External field effect on the anisotropy of dielectric permeability of KH_2PO_4 and $NaKC_4H_4O_6 \cdot 4H_2O$ crystals under high pressure

A.G.Slivka, V.M.Kedyulich, H.M.Guyvan

Uzhhorod National University, Department of Optics, 54 Voloshyn Str., 88000, Uzhhorod, Ukraine

Experimental studies of the effect of temperature, external pressure and electric field on the anisotropy of dielectric permeability of KH_2PO_4 and $NaKC_4H_4O_6 \cdot 4H_2O$ crystals are performed. The experimental data on the effect of electric field and pressure on the dielectric permeability of Rochelle salt are analyzed in the framework of phenomenological theory with the account of the vicinity of the phase transitions to the double critical point. Coefficients of thermodynamic potential expansion over the order parameter are determined as well as its pressure behaviour is described. The hydrostatic pressure increase is shown to shift the phase transitions in Rochelle salt away from the double critical point.

1. Introduction

Long time has passed since the discovery of ferroelectric activity in KH_2PO_4 (KDP) and $NaKC_4H_4O_6 \cdot 4H_2O$ (Rochelle salt) crystals. However, these materials still attract keen attention of researchers due to their specific physical properties related to a system of structural hydrogen bonds and their changes at phase transformations. Another common feature of these crystals consists in their piezoelectric properties revealed in the paraelectric phase.

Experimental studies of the anisotropy of physical properties and its variation under external pressure and electric field play an important role in understanding the microscopic mechanisms of phase transitions in ferroelectric crystals with hydrogen bonds. This is due to the fact that by applying external pressure once can continuously vary geometrical parameters of hydrogen bonds and violate their equivalence, which provides a unique possibility to study the role of hydrogen bonds in the mechanisms of structural phase transformations and dielectric response of crystals. Applying uniaxial strain as well as electric fields enables one to investigate the role of piezoelectric interactions in the phase transitions and in the formation of physical characteristics of these crystals.

In the 1990s the keypoint for further investigations of the KDP family crystals was related to theoretical studies by Stasyuk and Biletsky [1,2] which gave an impetus to another series of theoretical studies of these crystals and subsequently of other ferroelectric materials with hydrogen bonds including Rochelle salt crystals. The effective Hamiltonian for KDP crystals was obtained *ab initio* with the account of piezoelectric interaction within the framework of a modified model of proton ordering. This enabled a proper description of a number of available physical characteristics of crystals of this group as well as made it possible to predict some physical effects due to external fields which had not been experimentally studied before. Similarly, Stasyuk and Levitskii [3–5] have taken into account the piezoelectric interaction within the framework of a modified Mitsui model for theoretical description of experimental data for Rochelle salt. The results of these theoretical studies required experimental investigations of a number of key issues which led to more active experimental studies.

Here we report on the results of experimental studies of the effect of external pressure and electric field on the anisotropy of dielectric properties of KDP and Rochelle salt crystals. These results were obtained in a series of studies starting from 1999, having been initiated by the above mentioned theoretical papers.

2. Anisotropy of dielectric permeability of KDP and Rochelle salt crystals

Figure 1 shows the temperature dependences of longitudinal dielectric permeability of KDP crystals at atmospheric pressure and at the measuring field frequency of 1 kHz (curve 1) and 1 MHz (curve 2). No dispersion of transverse dielectric permeability has been revealed at these frequencies (curve 3). At the frequency of 1 kHz the dependence of longitudinal dielectric permeability is characterized by two anomalies: a maximum of $\varepsilon_c(T)$ at the phase transition temperature $T_C = 122$ K and a shoulder in the ferroelectric phase in the temperature range from 91 to 122 K, related to the domain wall dynamics [6]. At the measuring field frequency of 1 MHz a number of extrema is observed in the $\varepsilon_c(T)$ plot in the vicinity of the phase transition point (curve 2). This feature is related to the piezoelectric resonance in KDP crystal [7]. It cannot be related to the trend of the order parameter relaxation time in an order/disorder ferroelectric to the infinity, since in this case a single minimum in the temperature dependence of dielectric permeability at the phase transition temperature should be observed.
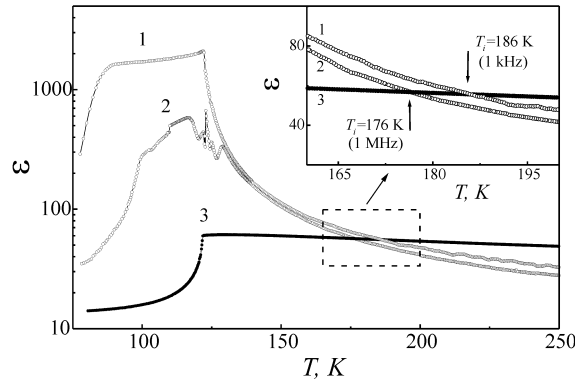

Figure 1. Temperature dependences of longitudinal dielectric permeability of KDP crystal at the frequency of 1 kHz (curve 1) and 1 MHz (curve 2) and transverse dielectric permeability (curve 3).

