Dielectric dispersion due to weak domain wall pinning in RbH₂PO₄

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Dielectric spectroscopy experiments are carried out in the ferroelectric phase of rubidium dihydrogen phosphate (RbH₂PO₄), within the frequency range 0.1 Hz $\leq f \leq 1$ MHz. A strong dielectric time decay (ageing) dominates after thermal equilibration. However, spectra taken after long dwelling times reveal a significant dispersion below the fundamental piezoelectric resonance frequency *f res* . The permittivity of the well-aged sample is found to decrease linearly with the logarithm of the frequency. Not detectable above the paraelectricferroelectric phase transition temperature T_c , the dispersion is observed both in the range T_c > $T>T_f$ \approx 117 K of the anomalously high domain wall contribution and in the low-temperature range $T < T_f$ corresponding to the frozen-in domain-wall response. The result indicates that the concept of weak pinning in the context of the theory of elastic interfaces in disordered media may be applicable to describe the interaction between the ferroelectric/ferroelastic domain walls in RbH₂PO₄ and randomly distributed impurities.

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I. INTRODUCTION

Rubidium dihydrogen phosphate (RbH_2PO_4) is a wellknown representative of $KH_{2}PO_{4}$ -type hydrogen-bonded ferroelectrics, whose dielectric properties attracted considerable interest due to the extremely large static permittivity values observed in the polar phase. A common feature of $KH₂PO₄$ -type single crystals is the peculiar dielectric anomaly accompanying the paraelectric-ferroelectric $(tetragonal-orthorombic)$ phase transition.¹ Upon cooling below the phase-transition temperature T_c =146 K, the permittivity parallel to the ferroelectric *c* axis remains anomalously high but decreases steeply below the temperature T_f \approx 117 K, limiting the so called "plateau range" $T_c > T$ $>T_f$.² This anomaly was interpreted³ in terms of dielectric contributions of dielectric domain walls, an activity which freezes out below T_f .^{4,5}

There have been repeated reports about Debye-like dispersion in the ferroelectric phase of KH_2PO_4 below the frequency f_{res} of the fundamental piezoelectric resonance.^{6–10} It is now well established that this low-frequency relaxation, representing a composite dispersion which includes at least three relaxational modes, is related to domain-wall motion. $6,7$ Thus the dispersion may be considered to be a ''fingerprint'' of the physical mechanism underlying the dielectric domainwall contribution. In view of the similar anomaly $\varepsilon'(T)$ observed in KH_2PO_4 and RbH_2PO_4 , it should be expected that the high domain-wall contribution is based on the same mechanism leading to a quite similar low-frequency dispersion in both crystals. However, dielectric data covering the low-frequency range of RbH_2PO_4 are not numerous. Though temperature scans $\varepsilon(T)$ show in the ferroelectric phase frequency dependence of the permittivity, 6 there is no information available about the type of the low-frequency dispersion and its relationship to the motion of ferroelectric domain walls in RbH_2PO_4 .

In this paper we present the results of a dielectric study of the low-frequency dielectric spectrum $(0.1 \text{ Hz} < f)$ \leq 1 MHz) within the ferroelectric phase of RbH₂PO₄. Here the low-frequency dielectric properties depend strongly on the time elapsed after thermal equilibration. To obtain information about the dispersion related to the metastable domain structure of RbH_2PO_4 , we carried out dielectric measurements after long dwelling times during which the major part of the time-dependent processes faded away. The results obtained on a well-aged sample provide clear evidence of a relaxational non-Debye dispersion in the ferroelectric phase of RbH_2PO_4 , which is characterized by a permittivity linearly decreasing with the logarithm of frequency. This dispersion type was recently detected in KH_2PO_4 ,¹⁰ where it is, however, almost hidden by the Debye-like constituents within the spectrum. Our observations indicate that the dynamics of ferroelectric domain walls both in RbH_2PO_4 and in $KH_{2}PO_{4}$, are creeplike, and comply with theoretical predictions for elastic interfaces in disordered media.¹¹

