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Features of the Behavior of Longitudinal Static and Dynamic Dielectric, Piezoelectric, and Elastic Characteristics of KH_2PO_4 and $NH_4H_2PO_4$ Crystals

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Abstract—Along with their electromechanical coupling coefficients, the longitudinal dielectric, piezoelectric and elastic characteristics in ferroelectric KH_2PO_4 and antiferroelectric $NH_4H_2PO_4$ crystals are calculated using a modified proton ordering model that considers piezoelectric coupling and the four-particle cluster approximation. The possibility of detecting piezoactivity in solid solutions of $K_{1-x}(NH_4)_xH_2PO_4$ is substantiated.

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INTRODUCTION

Ferroelectric crystals of the KH_2PO_4 family exhibit piezoelectric behavior that affects their physical behavior. This problem, however, has not been properly studied in the past. Descriptions of the dielectric features of the ferroelectric KH_2PO_4 family compounds using a common proton model is generally limited to examining their static behavior and high-frequency relaxation [1–3]. Studies of piezoelectric resonance on a model without piezoelectric coupling are meaningless. It should be noted that the only way to correct measurements of the high-frequency dielectric behavior of the KH_2PO_4 family compounds is to consider piezoelectric coupling. In contrast, the classical proton model is unable to describe the effects of unequal unclamped and clamped crystals, or the phenomenon of a crystal clamped in a high-frequency field.

A modified proton ordering model considering piezoelectric coupling and four-particle cluster approximation was used to obtain static and dynamic dielectric, piezoelectric and elastic behavior of the KH_2PO_4 family crystals in [4–8]. A detailed numerical analysis was made of the results, and the optimum sets of microparameters were found in order to describe well quantities of the experimental data.

In addition, a cluster theory of the thermodynamic and dynamic behavior of compounds of the $Rb_{1-x}(NH_4)_xH_2PO_4$ type was developed in [9–13]. If the parameters are selected properly, the theory describes experimental data in terms of quantity fairly well. Possible piezoelectric coupling in the crystal is ignored in the proposed theory, however, though the authors acknowledge its existence [14]. The results obtained in [9–13] for ferroelectric (FE) KH_2PO_4 (KDP) and antiferroelectric (AFE) $NH_4H_2PO_4$ (ADP) are reviewed briefly in this work. The electromechanical coupling factors of the crystals are estimated numerically. The possibility of detecting piezoelectric coupling in compounds of $K_{1-x}(NH_4)_xH_2PO_4$ type is discussed.

DIELECTRIC, PIEZOELECTRIC, AND ELASTIC CHARACTERISTICS

Dynamic behavior of the FE KDP and AFE ADP in the presence of piezoelectric coupling and deformation ε_6 are studied using a dynamic model of ferroelectric orthophosphate using four-particle cluster approximation, which is based on the stochastic Glauber model. The dynamic deformation process in the crystals is modeled with classical motion equations [7, 8]. On the basis of these techniques, we can write the longitudinal dynamic susceptibility of mechanically unclamped KDP and ADP crystals:

$$\chi_{33}^{\sigma}(\omega) = \chi_{33}^{\varepsilon}(\omega) + \frac{1}{R_6(\omega)}\frac{e_{36}^2(\omega)}{c_{66}^E(\omega)}, \tag{1}$$

where

$$\frac{1}{R_6(\omega)} = \frac{2}{k_6 l}\operatorname{tg}\frac{k_6 l}{2}, \quad k_6 = \frac{\omega\sqrt{\rho}}{\sqrt{c_{66}^E(\omega)}}, \tag{2}$$

and ω is a frequency of external field, ρ is a density of the crystal, and l is the length of a thin square crystal plate ($l = 1$ mm) made in the plane (001).

