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# Phase coexistence in DKDP with a thermal gradient parallel to the ferroelectric axis

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Received 6 August 1993, in final form 17 November 1993

Abstract. The coexistence of the paraelectric and ferroelectric phases is reported for thermally gradiented samples of DKDP; the shape of the phase front, the relevant domain textures and the dielectric properties are described. A phenomenological model allows us to evaluate the contribution of the domains to the dielectric constant.

## 1. Introduction

At a temperature around 215 K, KD<sub>2</sub> PO<sub>4</sub> (DKDP) crystals exhibit a first-order transition between a tetragonal paraelectric phase (space group  $\overline{42d}$ ), which is the high-temperature phase, and an orthorhombic ferroelectric and ferroelastic one (mm2). The ferroelectric domains appearing at the transition have not been studied as much as those in KH<sub>2</sub>PO<sub>4</sub> (KDP) (Bornarel 1987) but they look alike with permissible walls (Fousek and Janovec 1969) in (100) and (010) tetragonal planes. These domains are also mechanical twins and the importance of the mechanical energy in the domain texture of KDP type crystals has long been known (Bornarel and Lajzerowicz 1968). The DKDP paraelectric-ferroelectric (PF) transition has been studied by means of macroscopic measurements such as specific heat (Strukov et al 1968), dielectric susceptibility (Strukov et al 1972), polarization measurements (Sidnenko and Gladkii 1973), dielectric and electrocalorific properties (Reese 1969); x-ray (Andrews and Cowley 1986) or neutron (Nelmes et al 1985) diffraction studies also provide information integrating the whole sample. The first information on the phase front was presented (Zeyen et al 1976) with the intention of interpreting neutron diffraction measurements: the authors suggest that during the phase coexistence, paralelectric and ferroelectric stripes perpendicular to the c ferroelectric axis alternate in the sample. The validity of this prediction was confirmed by synchrotron x-ray topographies (Aleshko-Orzevskii 1982, 1983, 1992) and optical observations (Bachheimer et al 1981). These results demonstrated the greater importance of elastic energy than electrostatic energy at the transition: electrostatic energy is minimized by a phase front parallel to c, which avoids the charges on the front, elastic energy would be minimized by a phase front orientation perpendicular to c because the  $u_{xx}$  and  $u_{yy}$  values (10<sup>-4</sup>) are lower than the  $u_{zz}$  one (6.5 × 10<sup>-4</sup>) if a model based on the Khachaturyan work is used (Khachaturyan 1967, Bastie et al 1980). However, other models could obviously be applied to explain the observed phase front shapes.

Recently, optical observations performed in transmitted light along the three tetragonal axes simultaneously with dielectric measurements reveal different phase front shapes such as daggers, zig-zag or quasiplanar fronts (Bornarel and Cach 1991, 1993). These results need to be studied thoroughly. Firstly, concerning all phase front shapes, no part of the phase front makes an angle greater than  $22^{\circ}$  of arc with the (001) plane. Secondly, at the beginning of the coexistence in a PF transition, the dielectric constant along the *c* axis still follows a Curie–Weiss law and the small ferroelectric regions that appear seem to be monodomain ones (Bornarel and Cach 1993). As the phenomena in a phase coexistence are strongly dependent on experimental conditions—quality and shape of the sample, external applied field, temperature rate and gradient—systematical studies versus these parameters have been undertaken.

In the present paper results are reported on phase coexistence without external applied fields, and with a thermal gradient G parallel to the c ferroelectric axis, which favours a quasiplanar (001) orientation for the phase front. In the next section the experimental procedure and conditions are pointed out. In the following section the phenomena occurring during PF transitions and during ferroelectric-paraelectric (FP) transitions are described. The results of the simultaneous dielectric measurements are given (for the dielectric constant  $\varepsilon'$  and the loss constant  $\varepsilon''$  in the c direction). In the last section, a phenomenological model allows us to interpret the dielectric measurements, and the phase front and domain observations are analysed.