The temperature behaviour of dielectric permeability of Rochelle salt in different crystallographic directions at different measuring field frequencies ($f = 1$ kHz and $f = 1$ MHz) is shown in figures 2 and 3. The anomalous behaviour of $\varepsilon(T)$ in the vicinity of the phase transitions at $f = 1$ kHz is observed only along the a direction. Two characteristic maxima are observed at the phase transition temperatures $-18°$C and $+24°$C (figure 2) in the temperature dependence $\varepsilon(T)$ along the polar axis. The dielectric permeability values at the phase transition temperatures are different: 2200 and 4300, respectively. Along the c and b directions ($f = 1$ kHz) the dielectric permeability values are practically the same in the whole temperature range under investigation, except the high-temperature phase ($T > 25°$C). The temperature dependences of ε_b and ε_c reveal a monotonous increase of the dielectric permeability in the ferroelectric phase and in the high-temperature paraelectric phase. Since practically no anomalies are observed at the phase transition temperatures, this indicates that the main changes in the crystal structure, resulting in the appearance of spontaneous polarization, occur along the a direction. At the temperatures $T > 25°$C, the features in the temperature dependences of ε_b and ε_c are revealed, evidently related to the crystal lattice destruction and, as a consequence, an increase of the crystal ionic conductivity. These processes are irreversible and at temperatures above $50°$C they result in a loss of the Rochelle salt ferroelectric properties.
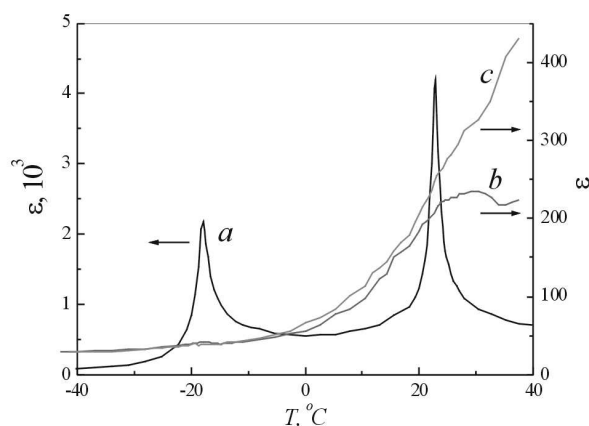
Figure 2. Temperature dependences of the dielectric permeability of Rochelle salt crystals for different crystallographic directions at the measuring field frequency $f = 1$ kHz.

In the temperature dependence of ε_a at the measuring field frequency $f = 1$ MHz a number of extrema is observed in the vicinity of the phase transition points (figure 3) which is related to the piezoelectric resonance, since the measuring field frequency is close to the resonance frequency of the crystal. The study of anisotropy of the crystal dielectric permeability consists in determining the orientation of the main ellipsoid axes along the crystallographic directions. It is also important to determine the variation of these parameters under temperature and external pressure. The orientational dependence of dielectric permeability which is a tensor in anisotropic crystals, is given by $\varepsilon = \varepsilon_{ij} l_i l_j$, where ε_{ij} are the dielectric permeability tensor components, l_i and l_j are the cosines of the angles between the direction, in which the dielectric permeability is being determined, and the Cartesian coordinate axes, in which the tensor is written.
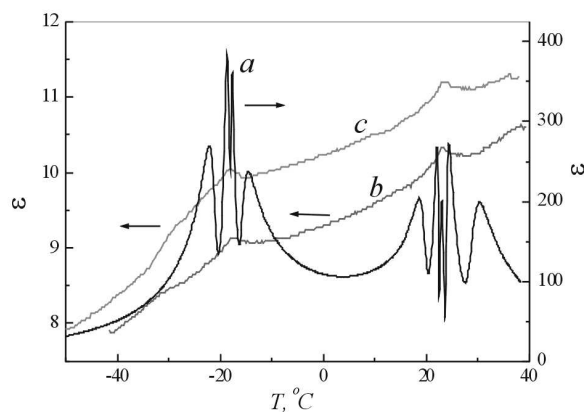

Figure 3. Temperature dependences of dielectric permeability of Rochelle salt crystals for different crystallographic directions at the measuring field frequency $f = 1$ MHz.

In the KDP crystal at room temperature the transverse dielectric permeability value is higher than the longitudinal one (see figure 1). The temperature decrease results in an increase of both values; however, the longitudinal dielectric permeability undergoes essential anomalous changes, its value sharply increasing with the approach to the phase transition point. Therefore, at the temperature $T_i = 186$ K ($f = 1$ kHz) the longitudinal and transverse dielectric permeability values become equal. The temperature of the isotropic point at the measuring field frequency of 1 MHz is $T_i = 176$ K (see the insert to figure 1). At the temperatures $T < T_i$ the longitudinal dielectric permeability value exceeds the transverse one.

