3.5 Spin-Orbital Ordering and Mesoscopic Phase Separation in Ca_2FeReO_6

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Novel phenomena caused by electrons localized in degenerate orbital states have been the central issue in the physics of transition metal oxides for the last few years. For e_g orbital systems, the degeneracy under cubic crystalline fields is often removed by large Jahn-Teller distortions of the oxygen octahedra. The physics of electrons localized in partly occupied t_{2g} orbitals is quite different, due to the relative weakness of the Jahn-Teller coupling, higher degeneracy, and additional symmetry of t_{2g} orbitals. In addition, these electrons may show unquenched orbital magnetic moments, and spin-orbit coupling usually plays an important role.

The double perovskite compounds A_2Fe(Mo,Re)O_6 (A = Ca, Sr, or Ba) have recently attracted much scientific and technological interest after the discovery of large tunneling magnetoresistance at room-temperature in most cases[1,2], which has been ascribed to half-metallic electronic band structures[1]. An intriguing exception is the insulating behavior found in the compound Ca_2FeReO_6 (CFRO)[3], revealing that the (Fe,Re)-based double perovskites are at the border of a metal-insulator transition[2]. Mössbauer studies in CFRO indicate the presence of Fe^{3+} ions[2], thus the oxidation state of Re ions is expected to be 5+, with 2 electrons in the *5d* t_{2g} orbitals. In order to search for possible manifestations of the Re t_{2g} orbital degree of freedom in this insulating compound, the nuclear and magnetic structures were investigated by neutron powder diffraction[4]. A number of remarkable observations were made: (i) mesoscopic phase separations, with coexisting monoclinic crystal structures below ~300 K and large changes in the phase proportions below ~160 K; (ii) magnetic-field control of the volume fractions in the phase-separated state at 100 K, with a suppression of the additional phases observed below ~300 K for an applied field of a few Tesla; (iii) a ferrimagnetic arrangement of Fe and Re magnetic moments below T_C = 521 K, with strong evidence for distinct moment directions for the competing phases; (iv) anomalous lattice parameter behavior below T_C and (v) a new Bragg peak below T_s, which is forbidden by the symmetry considered to model the nuclear structure. These results indicate that the Re^{5+} t_{2g}^2 electrons are localized on an atomic scale, and CFRO is a Mott insulator. The mesoscopic phase separations observed for this compound are ascribed to a competition between distinct spin- and orbitally-ordered insulating states.

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