II. EXPERIMENT

The RbH_2PO_4 crystal was grown by slow evaporation from an aqueous solution at $T=315$ K. X-ray topography and high-resolution x-ray diffraction were used to characterize the crystal quality. From x-ray topography (Lang method) we conclude that the dislocation-free areas of the crystal are large (the typical size is 1 cm in diameter), and observe characteristic fringes of the Bohrmann-effect (dynamic diffraction). The high-resolution x-ray-diffraction studies were carried out using $Cu_{K\alpha1}$ radiation from a fourfold Ge(222) monochromator $(\delta\lambda/\lambda \approx 5 \times 10^{-5})$. With a channel-cut $Ge(222)$ analyzer, we obtained a full width half maximum of 6.8" of the 4 0 0 reflection (rocking curve: ω scan) at room temperature. This value is in close agreement with the resolution limit of the instrument as we estimated it studying a perfect Si wafer. Thus we have established the high crystallographic quality of the RbH_2PO_4 crystal and can exclude that clustering of point defects is relevant, which would lead to a significantly higher dislocation density.

From the crystal, *c* plates $(4.0 \times 3.5 \times 2.2 \text{ mm}^3)$ were cut, and aluminum electrodes were evaporated onto the large faces. The sample was mounted into a liquid-nitrogen cryostate. Electrical contact was established by thin wires, so that the sample could vibrate freely during the dielectric measurements. The complex permittivity $\varepsilon^* = \varepsilon' - i\varepsilon''$ along the ferroelectric *c* axis was measured in the low-frequency range 0.1 Hz<f<100 kHz, with a computer-controlled dielectric spectrometer. The current through the sample, subjected to harmonic electric excitation, was amplified with a current amplifier whose output voltage was analyzed using a digital lock-in amplifier. Each time the measuring frequency or the gain and filter settings of the current amplifier were changed, a calibration measurement was carried out in order to compensate for its frequency and gain-dependent input impedance and the parasitic feedback capacity, respectively. Using a computer-controlled switch, the current amplifier input was connected instead of the sample with up to two out of eight low-loss reference capacitors $(2 \text{ pF} \leq C_{ref}^{(i)})$ ≤ 10 nF), so that the sample capacity was reproduced as well as possible. The transmission function of the amplifier was modeled by software in order to minimize the residual error. Switching the sample to the input terminal of a HP4192A impedance analyzer, the high-frequency part $(100 \text{ kHz} < f < 1 \text{ MHz})$ of the spectra was investigated in a typical four-wire configuration. The measurements were carried out with an amplitude $E_{ac} = 5$ V/m of the electric ac field. For the crystal examined, and within the experimental accuracy, the permittivity measured at this field level does not depend on E_{ac} within the whole temperature range investigated. The sample was cooled below the phase transition temperature T_c , with a constant cooling rate $\Delta T/T$ $=$ 3.3 K/h. The cooling run was interrupted several times to carry out spectroscopic investigations. After thermal equilibration, the dielectric spectra were recorded continuously at constant temperature. The temperature was stabilized during the dwelling time, with an accuracy $\Delta T \le 0.01$ K.

III. RESULTS

Approaching the phase-transition temperature upon cooling from above, T_c was determined from the dielectric data as the temperature for which deviations from the Curie-Weiss behavior of the low-frequency permittivity set in.^{12,13} If the cooling run was interrupted in the paraelectric phase, no significant time dependence $\varepsilon'(t)$ was detected at constant temperature. However, a strong time decay $\varepsilon'(t)$ was observed in the ferroelectric phase at frequencies below the piezoelectric resonance frequency $f_{res} \approx 50$ kHz. On the other hand, the rather slight temperature instability during the dwelling time caused large permittivity fluctuations due to the dielectric deaging of the sample, so that a high level of temperature stability turned out to be a condition for spectroscopic experiments.

We will show now that, due to the pronounced aging effect, unusually long dwelling times are required to obtain information about the dielectric dispersion in RbH_2PO_4 . First we present spectra $\varepsilon'(f)$ [Fig. 1(a)] taken at the temperature $T=144.7$ K, i.e., within the plateau range of RbH₂PO₄.

FIG. 1. Dielectric spectra obtained at $T=144.7$ K for the real (a) and imaginary (b) parts of the complex permittivity. Results for various dwelling times t_d are provided.

Data obtained for different dwelling times t_d are shown to demonstrate the significance of the time decrease $\varepsilon'(t)$. Since the measurements were carried out for decreasing frequency, the spectra measured immediately after thermal equilibration show even an apparent increase $\varepsilon'(f)$ due to the dielectric decay during the time $t_m \approx 130$ min, necessary to record the spectrum.

