Structure and Magnetic Properties of Cu3Ni2SbO6 and Cu3Co2SbO6 Delafossites with Honeycomb Lattices

The crystal structures of two Delafossites, Cu3Ni2SbO6 and Cu3Co2SbO6, are determined by high-resolution synchrotron powder X-ray diffraction. The Ni and Co are ordered with respect to Sb in the layer of edge sharing octahedra, forming magnetic layers with honeycomb geometry. High-resolution electron microscopy confirms ordering, and selected-area electron diffraction patterns identify examples of the stacking polytypes. Low temperature synthetic treatments result in disordered stacking of the layers, but heating just below their melting points results in nearly fully ordered stacking variants. The major variant in both cases is a monoclinic distortion of a 6-layer Delafossite polytype, but a significant amount of a 2-layer polytype is also present for the Ni case. The antiferromagnetic ordering with transitions, at 22.3 and 18.5 K for Ni and Co variants, respectively, is investigated by temperature and field dependent magnetization, as well as specific heat. The sharp magnetic transitions support the presence of well developed 2:1 ordering of the Co:Sb or Ni:Sb ions in the honeycomb layers. Neutron diffraction measurements at 4 K are used to determine the magnetic structures. For both the Ni and Co phases, the propagation vector is \( k = [100] \), and can be described as alternating ferromagnetic chains in the metal-oxide plane giving an overall antiferromagnetic “zigzag” alignment. While orientation of the magnetic moments of the Co is along the b-axis, the Ni moments are in the ac plane, approximately parallel to the stacking direction. Bulk magnetization properties are discussed in terms of their magnetic structures.