Relationship between ferroelectricity and magnetic structure of $PbCuSO_4(OH)_2$ with CuO_2 ribbon chains

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Measurements of magnetic and dielectric properties and neutron diffraction experiments have been studied for single- and poly-crystalline samples of PbCuSO4(OH)2 with quasi one-dimensional spin 1/2 chains formed of edge-sharing CuO4 square planes called CuO2 ribbon chains. The dielectric constant and polarization measurements indicate that a ferroelectric transition takes place simultaneously with a magnetic transition at the N
el temperature TN˜2.8 K (multiferroic) [1]. Based on the obtained results, a possible magnetic structure and relationship with the ferroelectricity are discussed.

For the systems with the CuO2 ribbon chains, due to the geometrical characteristic of the crystal structure, the nearest-neighbor exchange interaction J1 between spins is expected to be relatively weak and often ferromagnetic, and the second neighbor interaction J2 is antiferromagnetic. Under these situations, the non-trivial or helical ordering is often realized. LiVCuO4 is known as model systems of the CuO2 ribbon chains. LiVCuO4 has the helical magnetic structure, which induces ferroelectricity [2] as theoretically discussed in ref. 3. The quantum spin system can be considered as a new kind of multiferroics which may provide different aspects of physics from classical spin systems consisting of Fe3+ and Mn3+ with S > 1. In the present work, we have carried out experimental studies another CuO2 ribbon chain system PbCuSO4(OH)2 [4]. Neutron scattering measurements were carried out using the triple axis spectrometer HQR (T1-1) of the thermal guide installed at JRR-3 of JAEA in Tokai.

PbCuSO4(OH)2 has CuO2 ribbon chains along the b direction, whose structure is schematically shown in Fig. 1. a ’- and c ’- axes are defined to be suitable for the local structure of CuO2 ribbon chains. The present work has revealed the following features. (i) The evaluated |J2/J1| value from analyses of magnetic susceptibility(χ)?temperature(T) curves is ˜ 0.62 (J1= − 13 ± 3 K and J2=21 ± 5 K), which is theoretically expected to favor a helical type magnetic structure. (ii) Behavior of χ -T curves below TN indicates that the ordered spin components are almost parallel to the CuO2-plane (a ’b-plane). (iii) The spin flop transition at Hc˜2.5 T also indicates that the ordered spin components in the antiferromagnetic phase lie in the CuO2-plane (a ’b-plane). (iv) In the neutron diffraction measurements on a single crystal, 0 k ± δ 1/2 magnetic Bragg reflections with k=even, l=integer and δ ˜0.189 are observed below TN. Figure 2 shows the T-dependence of the peak intensities for 0 δ 1/2 reflection, indicating that the magnetic ordering grows with decreasing T below TN. Considering these results, we think that the magnetic structure of PbCuSO4(OH)2 is a kind of helical type structure with large components parallel to the CuO2-plane shown in the inset of Fig. 2. The helical axis e3 is parallel to the c ’ axis and the modulation vector Q of the incommensurate magnetic structure is parallel to the b axis (CuO2 ribbon chain). (v) The ferroelectric polarization P is observed along a ’. For PbCuSO4(OH)2, obtained results indicate that the equation P = Q × e3 holds, as was proposed by microscopic theories [3] where the ferroelectricity is originated from the helical type magnetic structure through the spin-orbit interaction.

References

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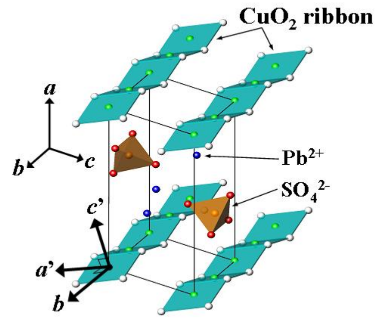

Fig. 1: Crystal structure of $PbCuSO_4(OH)_2$. The *a*'- and *c*'- axes are defined to be suitable for the local structure of the CuO_2 ribbon chains.

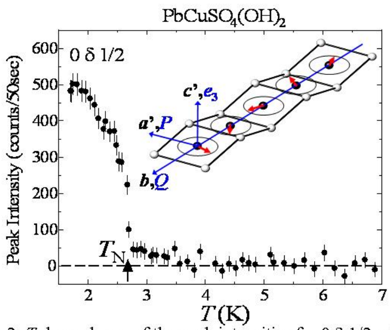

Fig. 2: *T*-dependence of the peak intensities for 0 δ 1/2 reflection (δ~0.189) for $PbCuSO_4(OH)_2$. The inset show the magnetic structure of the Cu^{2+} spins.

Fig. 1.