## 2. Experimental procedures

The DKDP crystals were grown by slow cooling of a supersaturated solution of KDP and heavy water. The observed transition temperature corresponds to a deuteration concentration of 83% for the crystal. The weak tapering angle of the crystal (a few degrees of arc), the room temperature optical studies and the dielectric properties confirmed the good quality of the sample. The sample was cut with a wire saw. The orientations of samples faces were verified with x-ray Bragg diffraction (accuracy of a minute of arc) and each face was polished on a wet silk cloth with an alcohol and heavy water solution. The sample dimensions were  $a_1 = 2.41$  mm,  $a_2 = 4.71$  mm, c = 9.02 mm. Semi-transparent gold electrodes were evaporated on the c faces. The sample is placed as shown in figure 1(a)in the helium gas exchange chamber of a cryostat, allowing optical observations in the three tetragonal directions simultaneously with dielectric measurements. The c ferroelectric axis is vertical, as is the thermal gradient G existing in the cryostat. This thermal gradient in the c (vertical) direction is a consequence of the cryostat design and changes linearly with the temperature rate R as indicated in figure 1(b). The thermal gradient G, directly measured by three platinum resistances, can be determined by the value of R. The G values cannot be exactly the same for a cooling cycle (PF transition) and for a heating one (FP transition) but |R| has always been lower than  $10^{-2}$  K min<sup>-1</sup> which induces G values equal to  $0.08 \pm 0.01$  K mm<sup>-1</sup>. The thermal gradient values in directions perpendicular to c are about  $10^{-3}$  K mm<sup>-1</sup> or under and this is considered to be negligible for the rest of the paper. The temperature may be regulated with an accuracy of a few mK. The sample capacitance and the dissipation factor measured with an impedance meter HP 4274 A allow us to calculate  $\varepsilon'$  and  $\varepsilon''$  values with a relative accuracy of  $3 \times 10^{-3}$  and  $10^{-2}$  respectively (measuring an electric field of  $10^{-1}$  V cm<sup>-2</sup> in amplitude and 4 kHz in frequency). As already described in a previous paper (Bornarel and Cach 1993), the observations along the  $a_1$  and  $a_2$  axes allow us to rebuild the phase front shape, and the observation along the c

axis gives information on the ferroelectric domain texture. Because of the great thickness of the sample in the c direction, it is not easy to resolve the domain texture finely, but the directions of the domain walls and the limits of domain species (regions with the same domain wall orientation) appear clearly.



Figure 1. (a) Schematic representation of the DXDP sample with the vertical thermal gradient G parallel to the c axis. The quasiplanar phase front perpendicular to c is shown. (b) Variation of the thermal gradient G versus the temperature rate R.

## 3. Results

#### 3.1. Description of the phase coexistence during a PF cycle

The phase coexistence during a cooling cycle is illustrated in figure 2 by two sequences of photographs. The first sequence concerns photographs taken along the  $a_1$  direction and shows the phase front displacement in the sample (figure 2(a)). The second one corresponds to photographs taken along the c direction, revealing the ferroelectric domain texture. The phase front appears in the sample's lower face, which is the cooler one, at a temperature of 211.9 K (all the temperatures given in this section result from the measurement performed by the platinum resistor near the lower c face of the sample). Sometimes it immediately crosses the whole sample in the  $a_2$  direction (the microscopic observation allows us to detect a ferroelectric region of a few micrometres), but usually the first ferroelectric nucleus is created in one of the lower corners of the sample. The phase front does not make an angle greater than 20° of arc with the (001) plane. Then, the dimension of the nucleus increases quickly in the  $a_2$  direction until the ferroelectric region overruns all the lower part of the sample as illustrated by the first photograph in figure 2(a). In this photograph the phase front traces appear on the  $a_1$  faces and figure 2(a) examples show that the phase front almost perpendicular to c can be quasiplanar or have a slightly distorted shape. In all cases, the angle between the phase front and the (001) plane, observed in section a1, does not exceed a few degrees of arc. The position of the phase front in the sample varies linearly

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Figure 2. PF phase coexistence observed during the PF transition: (a) photographs of the phase front observed along the  $a_1$  direction; (b) corresponding photographs of the domain texture obtained along the *c* direction. The photographs show the total sample faces; the magnification in (b) is greater than that in (a).



Figure 2. Continued.

