Weak ferromagnetism in magnetoelectrics $LiCoPO_4$ and $LiNiPO_4$

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Abstract

The carried out with using of a SQUID-magnetometer the magnetic researches of the antiferromagnetic magnetoelectric $LiCoPO_4$ and $LiNiPO_4$ monocrystals have shown that these compounds exhibited a weak ferromagnetism below the Neel temperature T_N(22 K and 20.8 K). The weak ferromagnetic moments (WFM) is directed along its antiferromagnetic axis in both crystals, but their temperature dependencies are different. Presence WFM in these magnetoelectrics of known by its anomalous magnetoelectric properties bring up a problem of redetermination of the magnetic structures of these antiferromagnetic crystals as their magnetic symmetry determined earlier by the neutron-diffraction studies, correspond to the compensated antiferromagnetism.

Key words: weak ferromagnetism; magnetoelectric; antiferromagnetic; $LiCoPO_4$; $LiCoPO_4$

The lithium orthophosphates is the well-known magnetoelectric crystals of the family of orthorhombic antiferromagnets with the olivine structure and having the general formula $LiMPO_4$ (where M = Fe^{2+}, Mn^{2+}, Co^{2+}, Ni^{2+}). They have been attracting attention because have unusual behavior of the magnetoelectric effect in a magnetic field and as yet unexplainedly [1,2]. The crystal structure of $LiCoPO_4$ and $LiNiPO_4$, like that of the other lithium phosphates of transition elements of the olivine family, has a symmetry described by the space group $Pnma$ (D^{16}_{2h}). In this structure the unit cell (a = 10.20 Å, b = 5.92 Å, c = 4.70 Å and a = 10.03 Å, b = 5.85 Å, c = 4.68 Å, respectively) contains four formula units, and the magnetic ions are crystallographically equivalent and occupy four c positions. The magnetic moments of the Co^{2+} ions are collinear and directed along the **b** axis and along the **c** axis for the Ni^{2+} ions, completely compensating each other [3]. The magnetic structure [2] of the crystal is described in a collinear four-sublattice model with the Shubnikov symmetry group Sh^{445}_{62} ($Pnma'$).

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The behavior of the magnetoelectric effect observed by the authors of Refs.[1,3] could be due to the existence of a weak ferromagnetic moment(WFM)in the crystal. However, previous studies of the magnetic properties of both polycrystalline and single-crystal $LiCoPO_4$ [2,3] and $LiNiPO_4$ [4] have not detected weak ferromagnetism.

On the top graph of figure 1 temperature dependencies of magnetization of LiCoPO4 crystal for different fields of cooling (FC) of the sample are submitted. On the middle graph of figure 1 moment M_0 calculated of dependencies of $M(T)/H$ for fields 10 and 0.5 kOe which represents temperature behavior WFM in this crystal along the axis antiferromagnetism is shown. In the bottom part of fig.1 the curve $M(H)/H$ is submitted at temperature 15 K on which are observed two jumps (vertical pointers) connected with reorientation WFM (M_0) in the applied magnetic field [5].

On Figure 2(a) shows the inverse magnetic susceptibility(M(T)/H) curve of the crystal $LiNiPO_4$ wich was obtained in the magnetic field 0.5 kOe directed along axis c. The inverse magnetic susceptibility shown in figure 2 is liner in the temperature range 50-300 K. Using the Curie-Weiss law $\chi(T) = C/(T + \theta)$ to fit to the mesured inverse susceptibility(solid line) yieds

