Competing Spin Structures and Magnetic Frustration in Osmium Double Perovskites Sr_2BOsO_6 (B = Fe, Co, Y, In, Sc)

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Magnetic double perovskite (DP) oxides $A_2BB'O_6$ with an ordered arrangement of B and B' ions are interesting materials with several potential applications and raise fundamental questions about the nature of the interactions determining their properties. In insulating 3d/5d DPs Sr_2FeOsO_6 (Fe^{3+}/Os^{5+}) and Sr_2CoOsO_6 (Co^{2+}/Os^{6+}), the interplay between superexchange and electron-lattice interactions stabilizes two competing antiferromagnetic spin structures in contrast to the ferrimagnetic spin structures in related systems. Only one type of magnetic ion (Os^{5+}, $5d^3$) occurs in compounds Sr_2BOsO_6 with B = Y, In, Sc, which reveal frustrated exchange interactions within the *fcc*-type osmium sublattice and adopt a type I antiferromagnetic spin structure.

Double perovskite (DP) oxides, $A_2BB'O_6$, which feature an ordered rock-salt-like arrangement of corner-sharing BO_6 and $B'O_6$ units in the crystal structure, offer ample possibilities for the design of new magnetic and magneto-electric materials. Peculiar properties arise if an electronically more localized 3d ion at the B site is combined with an electronically more delocalized 4d or 5d ion on the B' site. Prototype DP oxides in this field are Sr_2FeMoO_6 and Sr_2FeReO_6, which are half-metallic ferromagnets with Curie temperatures above room temperature and large magneto-resistance effects, making them promising candidates for applications in spintronics. Another key member in the DP family is Sr_2CrOsO_6, which is a ferrimagnetic insulator with an unusually high Curie temperature, T_C, of 720 K. This illustrates the potential for finding promising magnets among DPs. In order to elucidate the interplay of interactions and to search for new magnetic phenomena, we investigated the magnetic and electronic properties of DP oxides Sr_2BOsO_6 with two magnetic sublattices (B = Fe, Co) and a single magnetic sublattice (B = Y, In, Sc).

The Antiferromagnetic 3d-5d Double Perovskites Sr_2FeOsO_6 and Sr_2CoOsO_6

In contrast to ferro- or ferrimagnetic double perovskites, Sr_2FeOsO_6 [1-3] and Sr_2CoOsO_6 [4,5] turned out to be *antiferromagnetic* insulators. The materials were synthesized from binary oxides in sealed quartz ampoules. Magnetic susceptibility and heat capacity studies revealed two magnetic phase transitions for both compounds, namely, at T_{N1} = 140 K, T_{N2} = 67 K for FeOs and T_{N1} = 108 K and T_{N2} = 67 K for CoOs. The crystal and spin structures were investigated by temperature-dependent powder neutron diffraction experiments. Sr_2FeOsO_6 crystallizes in a tetragonal DP structure (space group *I-4m*), with buckling of the Fe-O-Os layers in the *ab* plane, but an ideal Fe-O-Os angle of 180° along *c*. The results from the neutron diffraction study of Sr_2FeOsO_6 are summarized in Fig. 1. The two antiferromagnetic spin structures formed below T_{N1} (AF1) and T_{N2} (AF2) feature a ferrimagnetic arrangement of the Fe and Os spins in the *ab* plane but differ in the spin sequence along the *c*-direction. Whereas all Fe and Os spins show parallel alignment along *c* in AF1, an up-up, down-down, etc., sequence is observed in AF2. The magnetic transitions are accompanied by structural modulations, which are reflected in an increased tetragonal distortion in the temperature range of the magnetic phase transitions (Fig. 1c,d). Electronic structure calculations suggest that the AF2 spin structure is stabilized by an alternation of short and long Fe-Os distances [2,6]. A step-like evolution of the magnetic moments at the transition metal sites in the magnetically ordered phases (Fig. 1b) is confirmed independently by the temperature dependence of the hyperfine fields in ^{57}Fe Mössbauer spectra [2,3], which also verify the presence of Fe^{3+} ($t_{2g}^3e_g^2$) and thus Os^{5+} (t_{2g}^3) ions in Sr_2FeOsO_6. The gradual evolution of the magnetic moments indicates that the magnetic order in AF1 is not fully developed.

