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# NONLINEAR ELECTROMECHANICAL PROPERTIES OF FERROELECTRIC $NaND_4SeO_4$ $2D_2O$

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Both theoretical and experimental study of nonlinear electromechanical properties for ferroelectric $NaND_4SeO_4.2D_2O$ were carried out above Curie temperature $T_c$. The minimum value of $C^E_{ikl}$ was found at temperature $T = T_c + (1.5 \div 2)$ K. It was shown that this temperature corresponds to the boundary of the strong fluctuation range.



The results of both theoretical and experimental investigations of the nonlinear electromechanical properties (NEMP) near phase transition (PhT) of $NaND_4SeO_4.2D_2O$ (NASeD(d)) are considered. The point group of symmetry in this crystal is 222, and $T_c \approx T_0 = 180$ K.[1,2]

As to nonlinear properties of this crystal, only optical[3] and electrostrictive[1] ones are known. The electrostriction proves to be an order higher than that of most other ferroelectrics even at room temperature. It is of interest to consider other NEMP (elastic and piezoelectric). In order to eliminate the strong influence of the domain structure[1] and to compare the experiment with the theory, the results will be presented only above Curie point.

## THEORY

Effective Hamiltonian (free energy density) for ferroelectrics possessing piezoelectricity in paraphase is given in Ref. 4. The equation of motion is

$$-\Gamma \frac{\partial P_k}{\partial t} = \frac{\partial H^{et}}{\partial P_k} + \tilde{\eta}(k, t); \tag{1}$$

assuming that $U_k{}^l$ follows ordinary equations of the elastic theory

$$\ddot{U}_k{}^l = ik \frac{\partial H^{et}}{\partial U_k{}^l} + ik\tilde{\vartheta}(k, t); \tag{2}$$

where $\Gamma$ is kinetic coefficient, $\tilde{\eta}$, $\tilde{\vartheta}$ are random forces, $U_k{}^l$ are vector components of ultrasonic displacements, $P_k$ is component of the polarization vector along polar axes. $H^{\text{eff}} = H_0 + \Delta H$, where $\Delta H$ is due to fluctuations.[4] In relaxation region we can put $H^{\text{eff}} \approx H_0$, $\tilde{\vartheta}$, $\tilde{\eta} = 0$.

The solution of Eqs. (1) and (2) at $U_k{}^i \sim e^{i\omega t}$ [4] gives the following expression for the third-order elastic constants:

$$\begin{aligned} C^E_{ijl} = &- q_{NN\xi} h_{Nj} h_{N\xi_1} (C^P_{\xi_1 l})^{-1} \{C^{E(2\omega)}_{\xi l} (L^\omega)^2 \\ &+ L^\omega L^{2\omega} C^{E(\omega)}_{\xi l}\} \\ &+ h_{N\xi\nu} h_{Nj} (C^P_{\xi\xi_1} C^P_{\nu\nu_1})^{-1} C^{E(\omega)}_{\nu_1 l} \{C^{E(2\omega)}_{\xi_1 l} L^\omega + \\ &L^{2\omega} C^{E(\omega)}_{\xi_1 l}\} \\ &+ C^P_{\xi_1\xi_2\xi_3} (C^P_{\xi_1\nu_1} C^P_{\xi_2\nu 2} C^P_{\xi_3\xi_3})^{-1} C^{E(2\omega)}_{\nu_1 l} C^{E(\omega)}_{\nu_2 j} C^{E(\omega)}_{\nu_3 l}; \end{aligned} \tag{3}$$

where $L^\omega = \Gamma^{-1}\tau(1 + i\omega\tau)^{-1}$; $\tau = \Gamma\xi_n$; $C^{E(\omega)}_{ij} = C^P_{ij} - \frac{1}{2} h_{Ni} h_{Nj} L^\omega$. $\varepsilon_N$ is dielectric susceptibility at constant strain. Corresponding expressions for electrostrictive and nonlinear piezoelectric coefficients are given in Ref. 4.

In fluctuation region $H^{\text{eff}} \approx \Delta H$. For ferroelectrics with dipole-dipole interaction the second order PhT is known to be Gaussian fixed point.[5] In this case the term for the direct interaction of polarization fluctuation can be omitted. Then $C_{ijl}$ is given by Ref. 4:

$$\begin{aligned} &C^E_{ijl} - C^P_{ijl} \sim \tau_1^{-\nu} \{f_1(\omega\tau_1) + i\omega f_2(\omega\tau_1)\}; \\ &\tau_1 = \Gamma[\varepsilon_N^{-1} - \tfrac{1}{2} h_{Ni} h_{Nj} (C^P_{ij})^{-1}]^{-1} \sim c\Gamma(T - T_c)^{-1}; \end{aligned} \tag{4}$$

Expressions for $f_1$ and $f_2$ which are smooth functions of $\omega\tau_1$ are given in Ref. 4. Critical exponent $\nu$ depends on indices $(ijl)$. For the crystals piezoactive in paraelectric phase $\nu = \frac{1}{2}$ when $i = j = l$, $\nu < 0$ when $i \neq j \neq l$ and $\nu \sim 0$ when $i \neq j = l$ and $(T - T_c) \gtrsim \frac{1}{2} h_{Ni} h_{Nj} [C^P_{ij} c]^{-1}$ (logarithmic increase). As in the vicinity of the PhT $C^E_{ij} \to 0$ and $L^\omega$ remains finite at $\tau_1 \to \infty$ Eq. (3) gives the decreasing of $C^E_{ijl}$ due to the relaxation mechanism. But Eq. (4) gives the abnormal increase for $C^E_{ijl}$ due to strong fluctuations. So, there must be minimum in the temperature dependence of $C^E_{ijl}$ the position of which determines the boundary of strong fluctuations region.

