Field anomaly of magnetic linear birefringence in magnetoelectric $LiCoPO_4$

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Abstract

Investigations of the magnetic field induced linear and circular birefringence in the antiferromagnetic magnetoeleclric crystal $LiCoPO_4$ were performed. A step-like change at H~20 kOe accompanied by magnetic hysteresis possessing high-field long tail was revealed. The behaviour of induced birefringence points to the magnetic field induced spin-reorientation phase transition to a non-collinear incommensurate structure.

Key words: magnetoelectric, antiferromagnetic, birefringence

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Introduction

Interest to the antiferromagnetic (AFM) magnetoelectric crystals of $LiCoPO_4$ is related to its magnetoelectric properties. Among known magnetoelectric materials including even materials with rare-earth elements the $LiCoPO_4$ crystal has exceedingly high value of the linear magnetoelectric effect [1]. $LiCoPO_4$ belongs to the olivine family of orthorhombic antiferromagnets and is isostructural with other members of this family, e.g., $LiNiPO_4$ and $LiMnPO_4$. At room temperature the space group of this crystal is Pnma. At T_N =21.9 K the crystal undergoes the paramagnetic-antiferromagnetic phase transition. The earlier neutron diffraction investigations [2] have indicated that at $T<T_N$ the $LiCoPO_4$ crystal is two-sublattice collinear AFM with coinciding magnetic and chemical cells and with the sublattice magnetic moments aligned along the ***b***-axis. According to [2] the Shubnikov group of this crystal is Sh_{62}^{445} = Pnma'. The important fact is that for obtaining a single AFM domain state required for an investigation of the magnetoelectric effect it is sufficient to performing in magnetic field only (without electric field) [1]. Moreover, in studies of the magnetoelectric effect a magnetic hysteresis of the "butterfly loop" type was observed near T_N. Such a loop is the signature of presence of a spontaneous magnetic moment, but the magnetic point symmetry mmm′ forbids the weak ferromagnetism. These experimental results still need conclusive interpretation.

The aim of the present work is to investigate behavior of magneto-optical properties of this magnetoelectric crystal. Here the behavior of the linear birefringence in magnetic field applied along the ***b***-axis is presented.

Experimental results and discussion

The setup for investigations of magneto-optic properties includes the monochromator, the photo-elastic modulator with the frequency 18 kHz and the lock-in amplifier. The given setup allows for performing investigations of magnetic linear and magnetic circular birefringence in the spectral region from 250 to 850 nm. Sample's temperature can be changed from 7 to 290 K, and intensity of magnetic field can reach 70 kOe. Magnetic field as well as the light propagation vector has been directed along the crystallographic ***b***-axis parallel to AFM vector. The sample of the $LiCoPO_4$ crystal has a right-angled parallelepiped shape of dimensions 1.22, 0.96, and 1.76 mm along ***a***, ***b*** and ***c*** axes (***a*** = 1.02, ***b*** = 0.59, ***c*** = 0.47 nm), respectively. The measurements were performed at the light wavelength λ = 610 nm. This wavelength was chosen reasoning from the transparency window of the sample and the desirability of the best sensitivity to changes of the phase shift between the polarized light modes propagating in the sample.