A complex magnetic ordering behavior with two distinct phases, AF1 and AF2, and a tetragonal (*I-4m*)-to-monoclinic (*$P2_1/n$*) phase transition at T_{N1} are found for Sr_2CoOsO_6 [4,5], where Co^{2+} ($t_{2g}^5e_g^2$) and Os^{6+} (t_{2g}^2) ions occur. Below T_{N1}, a spin structure with partially ordered Co^{2+} and Os^{6+} moments is formed, whereas, below T_{N2}, a canted non-collinear magnetic state occurs, where the Co moments are totally frozen. The Os moments still show spin-glass-like dynamics, which were studied by *ac* susceptibility and μ-SR experiments [5]. The detailed

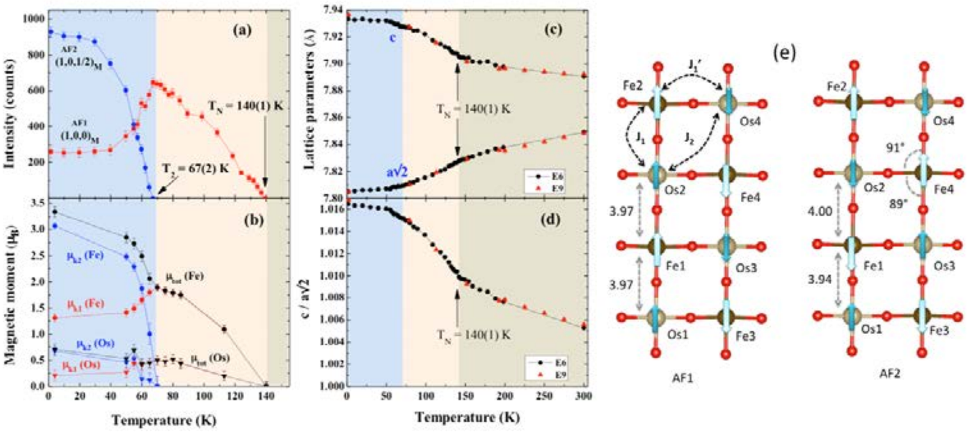

Fig. 1 (from Ref.[2]): Results obtained from powder neutron diffraction studies of Sr_2FeOsO_6. In (a,b), the evolution of the intensities of the magnetic Bragg reflections and of the magnetic moments is illustrated, and in (c,d), the increasing tetragonal structural distortion accompanying the magnetic phase transitions is shown. At the right, the spin structures of the AF1 and AF2 phases are compared.

spin structure of Sr_2CoOsO_6 is still a matter of discussion [7]. The peculiar spin structures of Sr_2FeOsO_6 and Sr_2CoOsO_6 are a manifestation of the large number of partly competing superexchange interactions, where long-range interactions between Os ions are particularly important [4,6]. This is a consequence of the large spatial extent of the Os 5d orbitals. In Sr_2CoOsO_6, the Co^{2+} and Os^{6+} sublattices are only weakly coupled because of the mismatch in orbital energies, which leads to weak nearest-neighbor Fe-Os interactions.

Magnetic Order and Frustration in Sr_2BOsO_6 (*B* = Y, In, Sc)

We have studied the structural and magnetic properties and the spin structures of DP oxides Sr_2BOsO_6 with B = Y, In, and Sc, where Os^{5+} ($t_{2g}{}^3$) ions are the sole magnetic component. [8] Both, Sr_2YOsO_6 and Sr_2InOsO_6 adopt the same type I antiferromagnetic spin structure as Sr_2YRuO_6, which was recently reinvestigated as a model system for magnetic frustration in an fcc-like lattice with antiferromagnetic interactions. The trend in the Néel temperatures (T_N = 52 K, 26 K, and 92 K for B = Y, In, and Sc, respectively) does not simply correlate with the chemical pressure effects by changing the size of the *B* ion. Instead, our results indicate that the detailed balance of exchange interactions is different in case of d^0 and d^{10} ions on the *B* site of the DP structure. Pronounced magnetic frustration is reflected in large values of 4-7 of the so-called frustration factor $|\theta_{CW}]/T_N$, where θ_{CW} corresponds to the Curie-Weiss temperature derived from the magnetic susceptibility data. The magnetism of the Os^{6+} ($5d^2$) compound Sr_2MgOsO_6 [9] is dominated by antiferromagnetic interactions as well, but the spin structure is not yet clear.

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