International Research Center for Elements Science - Advanced Solid State Chemistry -

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(D Sc)

Scope of Research

Novel inorganic crystalline materials that have new, useful or exotic features such as magnetoresistance, ferromagnetism and quantum spin ground state are synthesized. High pressure conditions are applied to obtain nonequilibrium materials with crystal structure and/or chemical composition unavailable at ambient pressures. Precise crystal structure analysis using X-ray or neutron beams as well as magnetic and electronic property measurements are performed to reveal and understand the physical properties of newly obtained materials.

Research Activities (Year 2007)

Publications

Saito T, Williams A, Attfield J P, Wuernisha T, Kamiyama T, Ishiwata S, Takeda Y, Shimakawa Y, Takano M: Neutron Diffraction Study of a Layered Cobalt Oxide $SrCo_6O_{11}$, *J. Mag. Mag. Mater.*, **310**, 1584-1586 (2007).

Ishiwata S, Terasaki I, Ishii F, Nagaosa N, Mukuda H, Kitaoka Y, Saito T, Takano M: Two-Staged Magnetoresistance Driven by the Ising-Like Spin Sublattice in $SrCo_6O_{11}$, *Phys. Rev. Lett.*, **98**, [217201-1]-[217201-4] (2007).

Shiraki H, Saito T, Yamada T, Tsujimoto M, Azuma M, Kurata H, Isoda S, Takano M, Shimakawa Y: Ferromagnetic Cuprates $CaCu_3Ge_4O_{12}$ and $CaCu_3Sn_4O_{12}$ with *A*-site Ordered Perovskite Structure, *Phys. Rev. B*, **76**, [140403-1]-[140403-4] (2007).

Zhang W, Okubo S, Ohta H, Saito T, Takano M: High-frequency ESR Measurements of the Co Spinel Compound $SiCo_2O_4$, *J. Phys. Cond. Mat.*, **19**, [145264-1]-[145264-6] (2007).

Oba N, Kageyama H, Saito T, Yoshimura K: Synchrotron X-ray Diffraction Study on the Square-lattice Antiferromagnets $(CuCl)LaNb_2O_7$ and $(FeCl)LaNb_2O_7$, *J. Mag. Mag. Mater.*, **310**, 1337-1339 (2007).

Tsujimoto Y, Baba Y, Oba N, Kageyama H, Fukui T, Narumi Y, Kindo K, Saito T, Takano M, Ajiro Y, Yoshimura K: 1/3 Magnetization Plateau in Spin-1/2 Square Lattice Antiferromagnet $(CuBr)Sr_2Nb_3O_{10}$, *J. Phys. Soc. Jpn.*, **76**, [063711-1]-[063711-4] (2007).

Presentations

Magnetic Frustration in the Layered Cobalt Oxide $SrCo_6O_{11}$, Saito T, Williams A, Attfield J P, Wuernisha T, Kamiyama T, Ishiwata S, Takeda Y, Shimakawa Y, Takano M, Gordon Research Conference on Solid State Chemistry II, Oxford UK, 4 September 2007.

Magnetic Phase Diagram of $SrCo_6O_{11}$ with Magnetization Plateaux, Saito T, Williams A, Attfield J P, Wuernisha T, Kamiyama T, Ishiwata S, Takeda Y, Shimakawa Y, Takano M, Spring Meeting of the Physical Society of Japan, Kagoshima Japan, 20 March 2007.

High Pressure Synthesis and Magnetism Research of Transition Metal Oxides, Saito T, The Ceramic Society of Japan, Kansai Branch Seminar, Kyoto Japan, 26 November 2007.

Grant

Takano M, Chemistry and Physics of 3d Transition Metal Oxides Equipped with Deep 3d Levels: Search for New Materials and New Functions, Grant-in-Aid for Scientific Research (S), 1 April 2005–31 March 2010.