The longitudinal dynamic susceptibility of the mechanically clamped crystal $\chi_{33}^{\varepsilon}(\omega)$ in (1), and the $e_{36}(\omega)$ and $c_{66}^{E}(\omega)$ for KDP, are determined as follows:

$$\chi_{33}^{\varepsilon}(\omega) = \chi_{33}^{\varepsilon 0} + \frac{\beta\mu_3^2}{2\upsilon} F^{(1)}(\omega), \quad \beta = \frac{1}{k_B T}, \tag{3}$$

$$e_{36}(\omega) = e_{36}^{0} + \frac{\beta\mu_3}{\upsilon} L_s(\omega), \tag{4}$$

$$\begin{aligned} c_{66}^{E}(\omega) &= c_{66}^{E0} + \frac{4\beta\psi_6}{\upsilon D_s} f_s + \frac{2\beta}{\upsilon D_s^2}(-\delta_{s6}M_{s6} \\ &+ \delta_{16}M_{16} + \delta_{a6}M_{a6})^2 + \frac{4\beta\psi_6}{\upsilon} L_s(\omega) \\ &- \frac{4\varphi_s f_s}{\upsilon D_s}\beta L_s(\omega) - \frac{2\beta}{\upsilon D_s}\big[\delta_{s6}^2 \operatorname{ch}(2z_6 + \beta\delta_{s6}\varepsilon_6) \\ &+ 4b\delta_{16}^2 \operatorname{ch}(z_6 - \beta\delta_{16}\varepsilon_6) + \delta_{a6}^2 2a \operatorname{ch}\beta\delta_{a6}\varepsilon_6^2\big], \end{aligned} \tag{5}$$

where k_B is the Boltzmann constant; μ_3 is the effective dipole moment of a hydrogen bond; υ is the volume of a primitive crystal cell consisting of two PO_4 tetrahedrons; ε_6 is a spontaneous deformation; ψ_6, δ_{s6}, δ_{16}, and δ_{a6} are deformation potentials; and $\chi_{33}^{\varepsilon 0}$, e_{36}^{0}, c_{66}^{E0} are the initial values of dielectric susceptibility, the piezoelectric coefficient, and the elastic constant, respectively:

$$\begin{aligned} L_s(\omega) &= -\psi_6 F^{(1)}(\omega) + \delta_{s6}F_s^{(1)}(\omega) \\ &+ \delta_{16}F_1^{(1)}(\omega) - \delta_{a6}F_a^{(1)}(\omega), \end{aligned}$$

while the expressions for $F^{(1)}(\omega)$, $F_s^{(1)}(\omega)$, $F_a^{(1)}(\omega)$, $F_1^{(1)}(\omega)$, are listed in [15]:

$$\begin{gathered} f_s = \delta_{s6}\cosh(2z_6 + \beta\delta_{s6}\varepsilon_6) - 2b\delta_{16}\cosh(z_6 - \beta\delta_{16}\varepsilon_6) \\ + \eta_s(-\delta_{s6}M_{s6} + \delta_{a6}M_{a6} + \delta_{16}M_{16}); \\ M_{a6} = 2a\sinh\beta\delta_{a6}\varepsilon_6, \\ M_{s6} = \sinh(2z_6 + \beta\delta_{s6}\varepsilon_6), \; M_{16} = 4b\sinh(z_6 - \beta\delta_{16}\varepsilon_6), \end{gathered}$$

$$\varphi_s = \frac{1}{1-\eta_s^2} + \beta\nu_c,$$

$$\eta_s = \frac{1}{D_s}(\sinh(2z_6 + \beta\delta_{s6}\varepsilon_6) + 2b\sinh(z_6 - \beta\delta_{16}\varepsilon_6)),$$

$$\begin{gathered} D_s = \cosh(2z_6 + \beta\delta_{s6}\varepsilon_6) + 4b\cosh(z_6 - \beta\delta_{16}\varepsilon_6) \\ + 2a\cosh\beta\delta_{a6}\varepsilon_6 + d, \end{gathered}$$

$$z_6 = \frac{1}{2}\ln\frac{1+\eta_s}{1-\eta_s} + \beta\nu_c\eta_s - \beta\psi_6\varepsilon_6,$$

$$a = e^{-\beta\varepsilon}, \quad b = e^{-\beta w}, \quad d = e^{-\beta w_1},$$

where ε, w, w_1 are the energy parameters of the Slater–Takagi model.

