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Influence of side electric potential on hysteresis loop parameters and electric permittivity in the Rochelle salt

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ABSTRACT

It has been shown that the application of an electric potential to relatively narrow side electrodes of a thin perpendicular parallelepiped Rochelle salt sample plate leads to the disappearance of the hysteresis loop. The effect is permanent and can be observed after the side potential disconnection. Moreover, a reduction of both zero-field longitudinal permittivity maxima at the critical points is visible then. A non-zero remanent polarization and a non-zero coercive field are then able to be observed only at temperatures higher than that of the lower critical point and lower than that of the upper critical one. No corresponding temperature shift in reduced permittivity maxima has been noticed. A transition to below the lower critical point for the next few hours does not lead to restoration of the original properties formerly lost during the side potential application in the ferroelectric phase. Such restoration is possible by annealing the sample above the upper critical temperature.

1. Introduction

Dielectric properties of uniaxial ferroelectric crystals (triglycine sulfate, triglycine selenate, the Rochelle salt) placed in an electric field not parallel to the ferroelectric axis have been investigated in several papers. Among other unexpected effects, hysteresis loop disappearance and electric susceptibility reduction have been observed in the ferroelectric phase as a result of prolonged application of a transverse electric field [1–7]. It was interesting that the crystals did not return to their original (i.e. those observed before the transverse field action) ferroelectric states after the transverse field disconnection. For example, a completely or partly reduced hysteresis loop could be then still observed at the temperature of the former application of the transverse electric field provided that the measuring electric field was not applied for too long time [1,2]. However, the original properties could be easily restored by annealing the crystal above the critical temperature. This phenomenon, the so-called transverse field effect, seems to be particularly interesting in the ferroelectric Rochelle salt in which there exist two critical points. On the temperature axis, the ferroelectric phase is situated between two paraelectric ones. The upper critical temperature and the lower one are $T_U \approx 297$ K and $T_L \approx 255$ K, respectively (in the experiment described below $T_U = 297.1\ \text{K} \pm 0.2\ \text{K}$ and $T_L = 255.4\ \text{K} \pm 0.2\ \text{K}$, see text and Fig. 4).

The impact of different factors on the physical properties of the Rochelle salt was investigated in many experiments. The influence of pressure [8,9], magnetic or longitudinal external electric field [10,11], γ- and X-ray irradiation [12–14], deuteration [15,16] and other admixture doping [17] can be cited here as examples. Some of these effects are not permanent, e.g. a crystal sample starts returning to its original state just after the pressure or the longitudinal electric field has ceased to be applied. Besides, the pressure effect and the one of the longitudinal electric field application (for not too high values of both the pressure and the electric field) are reversible. On the other hand, the crystal doping process is usually permanent and irreversible. Below we show the transverse field effect which is in some sense both permanent and reversible (see also Refs. [5,18]). For the consequences of the application of an electric field not parallel to the ferroelectric axis may be observed in the Rochelle salt crystal also after the field disconnection. On the other hand, as mentioned above, the original properties can be easily restored by annealing the sample above the upper critical temperature and therefore the application of an external electric field not parallel to the ferroelectric axis is required to permanently change the crystal again.

There arises a question of what will happen if the crystal previously influenced by a transverse field at temperatures $T_L < T < T_U$ is cooled down to below T_L. In other words, one can ask whether the restoration of the original shape of the hysteresis loop and electric permittivity values is a result of heating to a

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higher temperature (thermally activated effect) or it is only the consequence of a temporary transition to a state with a zero spontaneous polarization value, i.e. to the paraelectric phase (spontaneous polarization impact on the transverse field effect). The ferroelectric Rochelle salt, with its two critical points, seemed to be a good candidate for investigations that might give the answer to this question.

2. Experiment and results

The present paper shows the results of measurements of the hysteresis loop and electric permittivity in the Rochelle salt. The crystal was grown at Toyama University in Japan. The sample had the form of a perpendicular parallelepiped of dimensions 3.45 mm × 9.2 mm × 17.1 mm. Fig. 1 depicts the sample–electrode set as well as the coordinate system axes, with the ferroelectric *a* axis. Apart from two oppositely situated measurement silver paste electrodes, one can see also a pair of narrow, 1.3 mm wide silver paste side electrodes connected to each other by a fine silver wire. An alternating sine wave measuring signal was applied to the measurement electrodes, whereas a constant electric potential could be connected to both side electrodes. Due to such an electrode configuration, on one hand, the transverse field was clearly inhomogeneous inside the crystal. However, the advantage of such a shape of the sample is that it can be treated as a relatively thin crystal plate, i.e. its geometry is similar to that chosen in the majority of former classic experiments carried out in electric fields almost parallel to the ferroelectric axis. As it is shown below, the electric potential applied by means of the narrow side electrodes attached to the thin crystal plate may induce similar permanent changes in physical properties as in the case of the almost homogeneous transverse electric field [5,18]. The hysteresis loop parameters and permittivity values, modified by the additional V_S parameter, can be measured by means of the Sawyer–Tower circuit or the LCR meter more accurately than those shown in the former papers [5,18], where due to the crystal geometry ('thick' perpendicular parallelepiped samples) the measuring field was markedly inhomogeneous and the edge effects played a more significant role.