Stepwise Magnetoresistance and Magnetic Frustration in $SrCo_6O_{11}$

$SrCo_6O_{11}$ is a layered cobalt oxide with unique magnetic and electronic properties, first synthesized using a high pressure technique in our group. Its magnetization process shows a plateau region at the 1/3 of the saturated magnetization M_0, which is concomitant with the negative and stepwise magnetoresistance effect. It was found that $SrCo_6O_{11}$ comprises itinerant electrons and Ising-like local spins of $S = 2$ on separate crystallographic sites, having strong interactions with each other. The spin structure at the 1/3 magnetization plateau state was found to be ferromagnetic in the *ab*-plane and like ↑-↑-↓-↑-↑-↓ along the *c*-axis. The quick reorientation of the ferromagnetic layers from the ↑-↑-↓ (M/M_0=1/3; M= magnetization) manner to the ↑-↑-↑ (M/M_0=1) manner under magnetic field should result in a major decrease of the magnetic scattering of conduction electrons penetrating through the ferromagnetic layers, which explains the negative, sharp and two-stepped magnetoresistance. The ↑-↑-↓ magnetic structure in the 1/3 magnetization plateau state implies the existence of competing magnetic interactions along the *c*-axis, forbidding simple antiferromagnetism or ferromagnetism. Such a competition in the magnetic interactions produce magnetic frustration in $SrCo_6O_{11}$, resulting in rather complex magnetic phase diagram. Large magnetostriction is also observed at low temperatures. Spin, charge and orbital degree of freedom is strongly coupled with each other in $SrCo_6O_{11}$, giving rise to the unique physical properties.

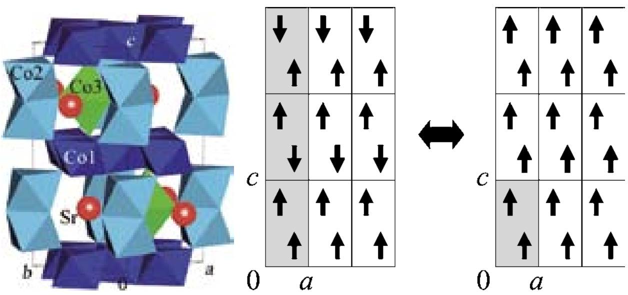

Figure 1. The crystal structure and spin structures of $SrCo_6O_{11}$. Up and down arrows represent Co(3) spins.

Magnetism of $CaCu_3B_4O_{12}$ (B = Ge, Ti, Sn) Perovskites

A-site ordered perovskites $CaCu_3Ge_4O_{12}$ and $CaCu_3Sn_4O_{12}$, which are isostructural to a well-known antiferromagnet (AFM) $CaCu_3Ti_4O_{12}$, were found to be ferromagnets (FM). $CaCu_3Sn_4O_{12}$ was newly synthesized under high pressure of 8 GPa. Though the crystal structure (*e.g.* lattice parameters, bond angles, bond lengths, *etc.*) of these materials changes systematically from $CaCu_3Ge_4O_{12}$ to $CaCu_3Ti_4O_{12}$ to $CaCu_3Sn_4O_{12}$, their magnetism changes in different manner, i.e. from FM to AFM to FM. Crystal structure analysis, magnetic measurements and electronic structure calculations were done to understand the origin of their magnetism. The special alignment of the CuO_4 planes in the crystal structure plays an important role in the magnetic properties of Cu^{2+} spins. Direct exchange interaction gives rise to the ferromagnetic behavior in $CaCu_3Ge_4O_{12}$ and $CaCu_3Sn_4O_{12}$, whereas involvement of Ti-3*d* orbitals produces the antiferromagnetic superexchange interaction in $CaCu_3Ti_4O_{12}$. This finding demonstrates that either ferromagnetic or antiferromagnetic behavior can appear within the same structural framework.

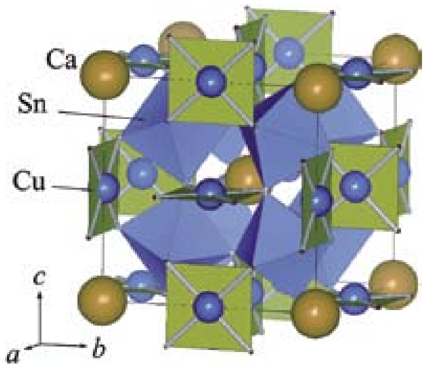

Figure 2. The crystal structure of $CaCu_3Sn_4O_{12}$.