Even for the longest dwelling times t_d =59 h examined, the dielectric decay is still evident (Fig. 2). However, spectra $\varepsilon'(f)$, obtained for long dwelling times $t_d > 20$ h, are *decreasing* functions of frequency. The permittivity decreases monotonically with time, and spectra were taken for decreasing frequency, so that we can exclude the aging effect as the cause of the frequency dependence, and thus obtain evidence of a small but distinct low-frequency dispersion in the ferroelectric phase of RbH_2PO_4 . To conclude as to the character of the dielectric dispersion, we first eliminate the influence of aging. In the limit $t_d \geq t_m$, the dielectric decay during the measuring time t_m is approximately linear. In addition, the strength of the time decay $\varepsilon'(t)$ is frequency independent within our frequency window, as it can be seen from Fig. 2.

FIG. 2. Logarithmic frequency dependency $\varepsilon'(f)$ observed at $T=144.7$ K after long dwelling times $t_d > 50$ h.

Hence it is easy to eliminate the aging effect on the dielectric spectra, taking into account the time decay $\delta \varepsilon'$ during t_m . Using the relationship

$$
\varepsilon'(f,t_0) = \varepsilon'(f,t) + \delta \varepsilon'(t-t_0)/t_m, \qquad (1)
$$

data $\varepsilon'(f, t_0)$ were obtained which refer to the starting time $t₀$ of the particular spectrum. Clearly, the aging effect does not distort the spectra for long dwelling times. We thus conclude that the dispersion in the ferroelectric phase of RbH_2PO_4 is characterized by a linearly decreasing permittivity on a logarithmic frequency scale, qualitatively similar to what is shown in Fig. 2 for the uncorrected data. In what follows we refer to this type of dispersion as logarithmic dispersion.

The frequency-dependent imaginary part $\varepsilon''(f)$ is shown in Fig. $1(b)$. The high-frequency branch of the dielectric loss spectrum is dominated by contributions from the resonance dispersion, whose resonance frequency increases with time. This increase is related to elastic domain-wall contributions to the compliance $1/c_{66}^E$ which, quite similar to the dielectric domain-wall response, decrease with time. At the lowfrequency end of the spectrum, an increase of ε'' with decreasing frequency is observed, which flattens out for longer dwelling time. We cannot exclude that the low-frequency dispersion $\varepsilon''(f)$ is related to the conductivity. However, the time dependence $\varepsilon''(t)$ points to a Debye-like dispersion region centered outside the observation window, whose relaxor strengths and/or relaxation frequency decrease with time. Within the central part of the loss spectrum, the contribution of the relaxational and resonance-type dispersions localized outside the observation window can be neglected. Nevertheless here we observe a nonzero, nearly frequencyindependent, dielectric loss which decreases with time.

We now analyze the temperature evolution of the dielectric spectra which is shown in Fig. 3 for the data corrected according to Eq. (1) . No significant low-frequency dispersion is discernible in the paraelectric phase close above T_c . Both logarithmic dispersion and dielectric aging set in upon cooling immediately below the phase transition temperature T_c .

FIG. 3. Dielectric dispersion close above T_c and at different temperatures within the plateau range of the ferroelectric phase: circles, $T=148.7$ K, $t_d=2$ h, and $\varepsilon'(0.1$ Hz)=1294; triangles, $T=145.7$ K, $t_d=44$ h, $\varepsilon'(0.1 \text{ Hz})=10745$; crosses, *T* $= 144.7$ K, $t_d = 59$ h, and $\varepsilon'(0.1 \text{ Hz}) = 4833$; squares, *T* $= 130.7$ K, $t_d = 82$ h, and $\varepsilon'(0.1$ Hz) $= 2254$. The data are normalized to the value $\varepsilon'(0.1 \text{ Hz})$, and corrected for aging (see the text).

At $T=145.7$ K, the strength $\delta \varepsilon'$ of the time relaxation is smaller and the strengths *s* of the logarithmic dispersion is larger as compared to the data measured at $T=144.7$ K, analyzed above. Therefore, in a narrow temperature range close below T_c , the logarithmic dispersion type is discernible even from the uncorrected data, taken immediately after temperature stabilization. On the other hand, the dispersion strength *s* is decreased upon further cooling to $T=130.7$ K and the average time constant of the time relaxation $\varepsilon'(t)$ becomes considerably larger. Although we are still within the plateau range, we did not obtain a saturation of the logarithmic dispersion strengths even for the longest dwelling time examined: t_d =82 h.

