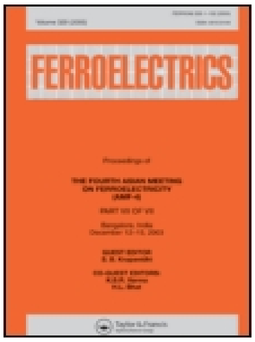

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DYNAMICS OF POLARIZATION REVERSAL IN PURIFIED Rb_2ZnCl_4

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Abstract The dynamics of polarization reversal in sinusoidal fields (0.1Hz≤f≤50kHz) was studied in the ferroelectric lock-in phase of purified Rb_2ZnCl_4 crystals. The frequency dependence of remanent polarization and coercive field E_c is qualitatively the same above and below $T^* \approx 160K$, where for all measuring frequencies E_c starts to increase rapidly with decreasing temperature. A model which treats sideways shifts of planar, nearly regularly arranged domain walls as the only mechanism of polarization reversal in Rb_2ZnCl_4 is applicable for the quantitative description of ferroelectric hysteresis loops in the whole temperature interval under investigation. The anomalous increase of E_c at T^* seems to be connected to a corresponding increase of the viscosity coefficient.

INTRODUCTION

Rb_2ZnCl_4 is known for its unusual "swan neck" shaped hysteresis curves at temperatures immediately below the incommensurate-commensurate phase transition temperature T_L which were reported by several authors[1-5]. Close below T_L, the dielectric properties in the lock-in phase seem to be connected with the peculiar domain structure which follows from the regular phase soliton lattice of the incommensurate phase[6,7]. Since the conversion of the solitons into the ferroelectric domain walls proceeds in a unique stripple mechanism[14,15], the spatial sequence of six domain states of the incommensurate phase is preserved below T_L. Theoretical considerations[10] predict that this sequence is retained even in electric fields usually used in repolarization experiments. Therefore it was suggested that the polarization reversal in

Rb_2ZnCl_4 takes place almost exclusively by sidewise shifts of a field independent number of planar domain walls and not simply via the steps nucleation-grow-coalescence of antiparallel domains as in proper ferroelectrics[11]. Basing on these conceptions, we have developed in our previous paper[5] a model for a quantitative description of the polarization reversal in purified Rb_2ZnCl_4 close below T_L.

Even in purified crystals however, the "swan neck" shape of the loops is lost during further cooling[4]. Below $T^* \approx 160K$, a strong increase of the coercive field determined at $f=50Hz$ was observed[12] as well as anomalies of a number of other quantities (see e.g. Ref 11-14). Moreover, a nonmonotonic field dependence of the permittivity was found at T^*. This was interpreted as the response of bounded domain pairs which may form at this temperature due to an oscillatory wall interaction potential[14]. Due to these results and the more convential shape of hysteresis loops at low temperatures[4], it may be supposed that the switching mechanism changes at T^* and the polarization reversal at low temperatures is dominated by the nucleation of new domains as it is known for proper ferroelectrics. We have studied the dynamics of polarization reversal in the lock-in phase of purified Rb_2ZnCl_4 crystals in sinusoidal measuring fields in the frequency range $0.1Hz \leq f \leq 50kHz$. Here we compare results obtained above and below T^*.

EXPERIMENTAL

The samples were prepared in the same way as described recently[5] from the same highly purified crystal. A Sawyer-Tower circuit in combination with an electrometer-amplifier and a current-voltage converter were used to measure polarization and switching current, respectively. The sinusoidal measuring voltage was stabilized to a constant amplitude $V_{max}=180V$ in the whole frequency range $0.1Hz \leq f \leq 50kHz$. The same measures were taken as in our previous study[5] to avoid dielectric heating of the sample. Before measuring at any given temperature was performed, the temperature was stabilized for 30 min in order to establish a stability $\Delta T < 0.1K$. The cooling/heating rate between any successive temperature spots was 2K/min.

