INFLUENCE OF MECHANICAL STRESS ON THE BIREFRINGENCE PROPERTIES OF $(N(CH_3)_4)_2ZnCl_4$ CRYSTALS

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The influence of applied mechanical stress on the birefringence of $(N(CH_3)_4)_2ZnCl_4$ crystal in the temperature region of its incommensurate phase was investigated using Senarmonth's method ($\lambda = 633$ nm). The value of pinning force for the samples with different defectiveness was experimentally determined.

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As it is known, the interaction of incommensurate (IC) structure with defects causes some essential properties of IC-phases. The temperature hysteresis of IC modulation wave vector [1], dielectric permittivity [2] and linear birefringence [3] may be related to such properties. The occurrence of temperature hysteresis is an evidence of the presence of some mechanisms preventing the processes of solitons creation and annihilation [1, 2]. One of such mechanisms may be soliton pinning on impurities, vacancies, dislocations and so on. Experimental studies of impure crystals confirm the above-mentioned mechanism of temperature hysteresis [4]. Irregular character of the pinning force acting on solitons breaks a regular distribution of solitons in the vicinity of T_c, and brings the solitons to metastable chaotic state. Apart from the pinning force $\boldsymbol{F}_{\text{pin}}$, the solitons are influenced by the elastic coupling force $\boldsymbol{F}_{\text{el}}$ acting between solitons and, in the case of a crystal sample in an external field, by the applied force $\boldsymbol{F}_{\text{appl}}$. Therefore, the total force $\boldsymbol{F} = \boldsymbol{F}_{\text{pin}} + \boldsymbol{F}_{\text{el}} + \boldsymbol{F}_{\text{appl}}$ should be irregularly distributed in the sample volume.

The aim of this paper is to study the influence of the applied mechanical stresses on soliton–defect interaction and an experimental determination of the pinning force $\boldsymbol{F}_{\text{pin}}$ value. Ferroelectric $(N(CH_3)_4)_2$ $ZnCl_4$ crystals were chosen as the objects of our investigation, since a smooth variation of IC-parameter δ is characteristic of the IC-phase of these crystals at the temperatures $T_c = 280$ K $\div$ $T_i = 296.6$ K. The samples were grown using a slow evaporation method at room temperature from aqueous solution of $ZnCl_2$ and $N(CH_3)_4Cl$ salts taken in stoichiometric ratio. The influence of mechanical stresses on soliton–defects interaction was studied from the measurements of the linear birefringence increment dependences by Senarmonth's method ($\lambda = 633$ nm). The axial mechanical stress was applied along the a axis, since the spontaneous polarization P_a in the ferroelectric phase is suppressed by σ_a ($T_1 = 276.3$ K $\div$ $T_c = 280$ K) [5]. The light propagates along the modulation axis c.

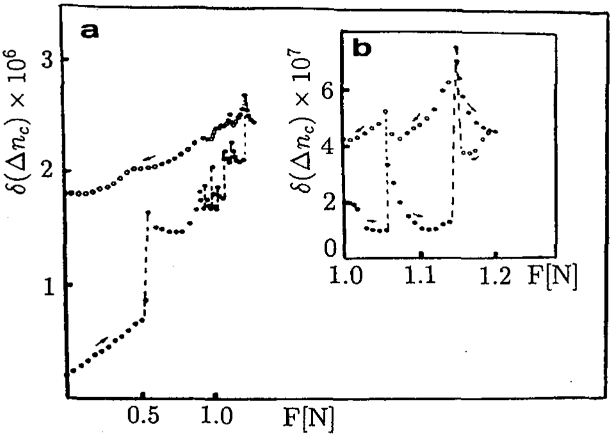

Fig. 1. The dependence of birefringence increment for the c-plate of $(N(CH_3)_4)_2ZnCl_4$ sample on the value of F_a applied along the a axis at $T = 288$ K (• — F_a increasing regime, o — F_a decreasing regime) (a) and the parts of corresponding curves in enlarged scale (b).

The experimental results are presented in Figs. 1–3. As it is shown in Fig. 1 for non-irradiated sample the $\delta(\Delta n)_c$ value increases at first. Then the $\delta(\Delta n)_c = f(F)$ dependence is characterized by a sequence of peaks which are similar to the ones in the temperature dependence $\delta(\Delta n)_i = f(T)$ at a low rate of temperature changes ($\partial T/\partial t = 60$ mK/h) [6]. The so-called "viscous" interaction manifests itself if the velocities of the IC-structure motion and defect diffusion become close and the optimum conditions for soliton–defect interaction arise [6]. The force–velocity dependence describing soliton motion [7] acquires a hysteresis character at "viscous" interaction.