The measurement technique was described in Ref.[19], though in the present paper two stripe-like electrodes were attached to the crystal instead of a single ring one. Just before the measurements the sample was rejuvenated at 308.2 K for 18 h. After that the classic measurements of remanent polarization and coercive field temperature dependences were carried out (the first stage of the experiment). The temperature controller Oxford ITC 503 was used. The amplitude and the frequency of the measuring field of the Sawyer–Tower circuit were 58 kV/m and 100 Hz, respectively. At the second stage, after annealing the sample at 308.2 K for an hour, the constant potential $V_s=500$ V was applied continuously to the side electrodes at a constant temperature of $T_A=289.6$ K. At the same time the measurement electrodes remained grounded, except for short intervals (of a few seconds each) when the hysteresis loop was observed. More and more reduced hysteresis loops were then measured for a few days by means of the same Sawyer–Tower method (with the same measuring field parameters as those used during the first stage). The side potential V_s was applied for 86 h, i.e. the time after which the hysteresis loop disappeared. Fig. 2 shows hysteresis loops observed at $T_A=289.6$ K before the side potential application (a) and 16 h (b), 40.5 h (c), 67 h (d) and 86 h (e) after it was switched on.

After the side potential disconnection at $T_A=289.6$ K, the sample was cooled down to 246.2 K, i.e. to below the lower critical point T_L, and then heated. The time period during which the temperature of the sample was lower than T_L was not controlled precisely. However, on the basis of the cooling and heating process analysis we can state that this term was not shorter than ten hours. In Fig. 3(a) and (b) one can see that the non-zero remanent polarization and the coercive field observed in the heating (solid circles) appear at a temperature higher than the lower critical point T_L. Moreover, they tend to zero again not at the upper critical temperature T_U but at a somewhat lower one, i.e. close to the temperature $T_A=289.6$ K at which the side potential was formerly applied. In this way quite new temperature dependences of the remanent polarization and the coercive field were obtained in the heating, starting from the lower paraelectric phase. It should be stressed that a similar, though not so convincing, reduced polarization dependence was shown in Ref.[18]. However, it was measured when the temperature was lowered just after switching off the transverse field, which means that the sample was not previously cooled to a temperature below T_L. The results shown in Fig. 3 indicate that the transition to a temperature below T_L for the next few hours does not lead to the rejuvenation of the sample and new properties previously induced by the potential V_s are preserved. Although the sample

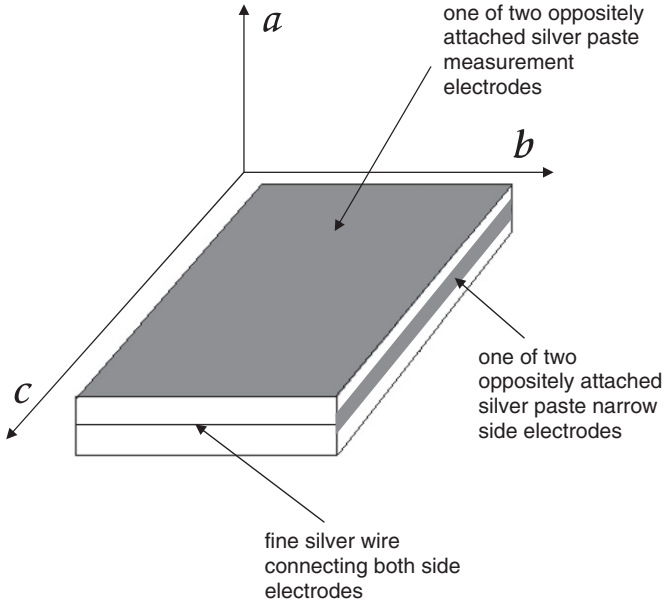

Fig. 1. Rochelle salt sample with painted four silver paste electrodes in the orthorhombic coordinate system (*a* —ferroelectric axis).

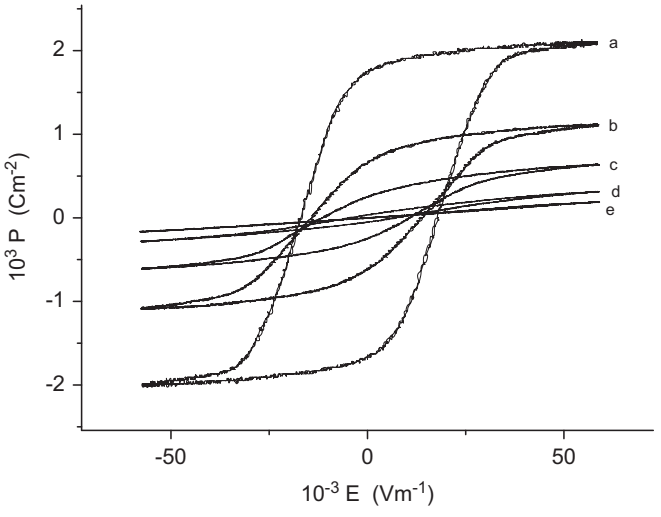

Fig. 2. Hysteresis loops observed at 289.6 K, before (a) and at different times: 16 h (b), 40.5 h (c), 67 h (d) and 86 h (e) after switching on the potential V_s.