RESULTS

Swan neck shaped hysteresis curves (Figure 1) which we have recorded close below T_L for low measuring frequencies f are characterized by an extremely small coercive feld $E_c < 10V/cm$ and a remanent polarization in the order of 30-50% of the polarization P_{max} obtained at the maximum value $E_{max}=2kV/cm$ of the measuring field. However, the coercive fields shows a peculiar temperature and frequency dependence. For all frequencies under consideration, the temperature dependence $E_c(T)$ suddenly changes its slope at T^* (Figure 2) as it was observed also for nominal pure crystals at f=50Hz[12]. A qualitatively similar increase of E_c, however, can be obtained in the lock-in phase for any temperature above and below T^* by increasing the frequency of the measuring field (Figure 3). Note that the critical frequency range where E_c starts to raise is shifted to lower values with decreasing temperature.

In what follows we discuss only saturated hysteresis curves ($P_r < P_{max}$, the low frequency branch in Figure 4) where P_r increases approximately logarithmically with increasing frequency. Once again, no qualitative difference is visible in the frequency dependence $P_r(f)$ above and below T^*. Therefore we assume that swan neck curves should appear also below T^* at extremely low measuring frequencies not tractable in experiment.

The model[5] we have recently developed to describe the dynamic P(E) dependence close below T_L treats the motion of planar, nearly regular arranged interacting domain walls in a viscous medium. Least square fits of data recorded below T^* (Figure 5) according to this model show a good agreement with the experiment both with respect to the polarization and the repolarization current. The only model parameter we found to have a significant anomaly at T^* is the viscosity coefficient which shows a steep increase below T^* comparable to those of the relaxation time of the small signal permittivity[15]. A more detailed analysis of the results of these fits will be given elsewhere[16].

The good agreement between experimentally recorded and calculated P(E) and I(E) hysteresis curves at low temperatures together with the qualitatively similar *frequency dependence* of E_c and P_r above and below T^* lead us to the conclusion that

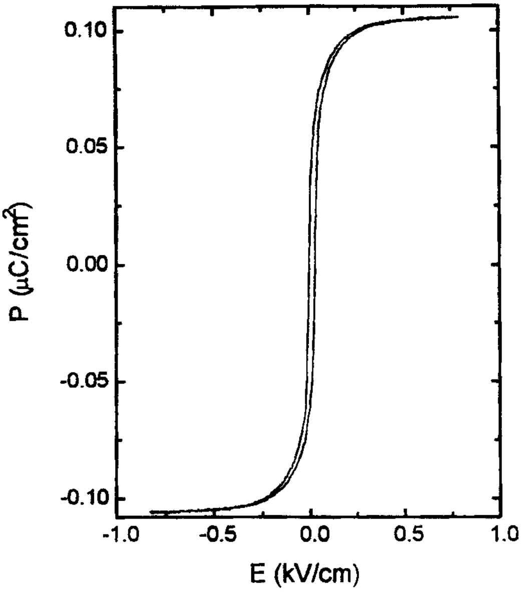

Figure 1: "Swan neck" shaped hysteresis loop recorded close below T_L (T= 191K; f=1Hz).

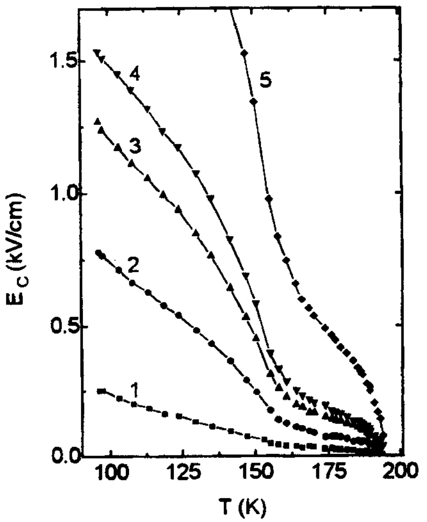

Figure 2: Temperature dependence of the coercive field determined on cooling for several measuring frequencies: 1 - 0.1Hz; 2 - 10Hz; 3 - 50Hz; 4 - 100Hz; 5 - 1kHz.

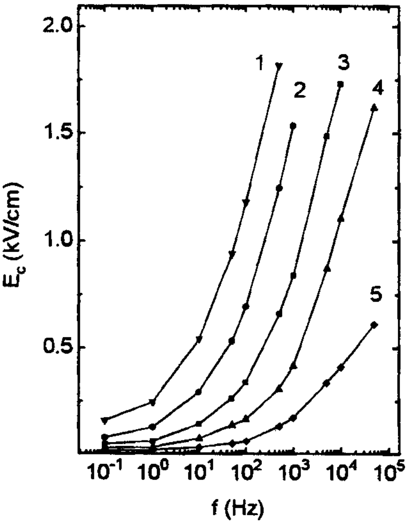

Figure 3: Frequency dependence of the coercive field determined on heating at several temperatures: 1 -123K; 2 - 147K; 3 - 158K; 4 - 174K; 5 - 186K.