According to the data of Ref. [8], the applied mechanical stress changes the soliton density x_0/d_0, where x_0 is the intersoliton distance, d_0 — the soliton width. It leads to changes in the soliton–soliton interaction energy $E_{ss} = a\exp(-\pi x_0/d_0)$ and the soliton–defect interaction energy $U_b = n_d x_0 E_b$ [9], where n_d denotes the defect concentration, E_b — the defect contact energy. In the presence of defects the soliton pinning will be observed at the condition $n_d x_0 E_b \approx a\exp(-\pi x_0/d_0)$. Therefore, the application and the release of mechanical stress will be accompanied by a hysteresis of $\delta(\Delta n)_c$ value and $\delta(\Delta n)_c = f(F)$ peaks positions. Hysteresis increases and peaks assymetry becomes more pronounced with increasing defectiveness (Figs. 1b, 2b, 3b).

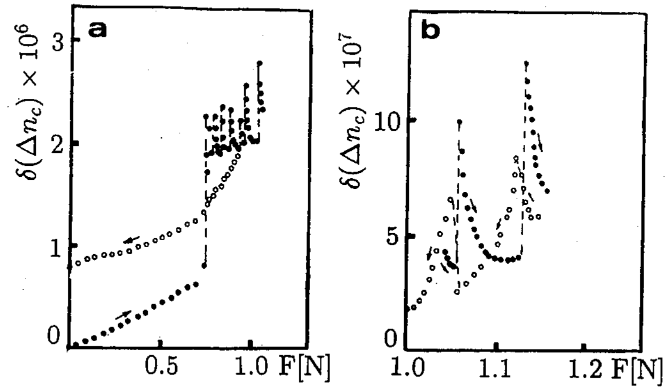

Fig. 2. The birefringence increment $\delta(\Delta n_c)$ dependence on F_a value at $T = 288$ K for $(N(CH_3)_4)_2ZnCl_4$ sample X-ray-irradiated (Mo-tube, $I = 13$ mA, $U = 40$ kV) during 2 hours (• — F_a increasing regime, ο — F_a decreasing regime) (a) and the parts of corresponding curves in enlarged scale (b).

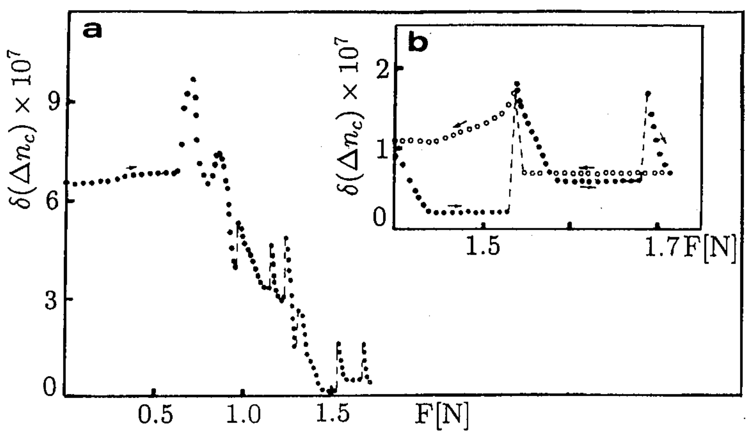

Fig. 3. The birefringence increment $\delta(\Delta n_c)$ dependence on F_a value (F_a increasing regime) at $T = 288$ K for $(N(CH_3)_4)_2$ $ZnCl_4$ sample in which the exponential distribution of defects due to X-ray-irradiation is created along the c axis (a) and the $\sigma(\Delta n_c)$ dependences on F_a value at 288 K for the same $(N(CH_3)_4)_2ZnCl_4$ sample (• — F_a increasing regime, ο — F_a decreasing regime) in enlarged scale (b).

On the basis of the above result the pinning force value may be estimated as $F_{\text{pin}} = F_{\text{k}}/n$, where n is the soliton density, F_{k} — the coercitive force. The $2F_{\text{k}}$ value is determined as the difference between the F_{appl} values for the corresponding peaks obtained on application and release of external stress (see Figs. 1b, 2b, 3b). The results $F_{\text{pin}} = 1.2 \times 10^{-8}$, 4×10^{-8}, 21×10^{-8} N are obtained for the Figs. 1b, 2b, 3b, respectively. It is obvious that an increase in the sample defectiveness increases the value of the pinning force. The values F_{pin} found in this paper are in agreement with those obtained in [7] equal to 10^{-8} N for $Ba_2Na \cdot Nb_5O_{15}$ crystals.

Thus, an axial mechanical stress σ_a leads to changes in the soliton density and, therefore, to changes in the conditions of soliton–defect interaction. The results obtained in this paper illustrate a set of hysteresis force–velocity dependences for description of soliton motion under mechanical field influence. Numerical values of F_{pin} are in agreement with the theoretical estimation [7]. We suppose analogous behaviour for other IC crystals.

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