An additional Si-diode was located at the cold head of the cryostat. The long term stability of this system was better than 0.1 K.

While the uniaxial stress was applied statically, pulsed electric fields were generated using a high-voltage generator (FUG 2HCN 12M-6000, HCB 20M-10000) in combination with a semi-conductor switch (Behlke HTS 81–03 GSM), triggered by a function generator (Rhode&Schwarz AFGU). This setup allows the application of almost rectangular HV-pulses of several kV with frequencies of up to 1 kHz.

Using the stroboscopic technique of data acquisition [2], the scattered intensity at each individual position of the spectrometer was registered within 100 time channels of equal width per period of the electric field. For 500 Hz-cycles, e.g., the period of 2 ms was thus divided in time channels of 20 μs each and the intensity was accumulated over one million cycles in order to obtain sufficient counting statistics. In order to save beam time, these experiments were performed at the center of the respective Bragg peaks only. It was confirmed that there was no variation neither in the position nor the width of the peak by taking rocking curves between different kinetic runs. Hence, we are confident that the peak-intensity can be used as a true measure of the integrated Bragg intensity which reflects the domain distribution.

Results and discussion

As described in [14], the domain fraction can be determined from the superlattice reflections of the type (*h/2 h/2 l/2*) with *h,l* odd since only domains with their symmetry axes along the *h*-directions contribute to their intensity. From the (1/2 1/2 7/2)-reflections, e.g., the volume fractions of the *x*-, *y*- and *z*-domains are obtained as:

$$\begin{aligned}
\phi_x &= \frac{I_{1/2\,7/2\,1/2} + I_{1/2\,1/2\,7/2} - I_{7/2\,1/2\,1/2}}{I_{7/2\,1/2\,1/2} + I_{1/2\,7/2\,1/2} + I_{1/2\,1/2\,7/2}} \\
\phi_y &= \frac{I_{7/2\,1/2\,1/2} + I_{1/2\,1/2\,7/2} - I_{1/2\,7/2\,1/2}}{I_{7/2\,1/2\,1/2} + I_{1/2\,7/2\,1/2} + I_{1/2\,1/2\,7/2}} \\
\phi_z &= \frac{I_{7/2\,1/2\,1/2} + I_{1/2\,7/2\,1/2} - I_{1/2\,1/2\,7/2}}{I_{7/2\,1/2\,1/2} + I_{1/2\,7/2\,1/2} + I_{1/2\,1/2\,7/2}}
\end{aligned} \tag{1}$$

Each individual sample exhibits a specific domain distribution if it is cooled below the transition temperature without any external influence. This is probably due to memory effects and internal strains produced during crystal growth or sample preparation. Therefore, the static domain distribution was determined for each sample under consideration. In most cases, there was a slight predominance of *z*-domains.

The time evolution of the (1/2 1/2 7/2)-reflection is shown in Fig. 3 during a 2 ms-cycle of a pulsed field of 4 kV/cm and a frequency of 500 Hz at 20 K for different applied stresses perpendicular to the field. It is clearly seen that the intensity increases strongly in the