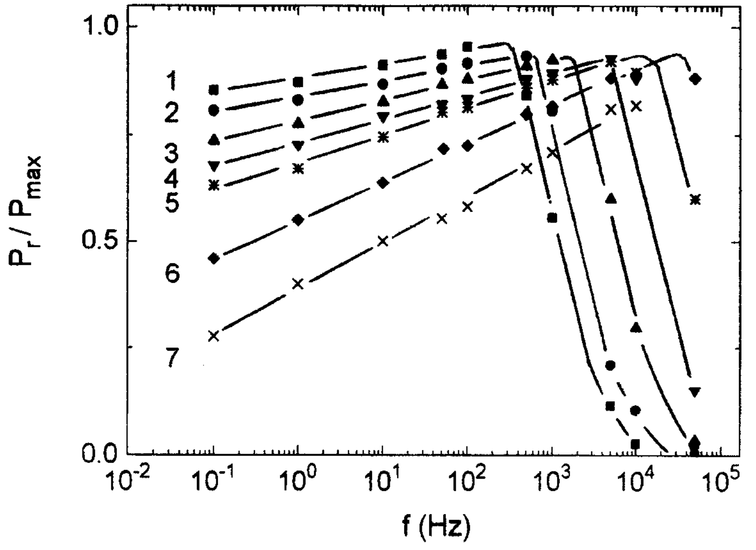

Figure 4: Frequency dependence of the normalized remanent polarization (see text) determined on heating for several temperatures: 1 - 124.7K; 2 - 137.5K; 3 - 150.6K; 4 - 156.9K; 5 - 166.0K; 6 - 185.1K; 7 - 191.0K.

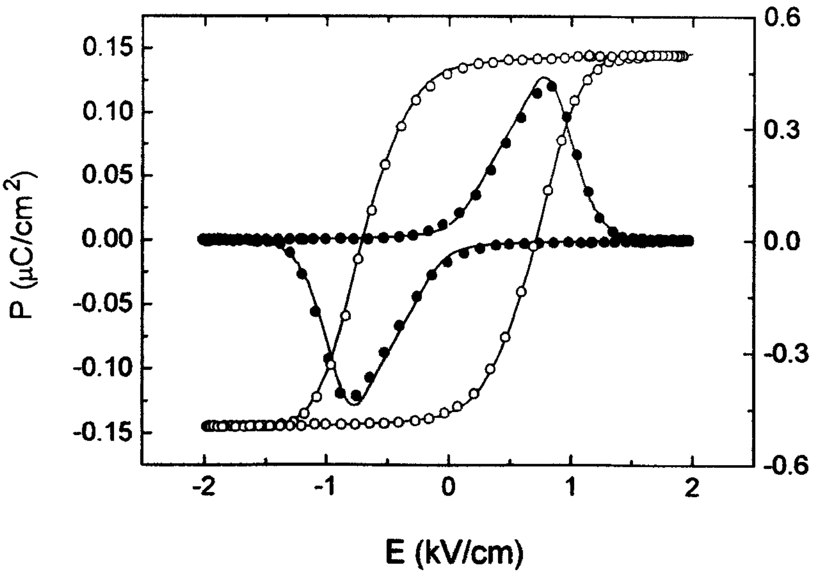

Figure 5: Ferroelectric hysteresis loops for polarization P and repolarization current I, respectively, recorded on heating at T=157K. The measuring frequency was 500 Hz. The lines are the results of a least square fit of the P(E) dependence according to our model (see text). The I(E) dependence was calculated without any adjustable fit parameter.

the predominant repolarization mechanism in Rb_2ZnCl_4 does not change at T^*. The polarization reversal at low temperatures seems to proceed as before almost exclusively by sidewise motion of a field independent number of domain walls. The steep increase of the viscosity coefficient may point to a change of the domain wall defect interaction at T^* what we will discuss elsewhere[16].

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