Sections of the indicating surface of the dielectric permeability tensor for the KDP crystal at atmospheric pressure and different temperatures are shown in figure 4. The results of angular dependence of dielectric permeability of KDP crystals in the (010) plane show that the main axes of the ε_{ij} tensor clearly coincide with the crystallographic axes of the crystal. At room temperature, the semi-major axis is directed along the a direction, while the semi-minor axis is along the c direction. The decrease of temperature at first results in the decrease of anisotropy of the crystal dielectric permeability. At $T_i = 186$ K the section of the indicating surface is a circle, which means that dielectric permeability ε is isotropic at this temperature. A further decrease of temperature again results in the anisotropy of dielectric permeability, its value being maximal at ferroelectric phase transition temperature. In this case the semi-major axis is oriented along the c direction while the semi-minor one – along the a direction. In the ferroelectric phase ($T < T_C$) the values of dielectric permeability along the c direction are by an order of magnitude higher than for the a direction. Thus, in KDP crystal at the temperature T_i an isotropic point is achieved, where the temperature inversion of the sign of dielectric permeability difference $\Delta\varepsilon = \varepsilon_a - \varepsilon_c$ is observed.
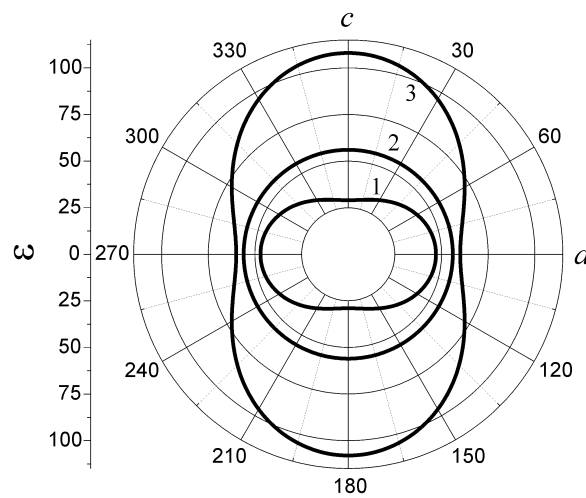

Figure 4. Temperature transformation of the section of dielectric permeability indicating surface by the ac plane of the KDP crystal, 1: $T = 275$ K, 2: $T = 186$ K, 3: $T = 150$ K.

We have also performed direct measurements of the ε value along different crystallographic directions for the $NaKC_4H_4O_6 \cdot 4H_2O$ crystal. The angular dependence of dielectric permeability of Rochelle salt in the (010) symmetry plane at atmospheric pressure and at different temperatures is shown in figure 5.

The obtained results show that the specific features of temperature transformation of sections of the indicating surface of dielectric permeability for KH_2PO_4 and $NaKC_4H_4O_6 \cdot 4H_2O$ crystals are caused by the anomalous temperature behaviour of dielectric permeability along the polar direction in the crystals. The orientation of the main axes is independent of temperature.

3. High-temperature anomaly of dielectric permeability in KDP

The temperature dependence of dielectric permeability ε_a of KDP crystal at atmospheric pressure at the measuring field frequency of 1 MHz is shown in figure 6. Three characteristic features can be seen in this dependence: a step of dielectric permeability at the phase transition temperature $T_C = 122$ K, a maximum in the $\varepsilon_a(T)$ dependence at the temperature $T_{max} = 130$ K, and an inflection point at $T_f = 172$ K. The existence of a maximum of dielectric permeability at $T > T_C$ was registered in $KMnF_3$ and $NH_4H_2AsO_4$ antiferroelectric crystals [8]. In our earlier papers [9–11] the first experimental studies of the effect of external hydrostatic pressure on the temperature dependence of transverse dielectric permeability ε_a for KDP crystal were described. The hydrostatic
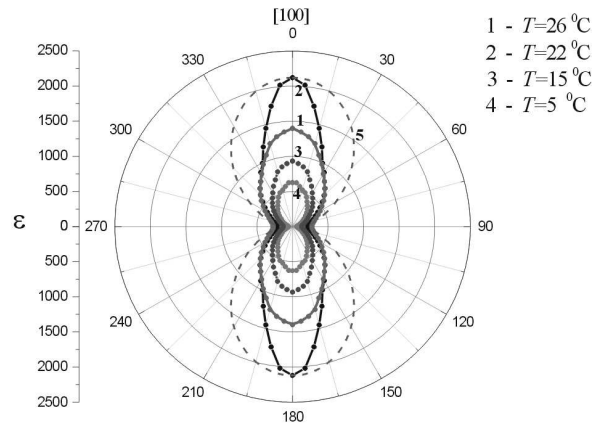
Figure 5. Temperature transformation of the section of the dielectric permeability indicating surface by the (010) plane of Rochelle salt crystal. Curve 5 shows the theoretical calculation data.

