Nonlinear off-diagonal magnetic susceptibility of LiNiPO₄

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Among orthorhombic (Pnma) olivines, suitable for cathodes of Li-ion batteries, LiNiPO₄ is a unique one, because it orders magnetically in two steps, i.e., at 21.8 K, the 2nd order transition to an incommensurate phase and then, at 20.9 K, the 1st order transition to an antiferromagnetic phase appear. We focused our studies on magnetic properties of the ordered phases and on the phase diagram. For a LiNiPO₄ single crystal, angular dependences of torque and magnetic moment were measured for magnetic field, B, rotating within a-c and b-c planes, for several |B| values, at several temperatures. We explained uncommon shapes of these dependences by assuming the magnetic moment induced along a, b, and c axes by B to be proportional not only to the B-component parallel to the particular axis but also to the square of the B-component perpendicular to it. Then, a very good agreement between the theoretical and the measured dependences was achieved, which confirmed validity of the model proposed. We called this effect the "nonlinear off-diagonal magnetic susceptibility". The temperatures of both transitions were found to decrease proportionally to B² for B parallel to c, and to be insensitive to B for B parallel to the a and b axes.