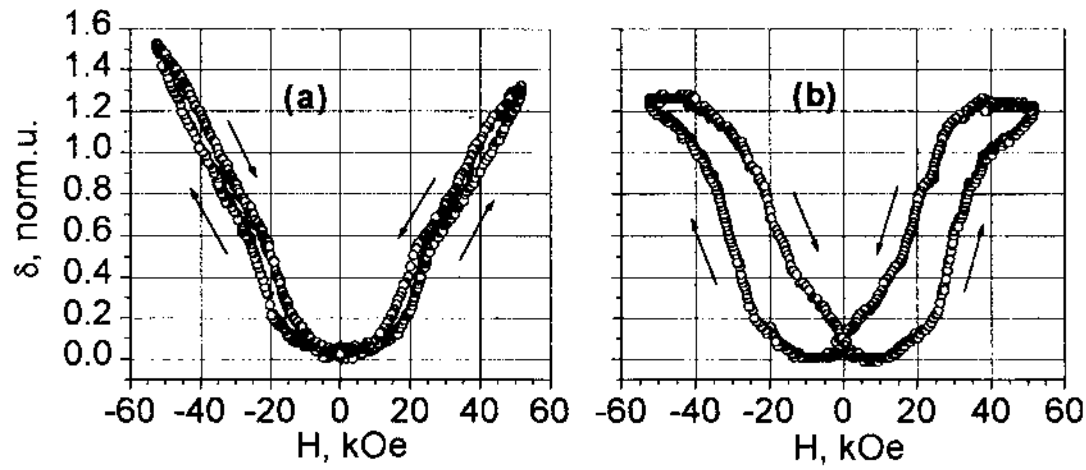

Fig.1 Magnetic field dependence of the phase shift corresponding to magnetic linear birefringence of light in $LiCoPO_4$ at T=15K (a), and T~T_N - 1K (b). The phase shift is shown in units normalized on the spontaneous change of the shift that occurred at the AFM ordering when temperature varied from T_N to 4.2K [3].

The figures 1 (a,b) show dependencies on magnetic field of that part of phase shift δ between the elliptically polarized light modes propagated in the magnetized sample , which is related to the linear birefringence. The curve on figure 1 (a) has the square-law character. A step-like anomaly is distinctively visible at the threshold field (H_t) of about 20 kOe. The step-like anomaly is accompanied by hysteresis with high-field tail reaching far in the region of magnetic fields, much higher than the threshold field.

Width of the hysteresis loop increases, when temperature approaches the Neel temperature. This property is illustrated in the figure 1(b). The hysteresis dependence takes the characteristic form of a "butterfly-loop", and its tails reach as far as beforehand.

Figure 2 shows dependence on magnetic field of that part of phase shift ρ which corresponds to the effect of magnetic circular birefringence, that is Faraday rotation. The dependencies exhibits the characteristic linear behavior. Within the measurement accuracy no step-like changes are observed on dependencies of circular birefringence at H_t.

The peculiarities of the magnetic field behavior of the linear birefringence observed in $LiCoPO_4$ are (i) high value of the induced birefringence comparing to the spontaneous magnetic birefringence and (ii) its step-like change in the relatively low fields that is accompanied by the peculiar hysteresis. So, the

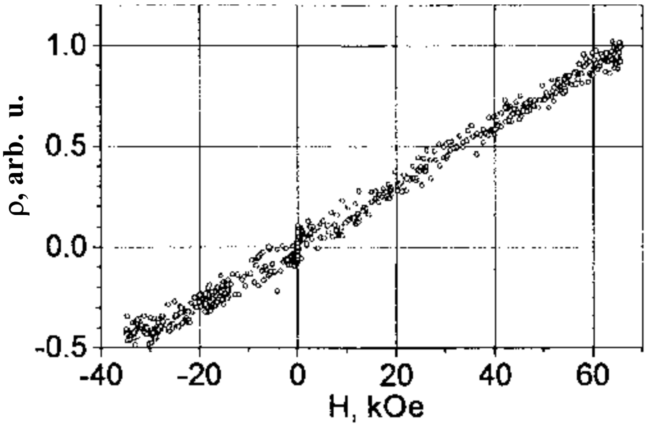

Fig.2 Magnetic field dependence of the phase shift corresponding to magnetic circular birefringence of light in $LiCoPO_4$ at T = 15 K. The phase shift is shown in arbitrary units.

change of birefringence equal to the spontaneous is observed in the field near 40 kOe, much lower than effective exchange field. These changes cannot be explained by possible small changes of the sublattice moments that is due to magnetic field at temperature different from T = 0K or is caused by mixing of quantum-mechanical states of Co^{2+} ions.

Observed peculiarities cannot be explained in the context of knowledge of the original collinear magnetic structure of $LiCoPO_4$. Behavior of birefringence may be clarified only assuming the presence of the spontaneous weakly non-collinear magnetic structure in $LiCoPO_4$. At the field H_t a spin-reorientation must take place that gives rise to a very weak increase of perpendicular to field components of magnetic moments. Characteristic hysteresis that extends to the high fields $H \gg H_t$ and its kinetic peculiarities let assume that the transition at H_t may be transition to the incommensurate non-collinear magnetic structure.

Conclusion.

The experimental data presented here leads to the conclusion that the structure of the crystals is more complicated than it was assumed in the earlier studies. It is more likely that spontaneous magnetic structure of $LiCoPO_4$ is weakly non-collinear, and in b-oriented magnetic field it transforms to a non-collinear incommensurate structure. It is possible that in close proximity to the Neel temperature the incommensurate phase is formed spontaneously. These inferences resulting from magnetooptical data are in accordance with the theoretical conclusions that in crystal $LiCoPO_4$ may exist a modulated non-collinear magnetic structure described by the Lifshitz-like invariants [4].

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