pressure increase to 6.5 kbar results in a shift of the $\varepsilon_a(T)$ curves towards lower temperatures, the T_C, $T_{\max}$, and T_f values varying linearly with pressure with the following coefficients: $\mathrm{d}T_C/\mathrm{d}p = -4.8 \cdot 10^{-2}$ K/MPa; $\mathrm{d}T_{\max}/\mathrm{d}p = -5.4 \cdot 10^{-2}$ K/MPa; $\mathrm{d}T_f/\mathrm{d}p = -5.4 \cdot 10^{-2}$ K/MPa. It was also shown that the temperature interval $T_{\max} - T_C$ becomes narrower with pressure.
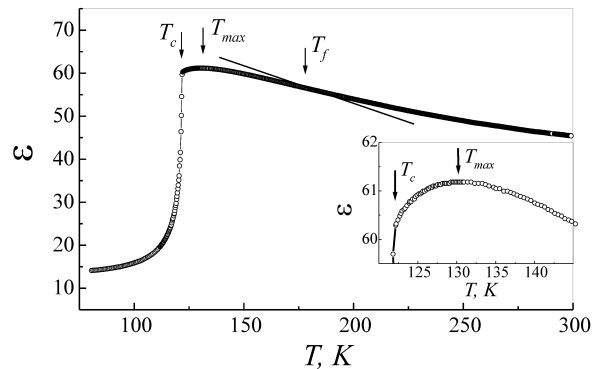

Figure 6. Specific features of the temperature dependence of transverse dielectric permeability for KDP crystal.

Figure 7 shows the temperature dependences of dielectric permeability of KH_2PO_4 crystal along the a side at the measuring field frequency $f = 1$ kHz (curve 1 – cooling, curve 2 – heating) and $f = 1$ MHz (curve 3 – heating). In the temperature range 250–300 K at $f = 1$ kHz an anomaly (a maximum) is observed, which is not revealed in the temperature dependence of dielectric permeability at $f = 1$ MHz, but is observed in the temperature dependence of dielectric loss angle tangent $\tan\delta(T)$. Temperature cycling (heating-cooling) leads to an increase of the temperature of maximum of dielectric permeability for the high-temperature anomaly (figure 8).

The change of the measuring field frequency results in considerable transformations of the high-temperature anomalies of transverse dielectric permeability in the paraelectric phase. While for KH_2PO_4 crystal at the measuring field frequency of 1 kHz the anomalies are observed both on $\varepsilon(T)$, and $\tan\delta(T)$ curves at the frequency of 1 MHz they are revealed only in the $\tan\delta(T)$ dependence. An increase of the hydrostatic pressure results in the suppression of high-temperature anomalies and their shift towards higher temperatures.

Low-frequency internal friction and shear modulus of KDP crystal in the paraelectric phase at
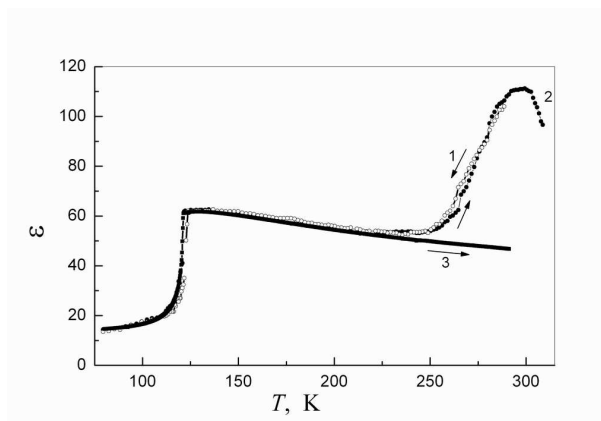
Figure 7. Temperature dependences of dielectric permeability of KDP crystal along the a side at the measuring field frequency $f = 1$ kHz (curves 1,2) and at the frequency $f = 1$ MHz (curve 3).
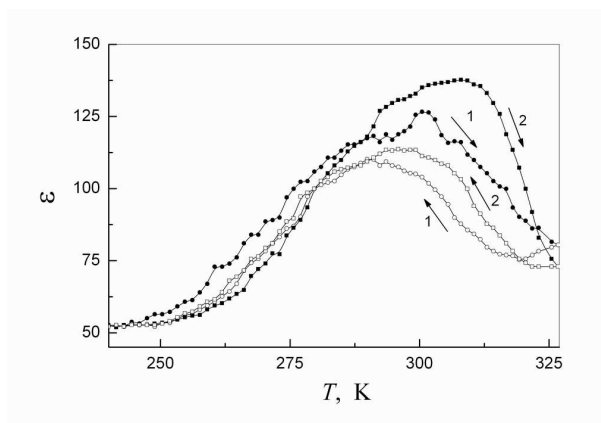

Figure 8. Temperature dependences of dielectric permeability of KDP crystal along the a side at the temperature cycling.

