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Hollandites: a novel class of oxides with unusual properties

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Hollandite with a general chemical formula $A_xM_8O_{16}$ ($x \leq 2$) is one of minerals. Its crystal structure consists of the M_8O_{16}-framework and A-cations at tunnel sites in the M_8O_{16}-framework. The most popular crystal symmetry is a tetragonal $I4/m$ in which the crystallographycally independent M site is unique, hence, $A_2^+M_8O_{16}$ is a mixed valent oxide with M^{3+}/M^{4+} = 1/3. We have synthesized $K_2V_8O_{16}$ [1] and $K_2Cr_8O_{16}$ [2] in both powder and single crystal form under high pressure and found metal-insulator (MI) transitions. The manner of MI transitions, however, is very different between both compounds. $K_2V_8O_{16}$ exhibits a first order MI transition at around 170 K. The magnetic susceptibility is reduced to a small value at the transition, suggesting the formation of V^{4+}-V^{4+} singlet pairs and V^{3+}-V^{3+}pairs in the low temperature insulator phase. The transition is accompanied by the structural change from a tetragonal to a monoclinic structure. The low temperature phase has a superlattice of $\sqrt{2}$ x $\sqrt{2}$ x 2, suggesting a charge ordering of V^{4+} and V^{3+}. On the other hand, $K_2Cr_8O_{16}$ is a ferromagnetic metal (or half-metal) with T_C = 180 K and shows a transition to an insulator at 95 K without any apparent structural change but retaining ferromagnetism. $K_2Cr_8O_{16}$ is quite unique in three aspects: It has a rare mixed valence of Cr^{3+} and Cr^{4+}; it has a metal (or half-metal) to insulator transition in a ferromagnetic state; and the resulting low temperature phase is a rare case of a ferromagnetic insulator. This discovery could open a new frontier on the relation of magnetism and conducting properties in strongly correlated electron systems.

References:

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