Along with the $e_{36}(\omega)$ and $c_{66}^{E}(\omega)$ values for an ADP crystal, the longitudinal dynamic susceptibility of a mechanically clamped crystal $\chi_{33}^{\varepsilon}(\omega)$ is

$$\chi_{33}^{\varepsilon}(\omega) = \chi_{33}^{\varepsilon 0} + \frac{\beta\mu_3^2}{\upsilon} F^{(1)}(\omega), \tag{6}$$

$$e_{36}(\omega) = e_{36}^{0} + \frac{\beta\mu_3}{\upsilon} L_a(\omega), \tag{7}$$

$$\begin{aligned} c_{66}^{E}(\omega) &= c_{66}^{E0} + \frac{4\beta\psi_6}{\upsilon D_a} L_a(\omega) - \frac{4\varphi_a f_a}{\upsilon D_a}\beta L_a(\omega) + \frac{4\beta\psi_6}{\upsilon D_a} f_a \\ &- \frac{2\beta}{\upsilon D_a}\big[\delta_{s6}^2 a' + \delta_{16}^2 4b' + \delta_{a6}^2(1 + \cosh 2x)\big], \end{aligned} \tag{8}$$

where $L_a(\omega) = -2\psi_6 F^{(1)}(\omega) + \delta_{s6}F_s^{(1)}(\omega) - \delta_{a6}F_a^{(1)}(\omega) + \delta_{16}F_1^{(1)}(\omega)$, the expressions for $F^{(1)}(\omega)$, $F_s^{(1)}(\omega)$, $F_a^{(1)}(\omega)$, $F_1^{(1)}(\omega)$ are listed in [16].

$$f_a = \delta_{s6}a' - \delta_{16}2b'\cosh x, \quad \varphi_a = \frac{1}{1-\eta_a^2} + \beta\nu_c(0);$$

$$\eta_a = \frac{1}{D_a}(\sinh 2x + 2b'\sinh x),$$

$$D_a = a' + \cosh 2x + d' + 4b'\cosh x + 1,$$

$$x = \frac{1}{2}\ln\frac{1+\eta_a}{1-\eta_a} + \beta\nu_a\left(k^z\right)\eta_a.$$

$$a' = e^{-\beta\varepsilon'}, \; b' = e^{-\beta w'}, \; d' = e^{-\beta w_1'},$$

where ε', w', w_1' are the antiferroelectric energy values of the Slater–Takagi model.

At the static limit ($\omega \to 0$), Eqs. (3)–(8) yield expressions for the isothermal static dielectric susceptibility of clamped crystals:

$$\text{KDP: } \chi_{33}^{\varepsilon} = \chi_{33}^{\varepsilon 0} + \bar{\upsilon}\frac{\mu_3^2}{\upsilon^2}\frac{1}{T}\frac{2\kappa_s}{D_s - 2\kappa_s\varphi_s}, \quad \bar{\upsilon} = \frac{\upsilon}{k_B}, \tag{9}$$

$$\text{ADP: } \chi_{33}^{\varepsilon} = \chi_{33}^{\varepsilon 0} + \bar{\upsilon}\frac{\mu_3^2}{\upsilon^2}\frac{1}{T}\frac{2\kappa_a}{D_a - 2\kappa_a\varphi_a}, \tag{10}$$

where

$$\begin{gathered} \kappa_s = \operatorname{ch}(2z_6 + \beta\delta_{s6}\varepsilon_6) + b\operatorname{ch}(z_6 - \beta\delta_{16}\varepsilon_6) - \eta_s^2 D_s, \\ \kappa_a = a' + b'\operatorname{ch}x. \end{gathered}$$

Isothermal piezoelectric coefficients are found in a similar manner:

In the case of KDP:

$$e_{36} = e_{36}^{0} + \frac{2\mu_3}{\upsilon}\frac{\beta\left(-2\kappa_s\psi_6 + f_s\right)}{D_s - 2\varphi_s\kappa_s}, \tag{11}$$

in the case of ADP:

$$e_{36} = e_{36}^{0} + 2\frac{\mu_3}{\upsilon}\frac{\beta(-2\kappa_a\psi_6 + f_a)}{D_a - 2\varphi_a\kappa_a}, \tag{12}$$

Table 1. Collections of optimum model parameters for KDP crystal

T_c	ε/k_B	w/k_B	ν_c/k_B	μ_{3-}, 10^{-18}	μ_{3+}, 10^{-18}	χ_{33}^{0}
(K)	(K)	(K)	(K)	(CGSE$q \cdot$ cm)	(CGSE$q \cdot$ cm)	
122.5	56.00	422.0	17.91	1.46	1.71	0.73

ψ_6/k_B	δ_{s6}/k_B	δ_{a6}/k_B	δ_{16}/k_B	$c_{66}^{0} \cdot 10^{-10}$	e_{36}^{0}
(K)	(K)	(K)	(K)	(d/cm^2)	(CGSEq/cm)
−150.00	82.00	−500.00	−400.00	7.10	1000.00

and the KDP and ADP crystals' isothermal elastic elements in a constant field are

$$c_{66}^{E} = c_{66}^{E0} + \frac{8\psi_6}{\upsilon}\frac{\beta\left(-\psi_6\kappa_s + f_s\right)}{D_s - 2\varphi_s\kappa_s} - \frac{4\beta\varphi_s f_s^2}{\upsilon D_s(D_s - 2\varphi_s\kappa_s)} - \frac{2\beta}{\upsilon D_s}\Big[\delta_{s6}^2\cosh\left(2z_6 + \beta\delta_{s6}\varepsilon_6\right) + \delta_{a6}^2 2a\cosh\beta\delta_{a6}\varepsilon_6 + \delta_{16}^2 4b\cosh\left(z_6 - \beta\delta_{16}\varepsilon_6\right)\Big] + \frac{2\beta}{\upsilon D_s^2}(-\delta_{s6}M_{s6} + \delta_{a6}M_{a6} + \delta_{16}M_{16})^2, \quad (13)$$

$$c_{66}^{E} = c_{66}^{E0} + \frac{8\psi_6}{\upsilon}\frac{\beta(-\psi_6\kappa_a + f_a)}{D_a - 2\varphi_a\kappa_a} - \frac{4\beta\varphi_a f_a^2}{\upsilon D_a\left(D_a - 2\varphi_a\kappa_a\right)} - \frac{2\beta}{\upsilon D_a}\left(\delta_{16}^2 4b'\mathrm{ch}x + \delta_{s6}^2 a' + \delta_{a6}^2 2\mathrm{ch}^2 x\right). \quad (14)$$

Assuming that $\sigma = \mathrm{const}$ for KDP and ADP crystals, the isothermal elastic constant for fixed polarization $c_{66}^{P} = c_{66}^{E} + e_{36}^2/\chi_{33}^{\varepsilon}$;, the isothermal piezoelectric strain coefficient $d_{36} = e_{36}/c_{66}^{E}$, and isothermal dielectric susceptibility $\chi_{33}^{\sigma} = \chi_{33}^{\varepsilon} + e_{36}d_{36}$ can be found on the basis of the well-known relations between elastic, dielectric and piezoelectric values [5, 6].

Table 2. Collections of optimum model parameters for ADP crystal

T_N	ε'/k_B	w'/k_B	$\nu_c(0)/k_B$	μ_{3+}, 10^{-18}	$\chi_{33}^{0\varepsilon}$
(K)	(K)	(K)	(K)	(CGSE$q \cdot$ cm)	
148	20	490.0	−10.00	2.10	0.23

ψ_6/k_B,	δ_{s6}/k_B,	δ_{a6}/k_B,	δ_{16}/k_B,	$c_{66}^{0}, 10^{-10}$	e_{36}^{0}
(K)	(K)	(K)	(K)	(d/cm^2)	(CGSEq/cm)
−160	1400	100	−300	7.9	10000

COMPARING THE NUMERICAL RESULTS AND EXPERIMENTAL DATA

Note that the theoretical results presented above are, strictly speaking, valid for deuterated crystals of DKDP and DADP. Considering the suppression of tunneling by short-term correlations [17], we shall assume that these results are correct for both KDP and ADP.