various external effects was studied in [12]. In the temperature range under investigation 80–325 K two distinct anomalies at the temperatures T_C and T^* are observed in the $Q^{-1}(T)$ and $G(T)$ dependences. Measurements of $Q^{-1}(T)$, $G(T)$, and $\varphi(T)$ (φ being the spontaneous torsion angle) were carried out at various rates of temperature, external mechanical strain, etc. The results of these studies have shown that the mechanism of the appearance of the high-temperature anomalies in the temperature interval 250–320 K is common for the KH_2PO_4 family crystals and practically does not depend on deuteration and isomorphic ion substitution which affect only the temperature position and magnitude of the anomalies revealed. The anomalies in high-temperature phase depend on the sample orientation, i. e. they are revealed not at an arbitrary orientation of the sample with respect to the crystallographic axes. Studies of temperature dependences of Q^{-1} [12] at different frequencies in the range from 6 Hz to 150 kHz have shown that the peak height at T^* varies inverse proportionally to the frequency, the shift of the Q^{-1} maximum along the temperature scale being insignificant. Such a behaviour of the internal friction maximum is typical of low-frequency relaxational processes related to phase transformations.

According to [13], in KDP-type crystals the role of impurities can be played by phosphate complexes (HPO_4^{-2}, $H_5P_2O_8^-$, H_3PO_4), present even in thoroughly purified solutions. Besides, the presence of a small concentration of uncontrollable impurities (Fe^{3+}, Cr^{3+}, Ca^{2+}, Al^{3+}) is assumed [14], responsible for local deformations which, in turn, lead to local symmetry distortion of the crystal matrix.

Since the volume fraction of inclusions is small (the average size of impurities is within 500–4500 Å and the total inclusion volume is from 1.5 to 5% in different crystals [13]), the dielectric permeability of such microheterogeneous system is determined mostly by the dielectric properties of the matrix, rather than the inclusions, which explains the absence of dielectric anomalies at the temperature T^* at certain experimental studies.

The data showing the anomalous behaviour of $\varepsilon(T)$ and $\tan\delta(T)$ dependences for KH_2PO_4 crystals obtained here agree with the results of low-frequency measurements of internal friction and shear modulus [12], related to a first-order phase transitions ($\bar{4}2m \longrightarrow mm2$), occuring in the inclusions of another phase whose formation is due to the uncontrollable defects.

4. Effect of external electric field and pressure on dielectric permeability of Rochelle salt

Temperature behaviour of dielectric permeability of Rochelle salt has been studied by many authors. The analysis of the available reference data shows a noticeable spread of the experimental values of dielectric permeability in different structural phases of Rochelle salt crystals [15]. Besides, variable character of the temperature dependence of inverse dielectric permeability in the paraelectric phase of these crystals is reported. These discrepancies, evidently, result from a different prehistory and quality of the samples as well as from different experimental conditions. In order to find possible reasons for such a situation, we have performed experimental studies of the effect of external factors (temperature, mechanical and electric fields, environment humidity) and experimental measurement conditions (temperature variation rate, preliminary thermal annealing, temperature mode of the measurements) on the dielectric properties of Rochelle salt crystals.

The value of ε and the character of its temperature dependence in the paraelectric phases most essentially depends on durable storage of Rochelle salt crystals in a humid air or on thermal annealing [17]. A crystal having not undergone thermal annealing, in the low-temperature phase is characterized by linear $\varepsilon^{-1}(T)$ dependence with the Curie-Weiss constant $C_W = 1.95 \cdot 10^3$ K. After preliminary thermal annealing the $\varepsilon^{-1}(T)$ dependence is nonlinear in both paraelectric phases and can be described by a single relationship $\varepsilon^{-1}(T) = -0.006 - 0.01t + 1.05t^2$ where $t = (T - T_m)/T_0$, $T_m = 275$ K, $T_0 = (T_{C2} - T_{C1})/2 = 276$ K which is in agreement with the results of [15] where the nonlinear dependence $\varepsilon^{-1}(T)$ in the paraelectric phases is explained within the framework of a phenomenological approach with the account of the vicinity of the phase transitions to the double critical point. Using the basic idea of [15], we have analyzed the obtained experimental data regarding the external electric field effect on the temperature dependences for Rochelle salt crystals. These results are illustrated in figures 9 and 10.
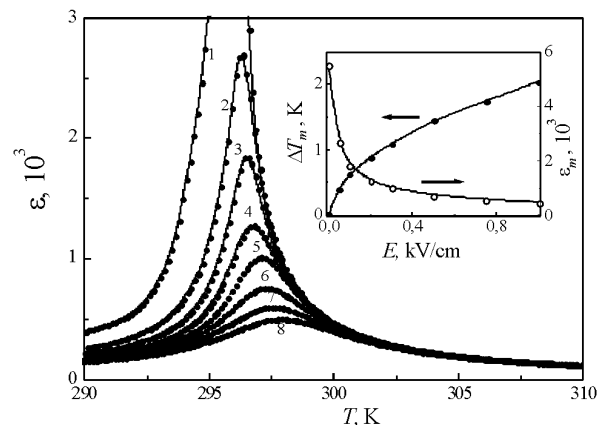