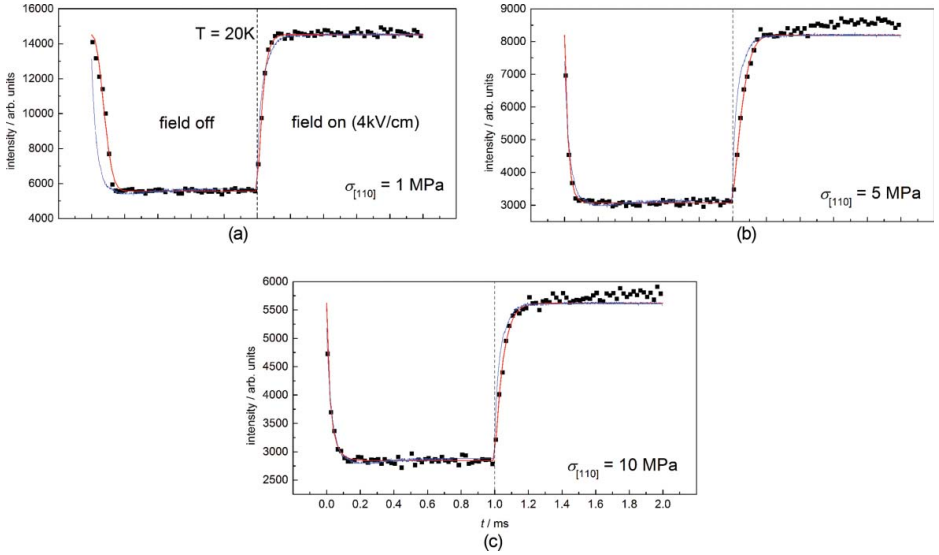

Figure 3. Time-evolution of the (1/2 1/2 7/2)-intensity during a 2-ms-cycle of a pulsed electric field of 4 kV/cm along [001] at 20 K for different competing mechanical stresses along [110]. The red lines are results of fits using a stretched exponential function while the blue lines correspond to the profile of the electric field.

field-on period almost instantaneously with the rising field (blue line) within about 100 μs. If the field is switched off, the stress bias guarantees a fast relaxation back to the initial state. Only at low stresses (1 MPa), there is a slight delay of the response. Moreover, the intensity levels decrease with stress corresponding to the fact that stress along [110] favors z-domains [14] which do not contribute to the (1/2 1/2 7/2)-reflection. Since the electric field along [001] is known to transform z-domains into x- and y-domains [5, 14] it acts against the bias stress and consequently its effect is reduced if the stress is large enough. This is reflected by the fact that the amplitude of the intensity variation becomes smaller.

The time evolution of the volume fractions of all domains are calculated using equation (1) from the time evolutions of the superlattice reflections. As an example, the data for the z-domain are shown in Fig. 4. The initial domain distribution of this sample without any external load was $\phi_x = 21.1\%$, $\phi_y = 36.2\%$ and $\phi_z = 42.7\%$. Without electric field, the fraction of z-domains is increased to about 60% at 1 MPa and 80% at 10 MPa under static conditions as indicated by the arrows at the left margin of Fig. 4. The experimental error for ϕ_z is reflected by the scatter of the time-resolved data and estimated as about 3–4%. The electric field of 4 kV/cm is able to switch almost all of them into x- or y-domains at low stress, while it is much less effective when acting against a stress of 5 or 10 MPa.

Careful inspection of Figures 3 and 4 yields that even if the crystal reacts almost immediately to the electric field perturbation (within some 100 μs) this does not lead directly to the equilibrium state: Rather, at a competing stress level of 5(10) MPa only about 20(40)% of the z-domains are transformed by the electric field after some 100 μs while another 10% are stabilized over a much longer time. The arrows at the right margin of Fig. 4 show the domain fractions which are obtained at equilibrium under static conditions. Obviously, there is another relaxation mechanism on a time scale much longer than milliseconds that leads to a gradual increase in (1/2 1/2 7/2)-intensity (see Fig. 3) and a corresponding reduction of

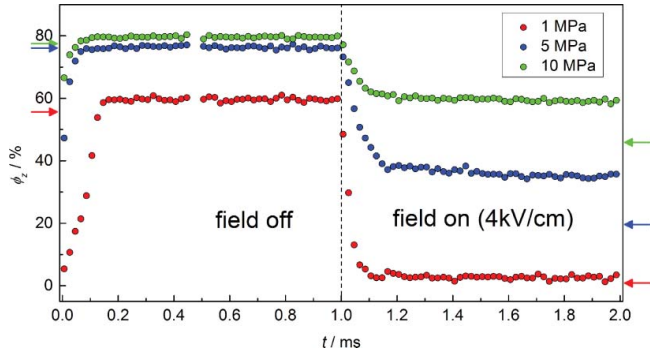

Figure 4. Time-evolution of the volume fraction of *z*-domains during a 2-ms-cycle of a pulsed electric field of 4 kV/cm along [001] at 20 K for different competing mechanical stresses along [110]. Arrows indicate the volume fraction under static conditions.

z-domains. This finding indicates that there are domain walls with different mobility that govern the kinetic behavior.