with the temperature as shown in figure 3 for successive PF cycles. If the temperature does not change, the phase front remains in the same place, and then moves again on cooling. This is obviously due to the important value of the thermal gradient G. The end of the PF transition resembles the beginning of the phase coexistence: in the  $a_1$  section the phase front near the upper sample face (001) disappears by moving in the  $a_2$  direction with a phase front angle of about 15° of arc in relation to the (001) face. The photographs of the c section shown in figure 2(b) give poor quality information on the ferroelectric domain texture because of the sample's great thickness in this direction. However, it can be noted that if the domains are not observable at the beginning of the PF transition, they are clearly visible when the ferroelectric region is larger. The variations of the species, the regions with the domains parallel to one of the two permissible orientations, are easily observable and domain rearrangements are manifest between photographs and in figure 2(b). If the cooling rate R is too large (for example  $10^{-1}$  K min<sup>-1</sup>), the temperature diffusion in the sample induces a non-planar phase front. In contrast, when R is lower than  $10^{-2}$  K min<sup>-1</sup>, the quasiplanar shape of the phase front is very reproducible for different PF transitions at a given position in the sample. This is demonstrated in figure 4: when the phase front is in a position defined by a d value in figure 3, its shape is the same for successive PF cycles (figure 3(a)), which proves that the local strains play a role in the phase front shape. Furthermore, this strain influence seems very sensitive as the domain textures are different in such cases, proving the good quality of the sample (figure 3(b)). In conclusion, the phase front in a DKDP sample with an important thermal gradient G parallel to the c ferroelectric axis has a quasiplanar shape perpendicular to c. The domain textures change with the thermal cycles, which proves the sample's good quality. The small distortions of the phase front related to the (001) plane are always smooth which suggests a small elastic surface energy of the phase front itself. These distortions are correlated with the local strains in the sample.

# 3.2. Description of the phase coexistence during an FP cycle

The FP transition appears to be very like the PF one along the  $a_1$  direction: the small paraelectric nucleus that appears in an upper corner of the sample has a triangular shape, with the angle between the phase front and the (001) face making about 15° of arc. The paraelectric region increases in the  $a_2$  direction until it overruns the uppermost part of the sample. Then, the quasiplanar phase front moves linearly with the temperature as shown in figure 3 for PF cycles, but obviously in the opposite direction. The disappearance of the ferroelectric region in the  $a_1$  section also follows a process inverse to that already described. The shape of the phase front is quasiplanar if the heating rate is lower than  $10^{-2}$  K min<sup>-1</sup> and the small distortions depend on their position in the sample as for the PF transition, and certainly on the local strains as well. To summarize, the observation of the phase front along the  $a_1$  direction gives similar results both in the FP transition and the PF one, with time and temperature inversion.

On the other hand, observations in the c direction are not similar: some domain arrangements can be observed if the PF-FP thermal cycle is performed very quickly, but if the experiment is carried out in good conditions, with stabilization for a few hours in the ferroelectric phase at low temperatures and a temperature rate always lower than  $10^{-2}$  K min<sup>-1</sup>, then the domain texture remains unchanged during the whole of the FP transition. The domain texture disappears only when scanned by the phase front. However, at the end of the FP transition, the optical contrast of the domains becomes softer, which is coherent with the decreasing of the birefringence near the transition and the widening of the domain wall width.



Figure 3. Position of the quasiplanar phase front, almost perpendicular to the c axis versus the temperature of the lower sample face.

## 3.3. Dielectric measurements during the phase coexistence

Typical measurement of the dielectric constant  $\varepsilon'$  and the loss constant  $\varepsilon''$  in the *c* direction are given in figure 5. During a cooling cycle, in the paraelectric phase,  $\varepsilon'$  follows a Curie-Weiss law  $\varepsilon' = M/(T - T_0)$  (with in the example of figure 5(a)  $M = 3740 \pm 70$  K and  $T_0 = 210.4 \pm 0.2$  K) and  $\varepsilon''$  exhibits small values (around 1). When the phase front appears in the sample at the 211.71 K temperature  $\varepsilon''$  increases a little, but  $\varepsilon'$  continues to follow the Curie-Weiss law until the domains become observable (until 211.45 K). Below 211.45 K the  $\varepsilon''$  value increases and  $\varepsilon'$  departs from the Curie-Weiss law. Each jump in the  $\varepsilon''$  curve versus the temperature corresponds to important modifications in the domain texture. The end of the phase coexistence with the disappearance of the phase front at T'' = 210.7 K is clearly visible on  $\varepsilon'(T)$  and  $\varepsilon''(T)$  curves. In the ferroelectric phase, the peaks or jumps below this temperature correspond to rearrangements of the domain texture, as has already been observed in KDP at a few degrees below the transition (Bornarel 1984).