Figure 9. Temperature dependences of the dielectric permeability of Rochelle salt crystals in the vicinity of the upper Curie point at different values of the longitudinal electric field E, kV/cm: 0 (1), 0.05 (2), 0.1 (3), 0.2 (4), 0.3 (5), 0.5 (6), 0.75 (7), 1 (8).
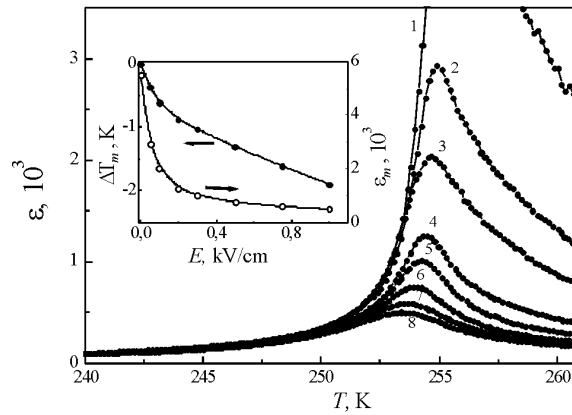
Figure 10. Temperature dependences of dielectric permeability of Rochelle salt crystals in the vicinity of the lower Curie point at different values of the longitudinal electric field E, kV/cm: 0 (1), 0.05 (2), 0.1 (3), 0.2 (4), 0.3 (5), 0.5 (6), 0.75 (7), 1 (8).

Within the framework of thermodynamic theory for a second-order phase transition, the expansion of thermodynamic potential in a series over the powers of the order parameter P is given by

$$F = F_0 + \frac{\alpha}{2}P^2 + \frac{\beta}{4}P^4 - PE, \tag{1}$$

where F_0 is the paraelectric phase energy, E is the electric field, α and β are the expansion coefficients. The α coefficient depends on temperature and changes its sign at the phase transition point. The β coefficient should be positive for a second-order phase transition. By minimizing the expression in equation (1) one obtains the equation of state of a unit volume of a ferroelectric

$$\alpha P + \beta P^3 = E. \tag{2}$$

Inverse dielectric permeability

$$\varepsilon^{-1} = \frac{\partial E}{\partial P} = \alpha + 3\beta P^2 \tag{3}$$

is a function of temperature and of the applied field magnitude. The extremum of this function with respect to temperature can be found as follows:

$$\frac{\partial \varepsilon^{-1}}{\partial \alpha} = 1 + 6\beta P \frac{\partial P}{\partial \alpha} = 0. \tag{4}$$

The $\frac{\partial P}{\partial \alpha}$ value can be found from equation (2), taking into account the fact of the applied field being independent of temperature:

$$\frac{\partial E}{\partial \alpha} = P + \alpha \frac{\partial P}{\partial \alpha} + 3\beta P^2 \frac{\partial P}{\partial \alpha} = 0. \tag{5}$$

Hence

$$\frac{\partial P}{\partial \alpha} = -\frac{P}{\alpha + 3\beta P^2}. \tag{6}$$

By substituting equation (6) into equation (5), one obtains the value of polarization, at which the extremum of the temperature dependence of dielectric permeability is observed:

$$P^2 = \frac{\alpha}{3\beta}. \tag{7}$$

Using equations (7) and (2), one can obtain

$$\alpha = 1.19\beta^{1/3}E^{2/3}. \tag{8}$$

The α coefficient in the vicinity of the upper and lower phase transitions for Rochelle salt crystals can be given by

$$\alpha = \alpha_T(T - T_2), \qquad \alpha = \alpha_T(T_1 - T), \tag{9}$$

where T_1 and T_2 are the temperatures of the lower and the upper phase transitions, respectively. By substitution of equation (9) into equation (8), one arrives at the expressions for the temperatures of dielectric permeability maximum under an electric field:

$$T_{m1} = T_1 - 1.19\alpha_T^{-1}\beta^{1/3}E^{2/3}, \qquad T_{m2} = T_2 + 1.19\alpha_T^{-1}\beta^{1/3}E^{2/3}. \tag{10}$$