The short time behavior of the domain redistribution can be described by a (stretched) exponential function as shown in Fig. 3[2]. In the field-off period, a relaxation time of about 100 μs can significantly be determined if the stress bias is as low as 1 MPa, while at higher stress levels the domain distributions follows instantaneously the decay of the electric field due to the enhanced driving force of the mechanical load. If the field is switched on, the mobile fraction of domain walls is characterized by a relaxation time of some ten microseconds if the stress is not too small.

If the competing uniaxial stress is removed, the field induced domain kinetics is strongly modified: As an example, Fig. 5 shows the comparison of the switching behavior at 15 MPa and 0 MPa under the influence of a pulsed electric field of 8 kV/cm. Here, a sample was used which had an initial domain distribution of $\phi_x = 33.5\%$, $\phi_y = 14.3\%$ and $\phi_z = 52.2\%$. At 15 MPa both stress and electric field are strong enough to induce domain redistribution between the equilibrium values of about 75% and 40% within 100 μs, i.e. synchronously to the electric field (upper part of Fig. 5). If the stress is removed, however, the electric field of 8 kV/cm is strong enough to suppress almost all of the *z*-domains immediately. In the field-off period, the back transformation is delayed by orders of magnitudes since the restoring force is strongly reduced without any applied uniaxial stress. Hence, only a few percent of *z*-domains are recovered immediately while the redistribution of another 10% needs about 100 s.

This behavior is related to the transition into the field induced ferroelectric phase: The hysteresis loops of the electric polarization observed by Hemberger et al. [7] indicate that ferroelectric domains are conserved after the field is removed. Hence, also the tetragonal domain structure is frozen to a large extent. This can be demonstrated by varying the amplitude of the electric field. We used a sample which had a large predominance of z-domains (92%) if no external perturbation was applied. At 20 K and a field amplitude of 2 kV/cm - which is still below the ferroelectric transition - only about 20% of the z-domains are

[2] While the time constant is obtained with sufficient accuracy, the stretching exponent is rather ill defined and will not be discussed in the following.

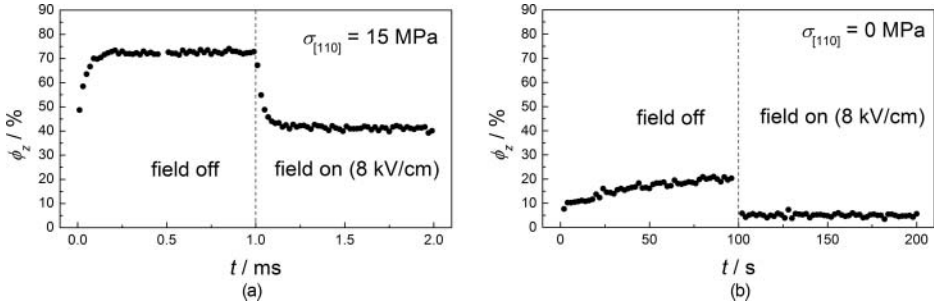

Figure 5. Time-evolution of the volume fraction of *z*-domains under the influence of a pulsed electric field of 8 kV/cm along [001] at 20 K with and without an applied mechanical stress along [110]. Please note the different time scales.

transformed reversibly as shown in Fig. 6 (upper curve). At an intermediate amplitude of 4 kV/cm, the crystal becomes ferroelectric and most of the z-domains are converted into *x*- and *y*-domains (lower curve of Fig. 4). The back transformation is, however, hindered due to the ferroelectric hysteresis and only 40% z-domains are recovered in the field-off period even under static conditions. Moreover, the kinetics is somewhat delayed with time constants $\tau_{on} = (303 \pm 36)\ \mu s$ and $\tau_{off} = (212 \pm 24)\ \mu s$.

This domain structure is preserved at zero field. On heating into the paraelectric phase above about 40 K, however, the pristine preference of more than 90% *z*-domains is reestablished as shown in Fig. 7. This finding indicates that the so-called coherent paraelectric phase [6] consists of polar clusters representing the remanent ferroelectric nanodomains with orthorhombic symmetry and polarization perpendicular to the tetragonal axis of structural domains [14]. Since the field induced polarization is along [001], only the *x*- and *y*-domains can be ferroelectric. Obviously, there is a well-defined transition into the paraelectric regime when the remanent polarization is destroyed near 40 K and the domain distribution is no longer influenced by electrostatic interactions.

