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

Stanislav Podchezertsev\textsuperscript{1}, Artem Korshunov\textsuperscript{1}, Alexander Malyshev\textsuperscript{1}, Alexander Kurbakov\textsuperscript{1}

\textsuperscript{1}Petersburg Nuclear Physics Institute, NRC Kurchatov Institute, Gatchina, Russian Federation
E-mail: stpcz@mail.ru

Quasi-two-dimensional magnetism is one of the most enthralling topics of modern solid state physics. Reduced dimension gives rise to plenty of new phenomena. One of the most intriguing cases 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 possible ground states [1]. Fine examples of realization for such structures are mixed honeycomb oxides. Such compounds are described with chemical formulas: \(\text{A}_2\text{M}_2\text{TeO}_6\) and \(\text{A}_3\text{M}_2\text{XO}_6\) where \(\text{A} - \) is alkali, \(\text{M} - 3d\)-metal and \(\text{X} \) 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 \(\text{Li}_3\text{Ni}_2\text{SbO}_6\) and \(\text{Na}_2\text{Ni}_2\text{TeO}_6\) compounds. For \(\text{Li}_3\text{Ni}_2\text{SbO}_6\) synchrotron diffraction experiment revealed a peak splitting that finally allowed to identify true space group to be \(\text{C}2/m\); additional diffuse scattering indicating stacking faults presence was also detected. Neutron diffraction pattern at RT for \(\text{Na}_2\text{Ni}_2\text{TeO}_6\) revealed an anisotropic peak broadening indirectly specifying a sample to be a possible mixture of a \(\text{P}6\text{3}2\text{2}\) and \(\text{P}6\text{3/mcm}\) politypes. Close to 90\(^\circ\) values of Ni-O-Ni bond angles shows presence of weak ferromagnetic interlayer interactions according 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 \(\text{Li}_3\text{Ni}_2\text{SbO}_6\) (on the left pic.) and \(\text{Na}_2\text{Ni}_2\text{TeO}_6\) (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 \(\text{k}=(1/2 1/2 0)\) for \(\text{Li}_3\text{Ni}_2\text{SbO}_6\) and \(\text{k}=(1/2 0 0)\) for \(\text{Na}_2\text{Ni}_2\text{TeO}_6\). For \(\text{Li}_3\text{Ni}_2\text{SbO}_6\) 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 \(\text{Na}_2\text{Ni}_2\text{TeO}_6\) 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.


Keywords: magnetic structure, oxides, Rietveld refinement