

Peculiar magnetic susceptibility and phase transitions in LiNiPO₄

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Among orthorhombic olivines, considered for application as cathodes of Li-ion batteries, LiNiPO₄ shows a unique set of properties, among them: (i) two-step formation of a quasi-2D antiferromagnetic structure via the 2nd order transition to an incommensurate phase at 21.8 K and the 1st order transition to the commensurate antiferromagnetic phase at 20.9 K, (ii) complex exchange interactions, and (iii) strong linear magnetoelectric effect.

Detailed studies of specific heat, magnetization and magnetic torque of a LiNiPO₄, aimed at:

- seeking for a transition to a multiferroic state,
 - constructing the phase diagram in the magnetic field, B , up to 9 T,
 - verifying whether the complex exchange interactions, strong anisotropy, and the quasi 2D magnetic structure lead to any uncommon macroscopic magnetic properties
- will be presented.

The specific heat studies, supplemented by the "slope analysis method", allowed to find a splitting of the specific heat anomaly associated with the 1st order transition, suggesting that these are two coupled transitions, one of which can be the ferroelectric one. Measurements of the specific heat, C_p , of a single crystal as a function of temperature, T , for a series of fixed B values allowed to determine analytical equations describing the phase transition lines in the T - B plane, and to model evolution of a shape of the specific heat anomalies accompanying the phase transitions in a powder sample under influence of B .

By measurements of angular dependences of magnetic torque and magnetization for B rotating within the a - c and b - c crystalline planes, for many fixed T and B values, we found a new effect, that we called "off-diagonal nonlinear magnetic susceptibility", i.e., we found that for each of the main crystallographic axis (a , b , and c), an additional component of magnetic susceptibility, proportional to the square of the perpendicular to this axis component of B exists. A phenomenological model of this effect, describing the experimental results very well, was proposed.