If a competing external stress along [110] is statically applied, no such abrupt temperature variation is observed. The domain kinetics is shown in Fig. 8 for different temperatures

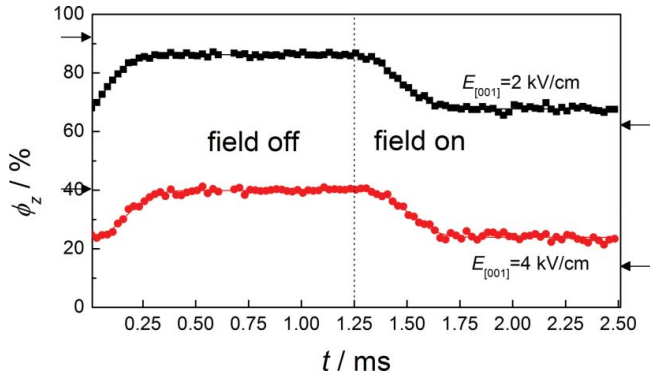

Figure 6. Time-evolution of the volume fraction of *z*-domains under the influence of pulsed electric fields with different amplitudes at 20 K. Arrows indicate the volume fraction under static conditions.
(Note that the rise time of the electric field is about 100 μs.)

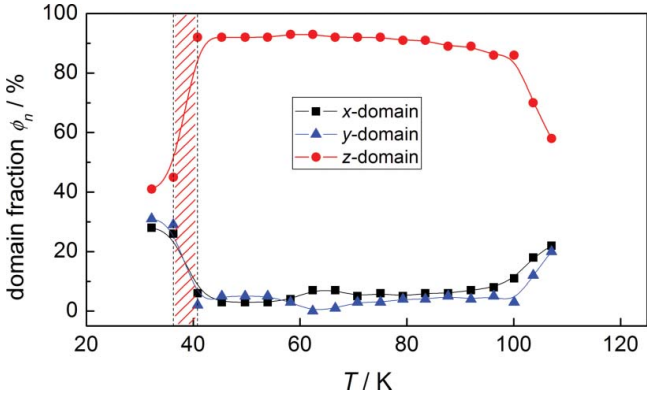

Figure 7. Temperature dependence of the domain distribution at zero field after having entered the ferroelectric phase at 30 K.

where a pulsed 8 kV/cm field was applied along with a mechanical stress of 15 MPa. It is clearly seen that the effect of the electric field in competition to the uniaxial stress decreases smoothly with increasing temperature in agreement with the findings of static experiments presented in [14]. While at 20 K, the fraction of *z*-domains decreases from 75% to 40%, the redistribution is limited to less than 10% at 65 K, leaving 65% of the *z*-domains unaffected. Due to the strong influence of the mechanical stress only 60% of the total volume is converted into *x*- and *y*-domains at 20 K. And only these become ferroelectric under the influence of strong enough electric fields [14]. In view of the existence of two types of hard and easy twin boundaries [11] it is likely that the redistribution of domains begins with the displacement of low-energy domain walls which are rather mobile and exhibit fast kinetics even if the ferroelectric phase is entered. Long-time relaxation processes are consequently associated with the displacement of the hard domain walls. Hysteresis effects are therefore less important and the temperature dependence does not show any anomaly similar to what is observed without any bias stress.