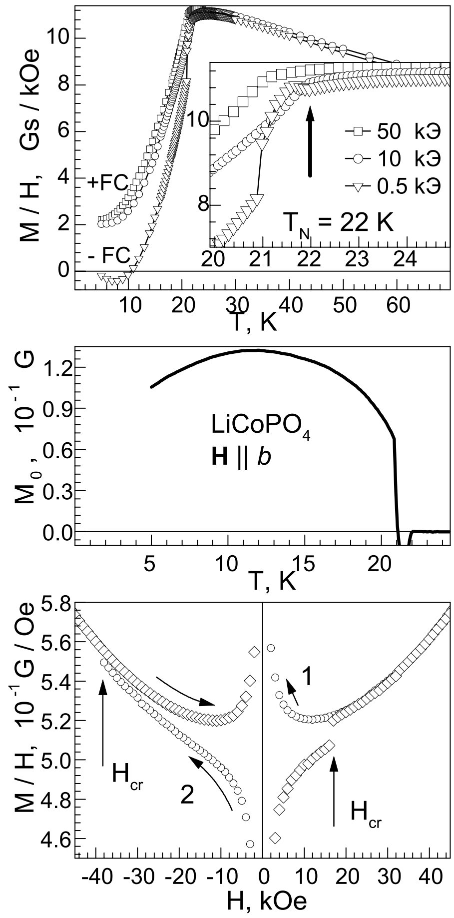

Fig. 1. a- Temperature dependence of the magnetization of the $LiCoPO_4$ crystal in an external magnetic field $\mathbf{H} \parallel \mathbf{b}$ with a field strength of 50,10 and 0.5 kOe ; b- temperature dependence of the WFM ($M_0(T)$) output curves M(T)for $H=10and0.5$ kOe; c- the $M(H)/H$ curves , obtained for a completely cycle of variation of the field from +5 to −5 T and back to +5 T at crystal temperature of 15 K.

an constant $C = 35\ KkOe/G$ and $\theta = -70K$. Figure 2(midle) shows the curves of the temperature dependence of the magnetization obtained in magnetic fields H of 50, 20, 10 and 0.5 kOe. On the curves is observed two critical point T_N and T_C. It can be testify about presence of a incommensurable phase in this crystal. Figure 2(down) shows the $M(H)/H$ curves for 19 K and 21.5 K, obtained for a completely cycle of variation of the field from +50 to −50 kOe and back to +50 kOe. The $M(H)/H$ curves are well approximated by the function $f(H) = M_0/H + \chi + \beta H^2$ (solid line). The presence of a nonzero spontaneous moment M_0 and the jumps of M_0 at H_{cr} (vertical pointers) is demonstrated by the $M(H)/H$ curves in Fig.2(c). The revealed hysteresis of the magnetization convincingly attests to the presence of a weak spontaneous magnetic moment(or WFM) in the $LiNiPO_4$.

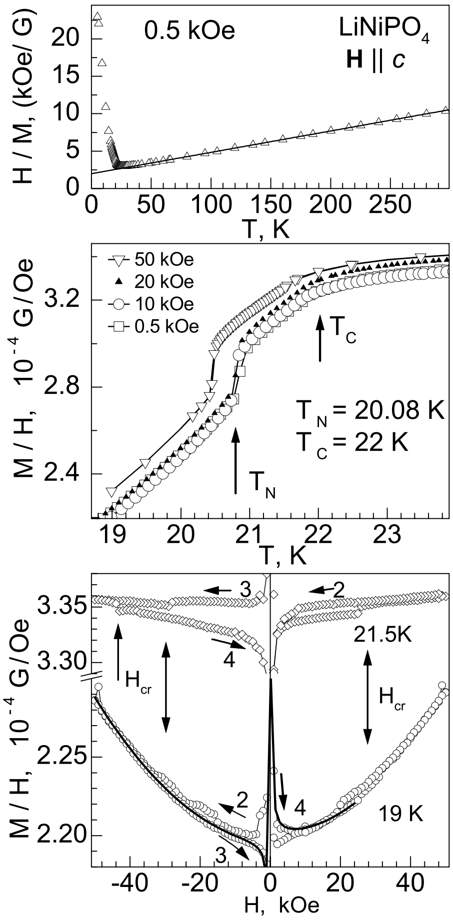

Fig. 2. a- Inverse magnetic susceptibility(M/H) versus temperature for $LiNiPO_4$ crystal in an external magnetic field $\mathbf{H} \parallel \mathbf{c}$ with a field strength of 0.5 kOe. The liner fit the data (solid line) in paramagnetic regim(50- 300 K), is the Curie-Weiss law; b- Temperature dependence of M/H in an external magnetic field $\mathbf{H} \parallel \mathbf{c}$ with a field strength of 50,20,10 and 0.5 kOe; c- the $M(H)/H$ curves at crystal temperature of 19 K and 21.5 K

We thank the organizing committee LT23 for financial support of this report.

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