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Domain Redistribution and Ferroelectric Phase Transition in $SrTiO_3$ Under the Influence of an Electric Field and Mechanical Stress

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Strontium titanate ($SrTiO_3$) is one of the best studied substances showing a displacive phase transition [1]. At ambient temperature, it exhibits a cubic perovskite structure with the space group Pm3m. On cooling, a Brillouin zone-boundary softmode drives the second-order transition to an antiferrodistortive phase at $T_C = 105$ K. The primary order parameter is the antiphase rotation of TiO_6-octahedra in adjacent unit cells. As a result, a paraelectric, tetragonal phase with the space group I4/mcm is formed. The tetragonal distortion c/a is slightly larger than unity and represents a secondary order parameter of the transition. Without an external perturbation none of the three possible tetragonal axes is favored over the others and the twin domains are evenly distributed.

The isostuctural barium titanate ($BaTiO_3$) shows a completely different behavior: Here, the condensation of a Γ-point softmode leads to a ferroelectric low temperature phase. A softmode with the same polar eigenvector is also observed in strontium titanate but shows only an incomplete softening [2]. This incipient ferroelectricity is also revealed by the high dielectric permittivity at low temperatures. It is assumed that long range ferroelectric order is suppressed by quantum fluctuations [3]. However, a polar phase can be induced by the application of an external electric field larger than 2 kV/cm below 40 K [4].

Both, an electric field as well as mechanical load are able to affect the distribution of tetragonal domains in $SrTiO_3$. A quantitative relation is, however, not yet available. Therefore, we investigated the domain distribution in a single crystal of strontium titanate while applying uniaxial stresses and electric fields simultaneously. A pressure cell was constructed that enabled us to apply a uniaxial force of up to 2 kN and voltages up to several kV at low temperatures in a closed cycle cryostat. The lattice dynamics and the intensities of two tetragonal superlattice reflections were studied at the thermal three axis spectrometer PUMA at the FRM II neutron source over a wide range of temperatures. In addition a larger set of 15 superlattice reflections was measured at the hot neutron diffractometer HEIDI at the same facility to allow a quantitative determination of the domain fractions at selected temperatures.

A model of the structure factor was employed to determine the influence of the three domains on the observed intensities. For example the reflection R1 (1/2 -1/2 3/2) depends exclusively on the volume fractions of domains with their tetragonal axes in x and y direction, respectively, and is independent of the third domain. Similarly, the reflection R2 (1/2 3/2 -1/2) is only associated with the x and z axes.

The left part of Fig. 1 compares the results under the influence of mechanical load in [110] direction with a reference measurement. At 100 bar, the *z* domain is favoured over the *x* and *y* domains due to the ratio of the lattice constants of $c/a > 1$. This is reflected by the reduction of reflection R1, which is about twice as large as the increase of reflection R2. By fitting the complete dataset of 15 superlattice reflections to our model we could determine a ratio of over 90 percent for the *z* domain at 80 K under comparable conditions. This finding is in accord with earlier measurements that proved a nearly complete redistribution of the domains already at moderate stress [5].

The changes in the domain fractions is partly irreversible. Even after the removal of the mechanical load, the intensities remain almost unchanged. We also observed that even in pristine samples there is no uniform domain distribution. In one specimen we determined a *z* domain with a volume fraction of about 50 percent while the other two domains showed a smaller ratio of 25 percent. This may be caused by residual stress originating in the crystal growth at elevated temperatures or by the sample preparation.

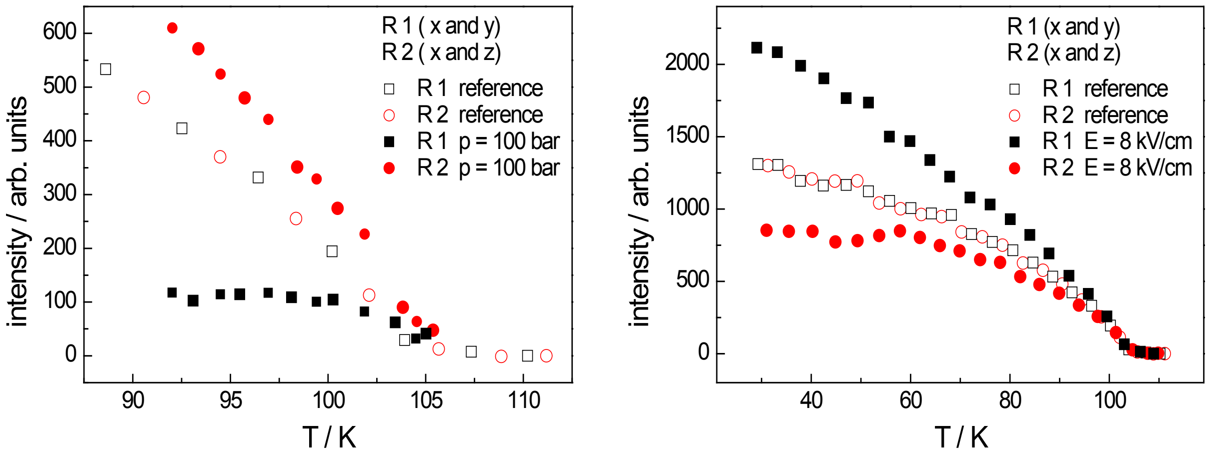

Fig. 1 Influence of mechanical stress in [110] direction (left) and an electric field of 8 kV/cm in [001] direction (right) on the intensity of the superlattice reflections.

As can be seen in the right part of Fig. 1, an electric field of 8 kV/cm along [001] leads to the opposite effect. The higher population of the *z* domain is reflected by the increase of the reflection R1. At sufficiently low temperatures, the electric field even overcompensates the effect of mechanical stress. Obviously, it is not possible to retain pressure oriented monodomain samples under the influence of an electric field along the tetragonal axis. Using the full set of 15 superlattice reflections, the redistribution of tetragonal domains is quantitatively determined. It is found that the electric field is even more effective than uniaxial mechanical stress. A direct coupling between the antiferrodistortive order parameter and the electric field is, however, prohibited by symmetry. The observed effect can probably be interpreted by a higher order coupling like piezoelectricity or electrostriction. This would cause a field induced mechanical strain that then leads to the changes in the domain distribution.

The lattice dynamics of strontium titanate are also affected by the electric field at low temperatures. The Γ-point softmode exhibits a strong history dependence. While after field cooling a broad intensity distribution is observed, two well defined components are visible when the electric field is switched on after zero field cooling. This may be connected to different domain configurations in both cases.

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