The time variation can be described by exponential functions with relaxation times τ_{off} in the field-off period being smaller than τ_{on} in the field-on period. As shown in Fig. 9, τ_{on} and

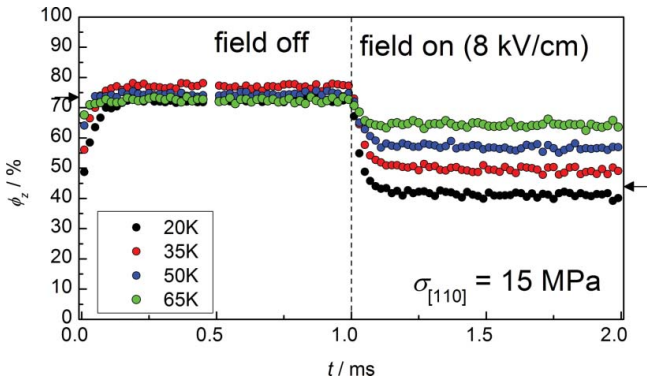

Figure 8. Temperature dependence of the field induced domain redistribution acting against mechanical stress along [110].

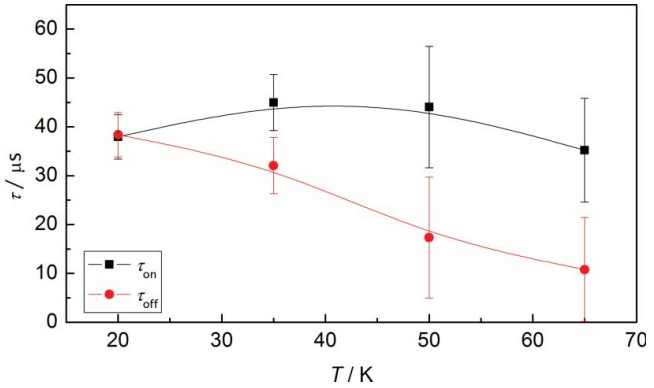

Figure 9. Temperature dependence of relaxation times for the field-induced domain-redistribution under a bias stress of 15 MPa.

τ_{off} are found to be identical at 20 K (38 μs). With increasing temperature, τ_{off} decreases significantly and reaches 10 μs at 65 K. This behavior is due to the fact that the permittivity and, hence, the capacity of the sample increases at low temperatures [13] and so does the RC time-constant of the electronic circuit. The domain redistribution thus follows directly the decreasing applied electric field. τ_{on}, on the other hand, is almost temperature independent and significantly larger than the charging time at 65 K. The driving force of the electric field of 8 kV/cm acting against a mechanical stress of 15 MPa is not large enough to allow for the immediate response of the crystal. This is not only reflected by the limited fraction of domains (about 8% at 65 K see Fig. 4) which are transformed by the field but also by the increased intrinsic relaxation time of about 30 ms.

Conclusion

The domain structure of $SrTiO_3$ can easily be affected by the application of uniaxial mechanical stresses or electric fields. While stress along [110] favors the tetragonal *z*-domains, electric fields along [001] can be used efficiently to form *x*- and *y*-domains. Moreover, a transition into the ferroelectric phase can be induced at sufficiently low temperatures. While on cooling below the antiferrodistortive transition at 105 K, each individual sample exhibits its characteristic domain distribution which seems to be determined by the conditions during crystal growth and preparation, the competition of stresses of the order 10 MPa and electric fields of several kV/cm are found sufficient to lead to an entire redistribution of domains. In real-time experiments with pulsed electric fields and different bias stresses, the kinetics of the domain relaxation is observed. Characteristic relaxation times of 10 to 100 μs are found. There are, however, at least two types of domain walls that can be distinguished by their mobility. If the field is acting against larger stresses, one part of the domains is transformed rapidly, while others need much longer times. The back transformation is strongly accelerated if the stress is increased. At higher temperatures, the electric field becomes less efficient and a smaller fraction of *z*-domains are converted with larger relaxation rate. If the electric field is strong enough to induce the ferroelectric phase below about 40 K, the z-domains are not fully recovered after removing the field due to hysteresis effects. Either application of

sufficiently strong competing mechanical stress or heating above 40 K leads to the pristine domain distribution.

If the field is applied along [001] only *x*- and *y*-domains can become ferroelectric. If *z*-domains are stabilized by mechanical stress along [110] these do not contribute to the overall polarization. In the field-off period, remanent polarization is associated with the existence of remaining *x*- and *y*-domains. The fact, that these are removed only after heating above the quantum-to-normal paraelectric phase supports the interpretation of the coherent- or quantum paraelectric phase as a glass-like structure with residual polar domains.

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