The freezing temperature $T_f \approx 117$ K of our sample was estimated from the temperature dependent data $\varepsilon'(T)$. This differs from the value $T_f \approx 100$ K derived from the result of a previous study, 6 which may be related to the different defect content¹⁴ or thickness¹⁵ of the specimens examined. Within the low-temperature range $T < T_f$, the timedependent relaxation is considerably smaller than the dispersion of the permittivity. The dielectric spectrum taken at *T* $=102.7$ K $<$ *T_f* (Fig. 4) applies to this range. It characterizes the dispersion in the temperature range of the ''frozen-in'' domain-wall response. As can be seen from the lowfrequency part of the spectrum, the logarithmic dispersion type is basically retained at low temperatures. Additionally, an indication for another polarization mechanism is obtained within the frozen-in state, leading to a relaxational-type dispersion with a wide distribution of relaxation times centered around the average time $\langle \tau \rangle \approx 5 \times 10^{-5}$ s. Since the Debyelike domain wall relaxation in KH_2PO_4 slows down if T_f is approached upon cooling,^{6,14} it may be assumed that similar Debye-like constituents within the spectrum of RbH_2PO_4 are hidden at elevated temperatures by the resonance-type dispersion.

FIG. 4. Dielectric dispersion at $T=102.7$ K taken below the freezing temperature T_f .

IV. DISCUSSION

The plateaulike temperature dependence of the anomalously high low-frequency permittivity ε' in the polar phase of RbH_2PO_4 represents a characteristic feature of $KH₂PO₄$ -type polydomain crystals.¹ In the plateau range of KH_2PO_4 , it is now confirmed^{6,7} that the observed lowfrequency dielectric dispersion⁸ results from domain-wall activity. The polarization mechanism related to the lowfrequency dispersion in RbH_2PO_4 is also clearly restricted to the polar phase, for we observe no dispersion above T_c . However, the domain-wall response in $KH_{2}PO_{4}$ was reported to be Debye-like, 8 whereas in the ferroelectric phase of RbH_2PO_4 we observe a logarithmic dispersion type. Thus the question arises of why the dielectric response of domain walls in the ferroelectric phase of both crystals manifests itself in quite similar temperature dependences but rather different dispersions of the low-frequency permittivity.

In a previous study, 10 we investigated the low-frequency dielectric spectrum within the plateau range of a purified KH_2PO_4 crystal. We detected a strongly non-Debye relaxational mechanism D_{ND} which is almost hidden by Debyelike constituents within the spectrum. The dielectric contribution ε'_{ND} due to D_{ND} is quite similar to the logarithmic dispersion we observe for a well-aged RbH_2PO_4 sample. In KH_2PO_4 , we interpreted D_{ND} to reflect the creeplike dynamics of domain walls predicted by the theory of elastic interfaces in disordered media.^{16,11} The theory treats local distortions of the elastic interfaces in an attempt to take advantage of concentration fluctuations of randomly distributed immobile impurities. Due to the competition between elasticity, disorder and thermal fluctuations, the smallest length L_c is predicted at which the interface can be weakly pinned, i.e., pinned on impurity fluctuations. On length scales $L \leq L_c$, the interface is either free or strongly pinned on isolated impurities. The free interface can respond to the external electric field on a length scale L_c thus providing an interface contribution $\chi_{DW} \sim L_c^2$ to the susceptibility.

The weakly pinned interface represents a system with a

large number of metastable states corresponding to local minima of its potential energy. Fluctuations on length scales *L* between different interface configurations require thermally activated jumps over energy barriers $E_b(L)$, which scale as $L^{2\zeta}$. For the interface dimension $d=2$, the pinning theory predicts roughness exponents $\zeta = 2/3$ and $\zeta \approx 2/5$ for random-field-type and random-bond-type impurities, respectively. In a dielectric experiment, the external electric field mediates fluctuations between different domain-wall configurations. Since the relevant energy barrier $E_c = E_b(L_c)$ for weak pinning at short times increases after the time t_c $t_0 = t_0 \exp(E_c/kT)$, the length scale L_c on which the interface is free to respond to the external field increases as $[\ln(t)]^{2\zeta}$ for $t > t_c$. The increase of L_c leads to a logarithmic dispersion of the interface susceptibility at low frequencies, which obeys the relationship 11

$$
\chi_{DW}(\omega) \sim \frac{L_\epsilon^2}{T_\epsilon} \left(1 + \frac{T}{T_\epsilon} \right)^{2/\phi} \left[1 + \frac{T}{T + T_\epsilon} \ln(1/\omega \tau_0) \right]^{1/\zeta}, \tag{2}
$$

where ϕ is the crossover exponent (random field ϕ =4/3, random bond ϕ =4/5), and T_e corresponds to the the smallest potential barrier at which the interface can be weakly pinned at zero temperature, where it is free on the length scale $L_{\epsilon} \sim T_{\epsilon}$.

