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Structure, magnetic and magnetoelectric properties of Ca(Co,Mn)Ge2O6 pyroxenes

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Compounds belonging to the Pyroxene family are well known as rock-forming minerals, and have thus drawn substantial interest by mineralogists. In this family of general chemical formula AM(Si, Ge)2O6, A is usually an alkali metal monovalent cation or a divalent alkaline earth cation, and B may be a trivalent or divalent transition metal cation. Among pyroxene compounds, the monoclinic clinopyroxenes are characterized by isolated one-dimensional chains of MO6 octahedra linked by edge-sharing. Due to this specific arrangement, clinopyroxene compounds where M is a magnetic transition metal cation have attracted considerable attention in recent years. Investigations revealed that these compounds present a rich diversity of intriguing low-dimensional magnetic properties. The existence and possible interplay of low dimensionality and magnetic frustration results in multiferroic and/or magneto- electric (ME) properties. We have undertaken the study of the CaCo1-xMnxGe2O6 (0<x<1) solid solution to investigate the effect of the Co/Mn substitution on the magnetic and ME properties by means of neutron diffraction, magnetic and magneto-electric measurements. In CaCoGe2O6, strong FM interactions within the M chains dominate the AFM coupling between the chains. In contrary, the magnetic structure of CaMnGe2O6 is made of AFM chains coupled ferromagnetically. These two commensurate magnetic structures adopt two different magnetic point group that allows for linear (for Mn) and bilinear (for Co) magnetoelectric effect. For the Co/Mn solid solution, a competition exists between FM and AFM coupling within and between the chains. This results in the apparition of a spin glass state above the long range magnetic transition and for certain composition in a magnetic structure described by two propagation vectors. We will present detailed investigation of the relationship between structure, magnetic structure and ME properties for this rich series of ME compounds.

Keywords: magnetoelectric, pyroxene, magnetic structure