Layered packing and two-dimensional magnetism in honeycomb-type mixed oxides

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Quasi-two-dimensional magnetism is one of the most enthralling topics of a modern solid state physics. Reduced dimension gives rise to plenty of new phenomena. One of the most intriguing case for 2D lattices is a hexagonal net of antiferromagnetically ordered spins. Minimal possible for 2D lattices coordination number (z=3), frustrated interactions of nearest neighbors with second and third neighboring spins and also a quantum fluctuations leads to a large variety of a possible ground states [1]. Fine examples of realization for such structures are mixed honeycomb oxides. Such compounds are described with chemical formulas: A2M2TeO6 and A3M2XO6 where A – is alkali, M – 3d-metal and X is for Sb or Bi cations. Crystal structure of these compounds is formed by alternating layers of Te/Sb/Bi and M oxygen octahedra forming a honeycomb ordering and alkali cations with different type of oxygen surrounding depending on a structural politype. So magnetic ground state in these compounds depend on the nature of cations, their spin, orbital and electronic states determined by the local environment and also by a type of superstructure ordering due to a complexity of in- and interlayer exchange interactions. Still no adequate description between crystal structure and magnetic properties for such compounds was established, so the aim of a present work was to investigate crystal and magnetic structures of Li3Ni2SbO6 and Na2Ni2TeO6 compounds. For Li3Ni2SbO6 synchrotron diffraction experiment revealed a peak splitting that finally allowed to identify true space group to be C2/m; additional diffuse scattering indicating stacking faults presence was also detected. Neutron diffraction pattern at RT for Na2Ni2TeO6 revealed an anisotropic peak broadening indirectly specifying a sample to be a possible mixture of a P6322 and P63/mcm politypes. Close to 90o values of Ni-O-Ni bond angles shows presence of weak ferromagnetic inlayer interactions according to Goodenough-Kanamori rules; distorted O-Ni-O bond angles indicate a trigonal crystal field presence at Ni sites. LT neutron powder diffraction revealed addition peaks associated with magnetic scattering appearing at temperatures below 15 and 27 K for Li3Ni2SbO6 (on the left pic.) and Na2Ni2TeO6 (on the right pic.) respectively. Magnetic structures for both compounds are determined to be a zig-zag ferromagnetic chains coupled antiferromagnetically in ab-plane. Propagation vectors are k=(1/2 1/2 0) for Li3Ni2SbO6 and k=(1/2 0 0) for Na2Ni2TeO6. For Li3Ni2SbO6 ferromagnetic coupling for chains from adjacent layers was found, revealing non-negligible interlayer interactions. With temperature decreasing Ni spins directed along c-axis, demonstrates a certain tilt aligning perpendicular to ab-plane at T = 1.5 K. For Na2Ni2TeO6 increased ionic radius of alkali cation leads to a suppression of interlayer interactions. Ferromagnetic chains are coupled antiferromagnetically, nevertheless magnetic moments exhibit an inclination indicating a presence of a small ferromagnetic component within interlayer interactions.


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