From the spectra obtained after long dwelling times, we conclude that the logarithmic dispersion in the ferroelectric phase of RbH_2PO_4 reflects the response of weakly pinned domain walls, just as the non-Debye dispersion in KH_2PO_4 . Accordingly, the domain-wall dynamics in both crystals is similar, i.e., creeplike. From this point of view, both crystals should be classified into a group of ferroelectrics in which a dielectric non-Debye dispersion arises due to a pinning of the domain walls, $17,18$ or of an incommensurate modulation¹⁹ in the ferroelectric and incommensurate phases, respectively. This stresses the significance both of the elastic domain wall stiffness and the impurity content for the dielectric properties of RbH_2PO_4 .

Our results show that the domain-wall responses in both KH_2PO_4 and in RbH_2PO_4 depend on the time elapsed after thermal equilibration. However, the aging in RbH_2PO_4 is much more pronounced, so that rather long dwelling times are required to obtain information about the spectrum. Hence we are restricted to the aged sample for which, within the central part of the loss spectrum [Fig. 1(b)], ε'' is nearly frequency independent. Here the relationship²⁰

$$
\varepsilon'' = -(\pi/2)[\partial \varepsilon'/\partial \ln(\omega)],\tag{3}
$$

valid for systems with a broad distribution of relaxation times, is nicely fulfilled. Therefore, the loss in this frequency range has to be attributed to the relaxational mechanism causing the logarithmic dispersion $\varepsilon'(f)$. Within the whole temperature range examined, we observe a time decay $\varepsilon''(t)$. However, our dwelling time was large enough to observe a saturation at temperatures close below T_c . Accordingly, data obtained for $T_c - T = 0.3$ and 1.3 K (Fig. 3) reflect the saturated value $s(t\rightarrow\infty)$. Assuming that Eq. (3) is also fulfilled for shorter times, the decay $\varepsilon''(t)$, observed immediately after thermal equilibration, would reflect the time decay of the logarithmic dispersion strength $s(t) = \partial \varepsilon'(t)/\partial \ln(\omega)$. In this case we can conclude from our data that the dispersion strength *s* of the unaged domain structure is more than one order of magnitude larger.

As the elasticity of the domain wall generally depends on temperature, 21 it should be expected that the interface rearranges itself after any temperature change with the defect environment. The system relaxes at constant temperature toward a more stable configuration which requires jumps over energy barriers separating metastable wall configurations. The dielectric aging we observe in RbH_2PO_4 may thus reflect the time decrease of the nonequilibrium length scale $L_c^{neq}(t)$, on which the domain wall is weakly pinned on impurity fluctuations.

We do not obtain direct evidence of Debye dispersion within the plateau range of RbH_2PO_4 which dominates the spectrum within the plateau range of $KH_{2}PO_{4}$. However, the low-frequency part of the loss spectrum [Fig. 1(b)] and the polydispersive Debye-like relaxational dispersion which superimposes the logarithmic dispersion type in the frozen in state $(Fig. 4)$ indicate Debye-like constituents within the plateau range of RbH_2PO_4 outside the frequency window of our spectrometer. In KH_2PO_4 , the Debye dispersion becomes polydispersive, and smears out upon cooling below the freezing temperature T_f .²² The logarithmic dispersion type, however, remains qualitatively unchanged in the frozen-in state. This observation complies with the concept of weak domainwall pinning, for which we assume no qualitative changes of the dispersion type at low temperatures. However, it is predicted that the stiffness Γ of the ferroelectric domain wall generally decreases with temperature. For a second-order phase transition, theory provides a critical behavior $\Gamma \sim (T_c)$ $(-T)^{3/2}$ of the defect-free crystal.²¹ Therefore, the temperature dependence of the parameter $T_e \sim \Gamma$ should be taken into account if the pinning theory is to be applied to a ferroelectric system. The decreasing logarithmic dispersion strengths we observe in RbH_2PO_4 upon cooling (Figs. 3 and 4) may hence be understood in terms of Eq. (2) as a consequence of the decreasing ratio T/T_e .