To calculate the physical characteristics of KDP and ADP crystals, our theory proposes that the physical characteristics of KDP and ADP crystals be calculated on the basis of the pre-defined parameters (Tables 1 and 2) found in [15, 16].

The volume of a primitive KDP crystal cell is taken to be $\upsilon = 0.1936 \times 10^{-21}$ cm^3, and $\upsilon = 0.2110 \times 10^{-21}$ cm^3 for ADP. Accordingly, the density of KDP and ADP crystals $\rho = 2.31$ g/cm^3 and $\rho = 1.804$ g/cm^3 [18].

The temperature dependence of calculated isothermal static dielectric permeability of unclamped $\varepsilon_{33}^{\sigma}$ and clamped $\varepsilon_{33}^{\varepsilon}$ crystals of KDP and ADP are presented in Fig. 1 together with experimental data. In the paraelectric phase, the value of $\varepsilon_{33}^{\sigma}$ grows hyperbolically in the vicinity of T_c and reaches very high values at $T = T_c$. Below T_c, when the temperature falls, $\varepsilon_{33}^{\sigma}(T)$ decays quickly. The Curie-Weiss law is thus valid in the temperature interval $\Delta T = T - T_c \approx 50$ K above the value T_c. Permeability $\varepsilon_{33}^{\sigma}$ for ADP is approximately 18% higher than $\varepsilon_{33}^{\varepsilon}$. This difference is insensitive to temperature.

The calculated temperature dependence of piezoelectric strain coefficient d_{36} for KDP and ADP crystals is presented in Fig. 2. Elastic constant c_{66}^{E} of the KDP crystal converges to zero when $T \to T_c$, while the function $c_{66}^{P}(T)$ does not reveal any anomalies in the vicinity of the phase transition. Unlike KDP, elastic constant c_{66}^{E} for ADP reaches finite values at $T = T_N$ and is insensitive to temperature.

As we can see in Figs. 1–3, the proposed theory adequately describes experimental data for static dielectric, piezoelectric, and elastic KDP and ADP

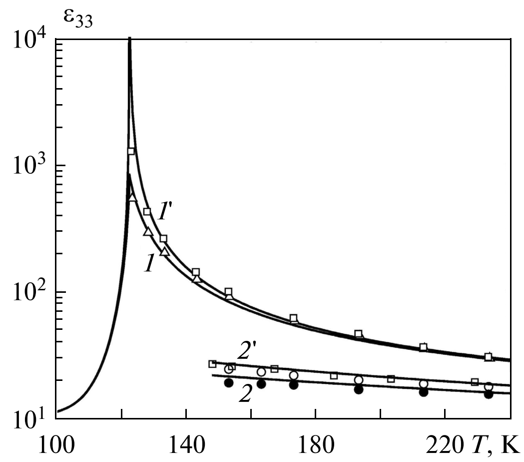

Fig. 1. Temperature dependence of dielectric permeability of clamped $\varepsilon_{33}^{\varepsilon}$ (*1*) and unclamped $\varepsilon_{33}^{\varepsilon}$ (*1*') KDP crystals, of clamped (*2*) and unclamped (*2*') ADP crystals. Curves correspond to the theoretical results; symbols mark experimental data adopted from referenced sources [18, 25].