## EXPERIMENTAL RESULTS AND DISCUSSION

The nonlinear piezoelectric and elastic coefficients were measured by resonant method. The bars of 22·5° and 45° Z-cuts and [112] cut were used. The temperature stability was better than 0.01 K. Nonlinear piezocoefficients were obtained from the resonance frequency shift due to the biasing field.[6] By applying these piezocoefficients to the second harmonic generation measurements[7] the third order elastic coefficients were calculated (Figure 1). The temperature dependence analysis of $S^E_{i66}$ ($i = 1, 2, 3$) and $d_{326}$ shows that in the region $(T - T_c) > 2$ K they increase with the critical index $\nu = 1$. It is in agreement with estimations for relaxation mechanism.[4] The dependence log $S^E_{i66}$ *vs.* log $(\Delta T)$ in the range of $(T - T_c) \approx 2$ K shows the break. Figure 2 gives the temperature dependence of the third order elastic moduli $C_n^E$ calculated from the experimental values of $S_n^E$. In the range $(T - T_c) \approx$

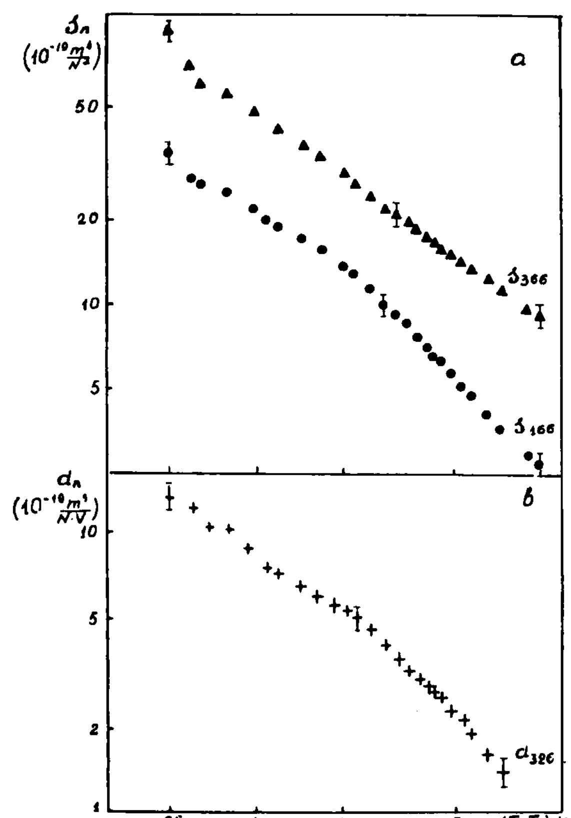

FIGURE 1 The temperature dependence of nonlinear coefficients, a) $S^E_{166}$ and $S^E_{366}$, b) $d_{326}$.

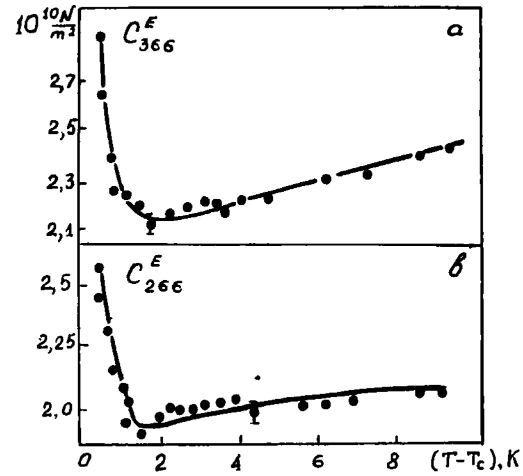

FIGURE 2 The temperature dependence of the third-order elastic moduli, a) $C^E_{366}$, b) $C^E_{266}$.

$(1.5 \div 2)$ K the minimum is observable. The temperature of $C^E_{i66}$ minimum coincides with that of $S^E_{i66}$ break. This leads to the conclusion that there are some changes in mechanism in this temperature range. The critical exponent $\nu \sim 0.1$ for $C^E_{266}$ at $(T - T_c) < 1.5$ K indicates the logarithmic increase in agreement with the theory. As Gruneisen constant of this crystal is high enough ($\gtrsim 10$) and $h^2_{36}/2C^p_{66}C$ value is small the last term makes the main contribution to Eq. (4). It results in decrease of $C^E_{Ujl}$. This decrease is not so great (because the value $\frac{1}{2}h_{36}/C^p_{66}\,C$ is small) at $(T - T_c) \gtrsim 2$ K.

Thus, the fluctuation range $T_{fl}$ in NASeD(d) is found experimentally to be $1.5 \div 2$ K.

It should be noted that such temperature dependence is likely to be observed for all crystals possessing the linear coupling between the strain and the order parameter in paraelectric phase. The study of the temperature dependence of the third order elastic moduli gives us the possibility of estimating the strong fluctuation range of the order parameter.

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