A heating cycle allows us to carry out a FP transition after remaining 10 h at low temperature and stabilization of the domain texture inducing lower values of  $\varepsilon'$ . As shown in figure 5(b) for temperatures just below the coexistence region,  $\varepsilon'$  follows approximately a Curie-Weiss law  $\varepsilon' = M'/(T_1 - T)$  with  $M' = 1821 \pm 5$  K and  $T_1 = 211.5 \pm 0.2$  K. The phase front appears at 210.66 K and with an increasing paraelectric region, decreasing  $\varepsilon''$ values are measured. During the phase coexistence  $\varepsilon'(T)$  curve leaves the low temperature Curie-Weiss law to join the paraelectric Curie-Weiss law at 211.30 K where the phase front disappears. In the paraelectric phase the  $\varepsilon'(T)$  curve fulfils the Curie-Weiss law well, whereas the  $\varepsilon''$  values become very small above 211.3 K.

For successive cooling and heating cycles (PF and FP transitions) what is reproducible and what is not in the  $\varepsilon'(T)$  and  $\varepsilon''(T)$  measurements? First, as already noted, the  $\varepsilon'$  value at a given temperature is always greater during a PF transition than the value at the same temperature during an FP transition, except in the paraelectric phase. This is illustrated in figure 6 where the inverse of  $\varepsilon'$  is drawn versus T for a PF cycle ( $R = -1.5 \times 10^{-2}$  K min<sup>-1</sup>, and for the temperature limits of the phase coexistence T''' = 211.195 K, T'' = 210.275 K) and a FP one ( $R = 1.5 \times 10^{-2}$  K min<sup>-1</sup>, T''' = 211.010 K, T'' = 210.275 K). This example leads us to recall that the thermal gradient in the present experiment cannot be exactly the same in a PF and an FP transition, so the measured temperature at the lower c face of the sample does not exactly correspond to the same physical situation. This is illustrated



B1

Cı



Figure 4. Shape of the phase front and domain texture during three successive PF transition: (a) photographs of the phase front in the  $a_1$  direction during PF cycles A, B, C in two different positions  $d_1$  and  $d_2$  (subscripts 1 and 2 respectively); (b) corresponding photographs of the domain texture. The photographs show the total sample faces; the magnification in (b) is greater than that in (a).





Figure 5. Variation of the dielectric constant  $\varepsilon'$  and the loss constant  $\varepsilon''$  versus the temperature T of the lower c sample face. Fine vertical lines show the phase coexistence temperature interval: (a) PF transition; (b) FP transition.

in figure 6 by the straight lines, which correspond to the Curie-Weiss, law fitted on the experimental  $\varepsilon'$  measurements in the paraelectric phase in both temperature cycles (PF or FP). The temperature is measured with an accuracy of a few mK, but the real uncertainty depends more on the sense of the temperature rate R (cooling or heating). The fact that the values of  $\varepsilon'(T)$  are higher in a PF cycle than a FP one is obviously due to the domain texture existence and so if these curves are relatively reproducible in FP transitions due to the stabilization of the domain texture, they may be very different from a PF transition to another. Figure 7 demonstrates this fact: with the same sample, the measured  $\varepsilon'(T)$  and  $\varepsilon''(T)$  curves are very different from those shown in figure 5(a) with a relatively similar cooling rate  $(-1.8 \times 10^{-2} \text{ K min}^{-1}$  in figure 5(a), and  $-2.4 \times 10^{-2} \text{ K min}^{-1}$  in figure 7). However, during the phase coexistence illustrated in figure 5(a), many small domain texture in the ferroelectric region remains unchanged up to the disappearance of the phase front in the sample. This event triggers off an important rearrangement of the domain texture with simultaneous abrupt modifications of  $\varepsilon'$  (drop) and  $\varepsilon''$  (jump) values.

Therefore, dielectric measurements can provide interesting information both on the phase coexistence and the domain texture. In this particular experimental situation with a thermal gradient in the sample, it seems necessary to distinguish contributions due to the front and domains from others due to the ferroelectric and paraelectric monodomain phases.

# 4. Discussion

Let us try to estimate the measured dielectric constant  $\varepsilon'$  versus the temperature in a sample such as that in figure 1 with the thermal gradient G parallel to the c axis.

It can be assumed, for a first-order transition close to the second-order one, that at constant temperature T, constant pressure and no external field applied, the simplest thermodynamical potential (for a monodomain sample) can be written as follows:

$$\Phi = \Phi_0 + \frac{A_0}{2}(T - T_0)P^2 - \frac{BP^4}{4} + \frac{CP^6}{6} + \cdots$$
 (1)



Figure 6. Reciprocal values of  $\varepsilon'$  versus the temperature T of the lower c face of the sample during a PF transition (open circles) and an FP transition (black circles). The temperature limits of the phase coexistence are shown by vertical lines (continuous for PF, dashed for FP). Two straight lines indicate the best fit obtained for the Curie-Weiss law in each case.