In order to find the dielectric permeability value at the maximum point substitute equations (7) and (8) into equation (3):

$$\varepsilon_m = 0.42\varepsilon_0^{-1}\beta^{-1/3}E^{-2/3}. \tag{11}$$

Taking into account the vicinity of the phase transitions in Rochelle salt to the double critical point [16], the coefficient of expansion of thermodynamic potential can be written in the form

$$\alpha = \alpha_{T1} + \alpha_{T2}(T - T_0)^2, \tag{12}$$

where $T_0 = (T_1 - T_2)$ is the midpoint of the temperature interval of the ferroelectric phase existence. By substituting equation (12) into equation (8), one can obtain different expressions for the shift of the temperatures of the dielectric permeability maxima for Rochelle salt crystals:

$$\Delta T_{m1} = A - \sqrt{1,19\alpha_{T2}^{-1}\beta^{1/3}E^{2/3} + A^2}, \qquad \Delta T_{m2} = -A + \sqrt{1,19\alpha_{T2}^{-1}\beta^{1/3}E^{2/3} + A^2}, \tag{13}$$

where $A = \sqrt{-\alpha_{T1}/\alpha_{T2}}$. Contrary to equations (10), in which according to equation (9) the α_T coefficients can be different, in equations (13) for Rochelle salt crystals the shifts of the temperatures of dielectric permeability maxima are the same in the absolute value and opposite in sign for the upper and for the lower phase transitions. Exactly this situation that was observed in [17] where the variation of the ΔT_m absolute value under the electric field for both phase transitions is practically the same within the error limits. Using the experimental data of the discussed paper, we have found the coefficients of expansion of thermodynamic potential in equation (1): $\alpha_{T1} = -7.4 \cdot 10^8$ J·m·C^{-2}, $\alpha_{T2} = 1.58 \cdot 10^6$ J · m · C^{-2}K^{-2}, $\beta = 11.34 \cdot 10^{13}$ J · m^5 · C^{-4}.

Using the experimental data of the studies of hydrostatic pressure effect on the Rochelle salt crystal dielectric permeability, carried out in our earlier paper [17], we have obtained the pressure dependences of α_{T1} and α_{T2} coefficients within the framework of phenomenological theory with the account of the phase transition vicinity to the double critical point. These dependences are shown in figure 11. With the pressure increase, an increase of the α_{T1} value is observed which indicates the phase transition shift away from the double critical point which in the Rochelle salt crystal corresponds to the negative hydrostatic pressure $p = -240$ MPa.

While studying the effect of transverse electric field on the upper phase transition in Rochelle salt crystals we found that the reproducibility of the experimental data for different samples is poor. Such a situation is a consequence of a non-uniform distribution of potentials between the sample plates which results in the formation of a longitudinal electric field as well. The experimental results of [16] enable the effect of this field to be estimated and the dependence of the transition temperature on the transverse electric field strength to be found. The recalculated values of the transition temperature shift due to the electric field are shown in figure 12. As one can see, the effect of transverse electric field, contrary to the longitudinal one, results in the phase transition temperature decrease for all Rochelle salt samples with a coefficient $\Delta T(E)/\Delta E = -0.5 \pm 0.1$ K·cm/kV. Theoretical calculations of the transverse electric field effect on the Rochelle salt dielectric properties performed within the framework of the Mitsui model [5], show that transverse external fields make the ferroelectric phase existence interval narrower. This is confirmed by our experimental data.
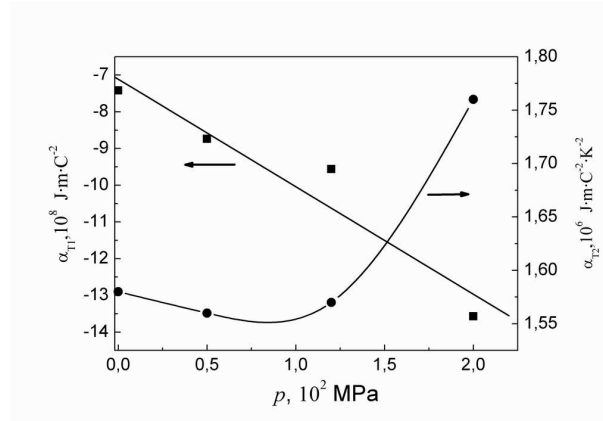
Figure 11. Pressure dependence of the thermodynamic potential expansion coefficients α_{T1} and α_{T2} of Rochelle salt crystals.
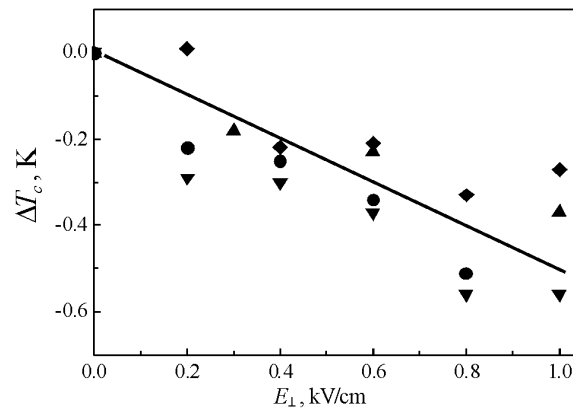

Figure 12. Dependence of the upper phase transition temperature on the transverse electric field strength for different Rochelle salt samples.