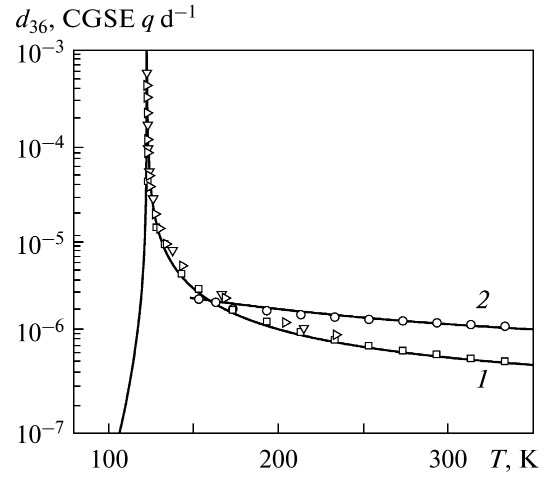

Fig. 2. Temperature dependence of piezoelectric strain coefficient d_{36} of KDP (*1*) and ADP (*2*) crystals. Curves correspond to theoretical results; symbols mark experimental data: ○, □ [18], ▽ [19], ▷ [20].

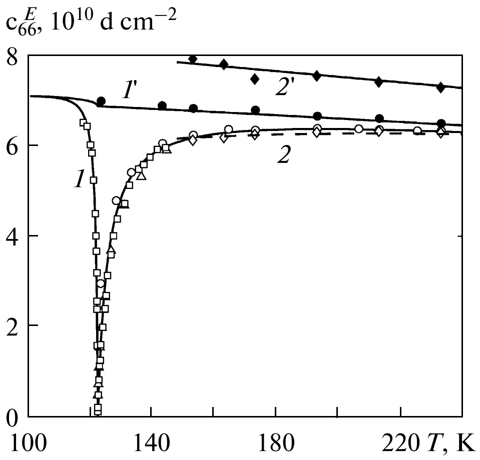

Fig. 3. Temperature dependence of elastic constants c_{66}^E and c_{66}^P of KDP (*1*, *1*', respectively) and ADP (*2*, *2*') crystals. Curves correspond to theoretical results, symbols mark experimental data: ○ [18], □ [21], △ [22], ◇ [18].

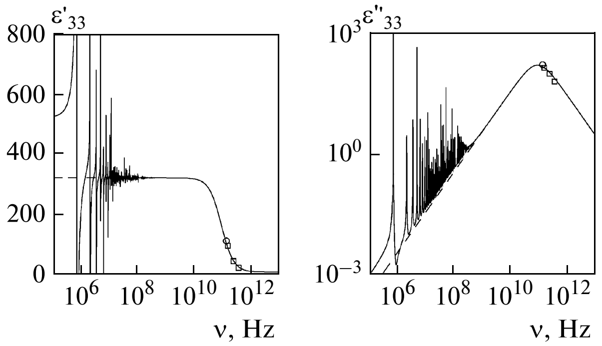

Fig. 4. Frequency dependence of real and imaginary parts of dielectric permeability of unclamped and clamped KDP crystals at $\Delta T = 5$ K: ○ [23], □ [24]. Curves represent theoretical results.

crystals. Note that studies of the temperature dependency of the difference $\varepsilon_{33}^{\sigma} - \varepsilon_{33}^{\varepsilon} = 4\pi e_{36} d_{36} = 4\pi d_{36}^2 c_{66}^E$ for KDP and ADP crystals is important in estimating the values of piezoelectric coefficients and elastic constants of ferroelectric compounds.

Let us now examine the results from calculations of the longitudinal dynamic characteristics of mechanically unclamped crystals of KDP and ADP. Diagonal deformations ε_i are ignored for the sake of simplicity. The frequency dependences of the real and imaginary parts of the longitudinal dielectric permeability of mechanically unclamped KDP crystals at $\Delta T = 5$ K and ADP crystals at $\Delta T = 28$ K are shown in Figs. 4 and 5. Resonance-type scattering is observed in the range of $3 \cdot 10^5$–$3 \cdot 10^8$ Hz. The resonance frequencies are inversely proportional to the sample size.