Figure 7. Variation of  $\varepsilon'$  and  $\varepsilon''$  versus T with an unchanged domain texture during the phase coexistence whose temperature limits are indicated by two vertical dashed lines.

with P the polarization as order parameter and  $A_0$ , B and C positive constants in temperature. The inverse of the dielectric susceptibility, estimated about  $1/\epsilon'$ , follows in the paraelectric phase a Curie-Weiss law

$$\frac{1}{\varepsilon_p'} = A(T - T_0). \tag{2}$$

The thermodynamical potential, which was a minimum for P = 0 in the paraelectric phase, adopts at the Curie temperature  $T_C$  the same value for P = 0 and for the spontaneous polarization  $P_s$  value given by

$$P_s^2 = \frac{B + [B^2 - 4A_0C(T - T_0)]^{1/2}}{2C}$$
(3)

with also

$$T_{\rm C} = T_0 + \frac{3}{16} \frac{B^2}{A_0 C}.$$
 (4)

In such a phenomenological model, the inverse of  $\varepsilon'$  in the ferroelectric region becomes

$$\frac{1}{\varepsilon_{\rm F}'} = 4A_0(T_1 - T) + \frac{2A_0^{1/2}B}{C^{1/2}}(T_1 - T)^{1/2}$$
(5)

with  $T_1 - T_0 = B^2/4A_0C$  the temperature interval where both phases, paraelectric and ferroelectric, can exist by metastability.

In the present case, the temperature of the sample is not homogeneous and varies with the position z in the c direction. The macroscopic capacitor C can be considered as the series combination of many elementary capacitances

$$1/\mathcal{C} = \int_0^e \frac{\mathrm{d}z}{\varepsilon_0 \varepsilon'(z)S} \tag{6}$$

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where e is the sample dimension in the c direction, S its section perpendicular to c, and  $\varepsilon'(z)$  the dielectric constant value of a monodomain region at the z position, i.e. at the temperature  $T_z = (T' - \Delta T/2) + (z/e)\Delta T$ . T' is the mean temperature of the sample;  $\Delta T$  is the difference between the upper and lower face temperatures. The distribution of the temperature inside the sample is considered to be linear in the c direction (G = DT/e). The dielectric constant  $\varepsilon'$ , when the phase front separating paraelectric region and the ferroelectric monodomain one (hypothesis) is at position d, can be obtained by

$$\frac{e}{\varepsilon'} = \int_0^d \frac{\mathrm{d}z}{\varepsilon_{\mathrm{F}}'(z)} + \int_d^e \frac{\mathrm{d}z}{\varepsilon_{\mathrm{p}}'(z)}.$$
(7)

When all the sample is in the paraelectric phase, (7) becomes

$$\frac{1}{\varepsilon'} = A_0(T' - T_0) \tag{8}$$

which is the classical Curie-Weiss law. This Curie-Weiss law (8) differs from that observed in isothermal conditions T by a translation in temperature of the curve. This translation depends on the thermal gradient (the measured temperature of the lower c face of the sample is equal to  $T'\Delta T/2$ ).

When all the sample is monodomain, in the ferroelectric phase,  $1/\epsilon'$  can be written

$$\frac{1}{\varepsilon'} = 4A_0(T_1 - T') + \frac{B}{6A_0C^2\Delta T} \times \left[ (4A_0C)^{3/2} + \left( \left( T_1 - \left( T' - \frac{\Delta T}{2} \right) \right)^{3/2} - \left( T_1 - \left( T' + \frac{\Delta T}{2} \right) \right)^{3/2} \right) \right].$$
(9)

Finally, when the phase front is in the sample, assuming that the temperature at the phase front level is the Curie temperature  $T_C$ , the dielectric constant during this phase coexistence can be described as follows:

$$\frac{1}{\varepsilon'} = \frac{1}{\Delta T} \left\{ \frac{B^4}{48A_0C^2} + \frac{A_0}{2} \left( T_{\rm C} - \left( T' + \frac{\Delta T}{2} \right) \right)^2 - \frac{3}{16} \frac{B^2}{C} \left( T_{\rm C} - \left( T' + \frac{\Delta T}{2} \right) \right) \right\} + 2A_0 \left( T_{\rm C} - \left( T' - \frac{\Delta T}{2} \right) \right)^2 + \frac{B^2}{4C} \left( T_{\rm C} - \left( T' - \frac{\Delta T}{2} \right) \right) + \frac{B}{6A_0C^2} \left( \frac{B^2}{4} + 4A_0C \left( T_{\rm C} - \left( T' - \frac{\Delta T}{2} \right) \right) \right)^{3/2} \right\}.$$
(10)

