

**$J_{\text{eff}} = 3/2$  ferromagnetic insulating state above 1000 K  
in a double perovskite osmate  $\text{Sr}_3\text{OsO}_6$**

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In magnetic insulating oxides, the cubic crystal symmetry is thought to be favorable for higher Curie temperatures ( $T_C$ ) [1], since the network morphology hosting the mechanisms of exchange interactions is subject to the crystal symmetry. Especially for perovskites, it is well known that the crystal symmetry dominates the exchange interactions, e.g.,  $\text{SrRuO}_3$  (pseudo-cubic perovskite, ferromagnetic metal) and  $\text{CaRuO}_3$  (orthorhombic perovskite, paramagnetic metal) [1]. In addition to the symmetry, adopting 5d transition metal oxides, where spin-orbit coupling (SOC) is enhanced, is a promising way to boost the long range ferri/ferromagnetic (FM) order. For example, it was shown for a pyrochlore osmate ( $\text{Cd}_2\text{Os}_2\text{O}_7$ ) that the SOC is responsible for its long range order [2]. Here we show that a highly B-site ordered cubic double-perovskite  $\text{Sr}_3\text{OsO}_6$ , which satisfies the above criteria, has the highest  $T_C$  of  $\sim 1060$  K among all insulators and oxides.

We synthesized the novel magnetic insulator  $\text{Sr}_3\text{OsO}_6$  (300-nm thick) by molecular beam epitaxy (MBE) on (001)  $\text{SrTiO}_3$  substrates [3]. Atomic-resolution scanning transmission electron microscopy (STEM) has revealed the cubic structure and excellent Sr/Os ordering on the B'/B-site (Fig. 1). Our chemical analyses using Rutherford backscattering spectroscopy and energy dispersive X-ray spectroscopy exclude the possibility of the contamination by magnetic impurities. The hexavalent state of Os is confirmed by X-ray photoemission spectroscopy. The magnetic properties were measured by a superconducting quantum interference device (Fig. 2). The results indicate that, in 5d systems, extraordinary high  $T_C$  is possible in spite of the long distance ( $5.81\text{\AA}$ ) between 5d ions.

Density-functional-theory calculations also elucidate the ferromagnetic ground state of  $\text{Sr}_3\text{OsO}_6$ , where the large SOC of the  $\text{Os}^{6+}$  5d<sup>2</sup> orbitals indeed drives the system toward an insulating state with  $J_{\text{eff}} = 3/2$ . The  $t_{2g\uparrow}$  states are split into effective total angular momenta of  $J_{\text{eff}} = 3/2$  (doublet) and  $J_{\text{eff}} = 1/2$  (singlet) states. The  $J_{\text{eff}} = 3/2$  states are fully occupied with two 5d electrons per  $\text{Os}^{6+}$ , resulting in an insulating state.

The combination of the cubic rock-salt-type ordering of  $\text{Os}^{6+}$  ions and the enhanced SOC found in 5d systems is key for the  $J_{\text{eff}} = 3/2$  insulating state with extraordinary high  $T_C$  above 1000 K in contrast to isoelectronic compound,  $\text{Ca}_3\text{OsO}_6$  (monoclinic,  $T_N = 50$  K) [4].

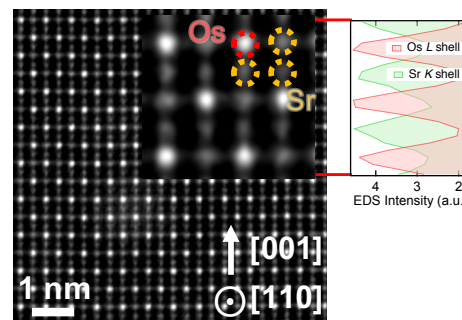


Figure 1 STEM image of the  $\text{Sr}_3\text{OsO}_6$  layer along the  $[110]$  direction. The inset show enlarged views together with EDS-STEM profiles along the  $[001]$  direction.

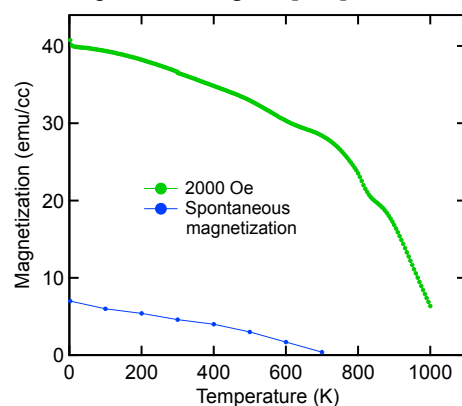


Figure 2 Magnetization as a function of temperature with  $H = 2000$  Oe for a  $\text{Sr}_3\text{OsO}_6$  film. Spontaneous magnetization as a function of temperature is also shown.

- [1] G. Koster, *et al.*, Rev. Mod. Phys. **84**, 253 (2012). [2] S. Calder, *et al.*, Nat. Commun. **7**, 1 (2016). [3] Y. K. Wakabayashi, *et al.*, Nat. Commun. **10**, 535 (2019). [4] H. L. Feng, *et al.*, J. Solid State Chem. **201**, 186 (2013).