Although it turned out that the domain wall responses in RbH_2PO_4 and KH_2PO_4 are qualitatively the same, it seems to be worthwhile to discuss the quantitative differences. From a crystallographic point of view, both crystals are nearly perfect, showing a rather low dislocation density. However, the KH_2PO_4 crystal, examined previously¹⁰ was additionally subjected to several recrystallization steps to reduce the impurity concentration.²³ The permittivity maximum at T_c is well known to decrease considerably with impurity concentration.²⁴ The high degree of purification of our KH2PO4 crystal hence manifests itself in a rather large value $(\varepsilon_{max} \approx 8 \times 10^4)$ which exceeds most of the literature data available for KH_2PO_4 . In addition, the typical value for the "threshold" ac field, $E_{ac}^{\star} \approx 10$ V/m above which the ac-field dependence of the permittivity of our $KH_{2}PO_{4}$ crystal becomes detectable, is almost two orders of magnitude smaller than values $E_{ac}^{\star} \approx 500$ V/m reported previously.¹⁴ On the other hand, the maximum permittivity value $\varepsilon_{max} \approx 10^4$ of our RbH_2PO_4 crystal is quite comparable to previous results.¹² In the plateau range of RbH_2PO_4 , we observe a significantly larger threshold $E_{ac}^{\star} \approx 150$ V/m, except in a narrow temperature interval close below T_c where the nonlinearity is considerably larger in both crystals $(RbH₂PO₄)$: $E_{ac}^{\star} \approx 4$ V/m; KDP: $E_{ac}^{\star} \approx 2$ V/m). Taking into account the differences between both crystals with respect to the level of nonlinearity and the height of the dielectric anomaly, our RbH_2PO_4 crystal should be classified, according to the pointdefect concentration, as a nominally pure system in contrast to the purified KH_2PO_4 crystal studied previously.¹⁰

The different level of impurity concentration is also visible in the logarithmic dispersion strength, which is considerably weaker in our RbH_2PO_4 crystal. Considering the ratio $R(T_c-T) = s^{(RDP)}/s^{(KDP)}$ within the plateau range of the aged crystals, we estimate, depending on temperature difference T_c –*T*, values $0.02 < R < 0.1$. The purification effect can be qualitatively understood in terms of the pinning theory, $¹¹$ </sup> from which the relationship $\chi_{DW} \sim 1/n_D$ between the interface susceptibility and impurity concentration can be derived. This result may be generalized to the domain-wall response of other ferroelectric crystals, insofar as in nominally pure crystals, on which the emphasis of the previous experimental work was apparently placed, rather small *s*-coefficients should be expected. In addition, the strength of the time-dependent dielectric relaxation in our RbH_2PO_4 crystal is, as compared to the absolute permittivity value and to the dispersion strength, significantly larger than in the KH_2PO_4 of the previous study.¹⁰ Though further experiments are underway to clarify the influence of purification on the aging phenomena, it may be assumed that generally rather long dwelling times, high-temperature stability, and highresolution measurements are required to detect the logarithmic domain-wall dispersion in nominally pure ferroelectric crystals.

To conclude, our dielectric experiments carried out on a well-aged sample revealed a non-Debye logarithmic dispersion in the ferroelectric phase of RbH_2PO_4 . This indicates that the concept of weak pinning in the framework of the theory of elastic interfaces in disordered media, $16,11$ applies to the interaction of the ferroelectric domain walls in RbH_2PO_4 with immobile impurities. It turned out that the dielectric domain-wall responses in RbH_2PO_4 and KH_2PO_4 $(Ref. 10)$ are essentially the same, and comparable to systems such as lead zirconate titanate ceramics 18 and $Sr_{1-x}Ca_{x}TiO_{3}$ single crystals,¹⁷ where the logarithmic type of the low-frequency dispersion is obvious. On the one hand, our results emphasize the significance of the temperature dependent stiffness of the domain walls in RbH_2PO_4 for the temperature dependence of dielectric properties. On the other hand, the crucial influence of the impurity content on the dielectric response of the domain wall becomes obvious, illustrating the experimental difficulties in proving the logarithmic dispersion type in nominally pure systems.

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