Using the values of  $T_0$ ,  $T_C$  and  $\Delta T$  given by the experiments, for each  $\varepsilon'(T)$  curve and the value previously calculated for  $B = 4.94 \times 10^{10}$  SI (Gladkii and Sidnenko 1972), it is possible to calculate with the help of (8), (9) and (10) the values of the dielectric constant  $\varepsilon'$ , called  $\varepsilon'_{mono}$  since this value corresponds to a monodomain sample in the ferroelectric region. Figure 8(a) shows the result corresponding to the case of the PF transition in figure 5(a). The difference between the measured  $\varepsilon'$  values and the calculated value  $\varepsilon'_{mono}$  is plotted versus the temperature in figure 8(b) (PF transitions) and 8(c) (FP transitions) with a





Figure 8. (a)  $\varepsilon'(T)$  curves: measured values (open circles) and values calculated using the phenomenological model (continuous line). (b, c) Difference between the measured values of  $\varepsilon'$  and the calculated values  $\varepsilon'_{mono}$  versus the temperature of which the origin corresponds to the phase front in the sample's lower c face: (b) PF transition; (c) FP transition.

temperature origin that corresponds to the phase front in the lower part of the c face of the sample.

It is interesting to discuss the physical meaning of the  $\varepsilon' - \varepsilon'_{mono}$  variation in relation to the optical observations of the phase front and domain textures. Firstly, it must be noted that the thermal gradient G equal to about 0.08 K mm<sup>-1</sup>, suffices to impose a phase coexistence situation as shown in figure 1. Then, the phase front can be blocked in a position d for a very long time. Without the thermal gradient or with weak G values, the phase coexistence period would depend only on the intrinsic properties of the sample (defects, thermal conductivity) and could happen in theory at an almost constant temperature. The gradient G is like an external applied field and the present results must be understood in this context.

On the phase front itself, the dielectric constant measurements do not give any information since the phase front is almost the same during all the phase coexistence and the domain contribution in  $\varepsilon'$  is very large. Figure 8(b) and 8(c) is compatible with the optical observations and previous results: during a PF cycle, many fluctuations and rearrangements of the domain texture induce jumps in  $\varepsilon''(T)$  curves and higher values of  $\varepsilon'(T)$  than those observed during the FP cycles. However, the beginning of the PF cycle is not totally explained (about 0.3 K below the phase front appearance, no domains are easily observed but  $\varepsilon' - \varepsilon'_{mono}$  increases as sharply as in a FP case, and even more in certain cases). As the  $\varepsilon'_F(T_C)$  value in a monodomain region is lower than  $\varepsilon'_P(T_C)$  (the usual discontinuity at the

transition), the  $\varepsilon'(T)$  curves in PF cycles are not compatible with a classical static model as presented:  $\varepsilon' - \varepsilon'_{mono}$  could remain equal to zero until the domain texture's appearance. Two explanations can be proposed. Firstly, there may be domain textures with such a small domain width that it is not possible to observe them by optical means. The first experiments of optical diffraction performed with a helium laser beam propagating along the *c* axis are not sufficient and other techniques must be studied such as  $\gamma$ -ray diffractometry (Bastie and Bornarel 1979) or synchrotron radiation. Secondly, the thermal gradient *G* suffices to play the role of an external field leading the new ferroelectric region to be monodomain. In this case, local measurements of birefringence could also provide information.

In conclusion, the effect of a thermal gradient G on the transition is illustrated. When G is parallel to c, the phase front is approximately perpendicular to c. This is a simple case, which demonstrates the importance of mechanical energy. However to acquire a better knowledge of the phase front, it is necessary to perform experiments in two directions: with different orientations of G relative to the c axis, and with decreasing values of G. These studies are in progress in our laboratory. The present paper confirms that the phase front itself does not have a strong energy per unit area because of its smooth shape in strained sample regions. Finally, the loss constant allows us to detect phase front modifications and domain arrangments. The contribution of domains to the dielectric constant is well confirmed. To apprehend an eventual contribution of the phase front itself towards dielectric properties, it is necessary to change the phase front area during the phase coexistence and as already noted, to change the G vector.

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