Unusual magnetic properties of layered $M\mathrm{CrS}_2$ compounds

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Abstract

A chromium sulfide AgCrS_2 has recently attracted considerable attention due to its multiferroic properties and unusual double-stripe magnetic order which appear below the Néel temperature of 50 K. AgCrS_2 belongs to a family of layered $M\mathrm{CrS}_2$ (M^+=Li, Na, K, Cu, Ag, Au) compounds in which magnetic Cr^{3+} ($3d^3$) ions form a triangular lattice. Although charge and orbital degrees of freedom in these compounds are frozen, geometrical frustrations of magnetic interactions, inherent in the triangular lattice, lead to fascinating magnetic properties: depending on the size of an M^+ ion magnetic order in Cr planes changes from non-collinear 120° (Li) antiferromagnetic, to double stripes (Ag, Au), and, finally, to ferromagnetic (K). In order to understand this strong variation of the magnetic properties we calculated band structures and total energies $E(\mathbf{q})$ as a function of a wave vector $\mathbf{q}$ for various spin spiral structures. Effective exchange coupling constants j between Cr spins are then estimated by fitting $E(\mathbf{q})$ to an appropriate classical Heisenberg model. We found that depending on the M size the nearest neighbor coupling j_1 changes from strongly antiferromagnetic in LiCrS_2 to ferromagnetic in KCrS_2, whereas the coupling j_3 between the 3-rd Cr neighbors is strong and remains nearly constant in all the compounds. In AgCrS_2 with $j_1 \ll j_3$ the double stripe magnetic order is stabilized by monoclinic distortions of the crystal structure. We discuss the microscopic origin of various j and show that similar considerations help to understand the magnetic properties of other frustrated Cr compounds $A\mathrm{Cr}_2\mathrm{S}_4$ with a spinel structure.