5. Conclusions

Based on the experimental studies performed, we discuss the regularities of variation of anisotropy of dielectric permeability of KH_2PO_4 and $NaKC_4H_4O_6 \cdot 4H_2O$ crystals due to temperature, pressure, and electric field. In KDP crystals we experimentally observed new high-temperature anomalies (250–300 K) in the temperature dependences of transverse dielectric permeability and dielectric loss angle tangent which are due to the phase transition that occurs in local domains of nanometric size with the $\overline{4}2m \longrightarrow mm2$ symmetry change. Under external hydrostatic pressure, the temperatures of these phase transitions increase. Within the framework of Landau phenomenological theory, the experimental results for temperature dependences of longitudinal dielectric permeability of Rochelle salt crystals under external longitudinal and transverse electric field are analyzed. The coefficients α_{T1} and α_{T2} of thermodynamic potential expansion over the order parameter are calculated with the account of the vicinity of phase transitions in Rochelle salt to the double critical point. The increase of the α_{T1} absolute value with hydrostatic pressure p shows that the phase transitions in Rochelle salt shift away from the double critical point which corresponds to $p = -240$ MPa. It is shown experimentally that the effect of transverse electric field, contrary to the longitudinal one, results in the upper phase transition decrease with a coefficient $\Delta T(E)/\Delta E = -0.5 \pm 0.1$ K·cm/kV.

References

1. Stasyuk I.V., Biletskii I.N., Bull. Acad. Sci. USSR. Phys. Ser., 1983, **4**, 78.
2. Stasyuk I.V., Biletskii I.N., Styahar O.N., Ukr. Phys. J., 1986, **31**, No. 3, 567.
3. Levitskii R.R., Zachek I.R., Moina A.P., Verkholyak T.M., J. Phys. Studies, 2003, **7**, 106.
4. Moina A.P., Levitskii R.R., Zachek I.R., Phys.Rev.B, 2005, **71**, 134.
5. Stasyuk I.V., Velychko O.V., Ferroelectrics, 2005, **316**, 51.
6. Nakamura E., Kuramoto K., J. Phys. Soc. Jpn., 1988, **57**, No. 6. 2182.
7. Levitskii R.R., Slivka A.G., Moina A.P., Lukach P.M., Guivan A.M., J. Phys. Studies, 2002, **6**, No. 2, 197.
8. Gesi K., Ozawa K., J. Phys. Soc. Jpn., 1984, **53**, No. 12, 4405.
9. Levitskii R.R., Slivka A.G., Moina A.P., Lukach P.M., Guivan A.M. Preprint arXiv:cond-mat/0012053v1, 2000.
10. Kedyulich V.M., Slivka A.G., Gerzanich E.I., Guivan A.M., Conden. Matter Phys., 2002, **5**, No. 4(32), 761.
11. Kedyulich V.M., Slivka A.G., Gerzanich E.I., Guivan A.M., Lukach P.M., Conden. Matter Phys., 2003, **6**, No. 2(34), 271.
12. Gridnev S.A., Kravchenko S.A., Fiz. Tverd. Tela (Leningrad) [Sov. Phys. Solid State], 2000, **42**, No. 11, 2074.
13. Suvorova E.I., Klechkovskaya V.V., Kristallografiya, 1991, **36**, No. 3, 729.
14. Suvorova E.I., Klechkovskaya V.V., Ferroelectrics, 1993, **144**, 245.
15. Levitskii R.R., Zachek I.R., Verkholyak T.M., Moina A.P., Phys. Rev. B, 2003, **67**, 174112.
16. Kryukova E.B., Sobyanin A.A., Zh. Eksp. Teor. Fiz. [Sov. Phys. JETP] , 1987, **51**, No. 12, 2090.
17. A.G. Slivka, V.M. Kedyulich, R.R. Levitskii, A.P. Moina, M.O. Romanyuk, A.M. Guivan., Condens. Matter Phys., 2005, **8**, No. 3(43), 623.

Вплив зовнішніх полів на анізотропію діелектричної проникності кристалів KH_2PO_4 та $NaKC_4H_4O_6 \cdot 4H_2O$

О.Г.Сливка, В.М.Кедюлич, Г.М.Гуйван

Ужгородський національний університет, кафедра оптики Вул.Волошина, 54, 88000, Ужгород, Україна

Проведено експериментальні дослідження впливу температури, зовнішнього тиску та електричного поля на анізотропію діелектричної проникності кристалів KH_2PO_4 та $NaKC_4H_4O_6 \cdot 4H_2O$. У рамках феноменологічної теорії та з врахуванням близькості фазових переходів до подвійної критичної точки проведено аналіз експериментальних результатів по впливу електричного поля та тиску на діелектричну проникність сегнетової солі. Визначено коефіцієнти розкладу термодинамічного потенціалу по параметру порядку і їх баричну поведінку. Показано, що збільшення гідростатичного тиску віддаляє фазові переходи в сегнетовій солі від подвійної критичної точки.

Ключові слова: *сегнетоелектрики, фазовий перехід, діелектрична проникність, температура, тиск, електричне поле*

PACS: *77.80.Bh, 77.22.Ch, 77.84.Fa*