Temperature dependency of electromechanical-coupling coefficients $k_3^2 = \left(\varepsilon_{33}^{\sigma} - \varepsilon_{33}^{\varepsilon}\right)/\varepsilon_{33}^{\sigma}$ of KDP and ADP crystals are displayed in Fig. 6. The value k_3^2 for KDP reaches its maximum at transit point and decays dramatically when pulled away from T_c point, especially in ferroelectric phase. The ADP's coefficient k_3^2 reaches maximum of approximately 0.35, when $T =$

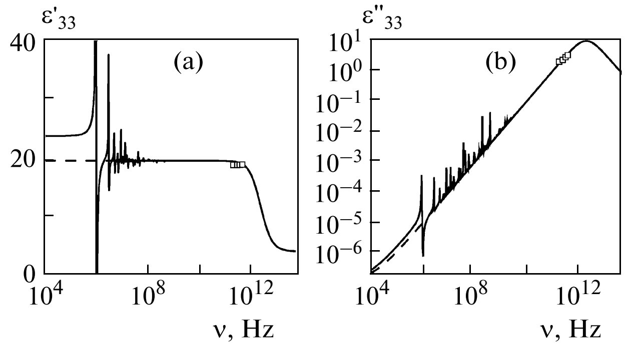

Fig. 5. Frequency dependence of real and imaginary parts of dielectric permeability of unclamped and clamped (dashed line) ADP crystals at $\Delta T = 28$ K: □ [26]. Curves represent theoretical results.

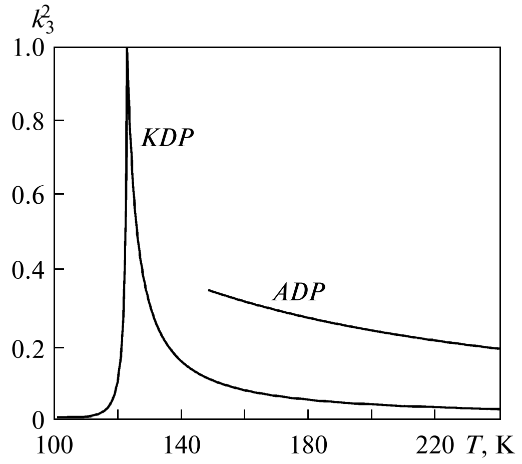

Fig. 6. Temperature dependence of the electromechanical coupling coefficient of KDP and ADP crystals.

T_N, and descends slowly with temperature growth. Meanwhile, the ADP's value of k_3^2 in paraelectric phase is substantially higher than that of KDP.

The results of this section indicate that piezoelectric coupling in crystals of the KH_2PO_4 family leads to the difference between the dielectric permeability of mechanically unclamped and clamped crystals, and determines their piezoelectric modules and piezoelectric resonance. Unfortunately, similar theoretical and experimental studies have not been undertaken on irregular compounds such as $K_{1-x}(NH_4)_xH_2PO_4$. An exception was described in [14]. In order to prove there is piezoelectric coupling in a compound, it is crucial to examine $\varepsilon'_{33}(\nu, T)$ and $\varepsilon''_{33}(\nu, T)$ over wide temperature and frequency ranges, and to estimate the electromechanical coupling coefficient and difference in permeability for unclamped and clamped crystals. The latter indicates the presence of non-zero piezoelectric modules in a compound. A detailed study of the listed characteristics as a function of NH_4 ionic cluster density in these compounds would also be of interest.

CONCLUSIONS

The expressions of permeability of unclamped and clamped crystals, the piezoelectric strain coefficient, the elastic constant, and the dynamic behavior of KDP and ADP crystals were revealed by considering piezoelectric coupling. Calculating the temperature dependency of these characteristics demonstrated the considerable difference between $\varepsilon_{33}^{\sigma}$ and $\varepsilon_{33}^{\varepsilon}$ measured experimentally for KDP crystals, along with minor differences for ADP. The optimum sets of parameters and initial characteristics of the crystals under study were found, and were helpful in describing the available experimental data. The phenomena of clumping and piezoelectric resonance in the studied crystals have been described for the first time. Similar research for compounds of type $K_{1-x}(NH_4)_xH_2PO_4$ will produce important data whether or not they contain piezoelectric coupling.

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