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## Neutron diffraction study of double tungstates $\text{Li}_2\text{M}^{II}(\text{WO}_4)_2$ (M=Co and Ni)

Neutron diffraction study of double tungstates  $\text{Li}_2\text{M}^{II}(\text{WO}_4)_2$  (M=Co and Ni)

The isostructural  $\text{Li}_2\text{Co}(\text{WO}_4)_2$  and  $\text{Li}_2\text{Ni}(\text{WO}_4)_2$  crystallize in the triclinic structure (space group P-1, Z = 1). The crystal structures can be viewed as alternating layers of WO layers and Li-MO layers along the b-axis. The WO layers built up of edge-sharing  $\text{WO}_6$  octahedra forming infinite zigzag chains along the a-axis as in many other tungstates. The edge-sharing  $\text{LiO}_6$  octahedrons also form infinite chains extending along the c-axis. The divalent ions,  $\text{Co}^{2+}$  and  $\text{Ni}^{2+}$ , are locating in the octahedral oxide environment, in the mesh forming by crossing LiO and WO chains, isolated from each other. Due to the lack of direct connection of the  $\text{MO}_6$  octahedral, the magnetic coupling between the nearest neighboring M cannot through the strong M-O-M superexchange path (SE). Nevertheless the magnetic coupling can be achieved through more complex pathways, which is bridging by the oxygen ions in  $\text{LiO}_6$  or  $\text{WO}_6$ . In  $\text{Li}_2\text{M}^{II}(\text{WO}_4)_2$  the weak M-O-O-M pathways, known as super-superexchange (SSE) form complex network with magnetic frustrations, gives raise to complex magnetic behaviors.

The temperature dependent magnetic susceptibility and heat capacity clearly mark the two magnetic transition temperatures of each of the two compounds. In  $\text{Li}_2\text{Ni}(\text{WO}_4)_2$  the two magnetic transition temperatures are  $\text{TN}_1 = 12.8$  K and  $\text{TN}_2 = 17.5$  K, whereas the slightly lower  $\text{TN}_1 = 7.2$  K and  $\text{TN}_2 = 9.5$  K for  $\text{Li}_2\text{Co}(\text{WO}_4)_2$ . The magnetic structures have been determined from neutron powder diffraction measurement conducted on Wombat and Echidna at ANSTO. For both compounds the magnetic correlation develops below  $\text{TN}_1$ , as an incommensurate modulation and the magnetic moments order into commensurate super cells below  $\text{TN}_2$ . Both compounds possess collinear antiferromagnetic spin configurations in the magnetic order phases. Surprisingly the magnetic unit cells of the two isostructural compounds are different. The magnetic unit cell of  $\text{Li}_2\text{Ni}(\text{WO}_4)_2$  is (2a b 2c), and the magnetic moment of  $\text{Ni}^{2+}$  at 3 K is  $1.925 \mu\text{B}$ , roughly pointing along the a direction. Whereas a much larger magnetic unit cell of (2a 4b 4c) is realized in  $\text{Li}_2\text{Co}(\text{WO}_4)_2$  below  $\text{TN}_2$ . Co spins in the {+ + -} sequence are observed when moving to the neighboring magnetic sites along b and c-axis. The magnetic moment of  $\text{Co}^{2+}$  is  $2.836 \mu\text{B}$  at 5.5 K closely align along the a-axis as well. The different magnetic ground states in the two compounds might be resulted from to the slight differences in the crystal structure and magnetic couplings.

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