Competing Spin Structures and Magnetic Frustration in Osmium Double Perovskites $\text{Sr}_2\text{BO}_6$ ($B = \text{Fe}, \text{Co}, \text{Y}, \text{In}, \text{Sc}$)

Peter Adler*, Avijit Kumar Paul, Manfred Reehuis¹, Angelina Sarapulova, Binghai Yan, Sudipta Kanungo, Walter Schnelle, Liu Hao Tjeng, Claudia Felser, Martin Jansen

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 $\text{Sr}_2\text{FeOsO}_6$ ($\text{Fe}^{3+}/\text{Os}^{5+}$) and $\text{Sr}_2\text{CoOsO}_6$ ($\text{Co}^{2+}/\text{Os}^{6+}$), 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 ($\text{Os}^{5+}, 5d^0$) occurs in compounds $\text{Sr}_2\text{BO}_6$ with $B = \text{Y}, \text{In}, \text{Sc}$, 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 $\text{Sr}_2\text{FeMoO}_6$ and $\text{Sr}_2\text{FeReO}_6$, 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 $\text{Sr}_2\text{CrOsO}_6$, 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 $\text{Sr}_2\text{BO}_6$ with two magnetic sublattices ($B = \text{Fe}, \text{Co}$) and a single magnetic sublattice ($B = \text{Y}, \text{In}, \text{Sc}$).

The Antiferromagnetic 3d-5d Double Perovskites $\text{Sr}_2\text{FeOsO}_6$ and $\text{Sr}_2\text{CoOsO}_6$

In contrast to ferro- or ferrimagnetic double perovskites, $\text{Sr}_2\text{FeOsO}_6$ [1-3] and $\text{Sr}_2\text{CoOsO}_6$ [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. $\text{Sr}_2\text{FeOsO}_6$ 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 $\text{Sr}_2\text{FeOsO}_6$ 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 $^{57}\text{Fe}$ 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 $\text{Sr}_2\text{FeOsO}_6$. 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 $\text{Sr}_2\text{CoOsO}_6$ [4,5], where Co$^{3+}$ $(t_{2g}^3e_g^2)$ and Os$^{6+}$ $(t_{2g}^3)$ 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 $\mu$-SR experiments [5]. The detailed
spin structure of Sr$_2$CoOsO$_6$ is still a matter of discussion [7]. The peculiar spin structures of Sr$_2$FeOsO$_6$ and Sr$_2$CoOsO$_6$ 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$_2$CoOsO$_6$, 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$_2$BOsO$_6$ (B = Y, In, Sc)**

We have studied the structural and magnetic properties and the spin structures of DP oxides Sr$_2$BOsO$_6$ with B = Y, In, and Sc, where Os$^{5+}$ ($t_2g^3$) ions are the sole magnetic component. [8] Both, Sr$_2$YOsO$_6$ and Sr$_2$InOsO$_6$ adopt the same type I antiferromagnetic spin structure as Sr$_2$YRuO$_6$, 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$^8$ 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 $\theta_{CW}$ corresponds to the Curie-Weiss temperature derived from the magnetic susceptibility data. The magnetism of the Os$^{6+}$ (5d$^2$) compound Sr$_2$MgOsO$_6$ [9] is dominated by antiferromagnetic interactions as well, but the spin structure is not yet clear.

**References**


* adler@cpfs.mpg.de
1 Helmholtz-Zentrum Berlin für Materialien und Energie, 14109 Berlin, Germany