## $J_{\text{eff}} = 3/2$ ferromagnetic insulating state above 1000 K in a double perovskite osmate synthesized by molecular beam epitaxy

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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., SrRuO<sub>3</sub> (pseudo-cubic perovskite, ferromagnetic metal) and CaRuO<sub>3</sub> (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  $(Cd_2Os_2O_7)$  that the SOC is responsible for its long range order [2]. Here we show that a highly B-site ordered cubic double-perovskite Sr<sub>3</sub>OsO<sub>6</sub>, which satisfies the above criteria, has the highest  $T_{\rm C}$  of ~1060 K among all insulators and oxides, and the highest magnetic ordering temperature in any compound without 3d transition elements.

We synthesized the novel magnetic insulator  $Sr_3OsO_6$ (300-nm thick) by molecular beam epitaxy (MBE) on (001)  $SrTiO_3$  substrates [3]. Atomic-resolution scanning transmission electron microscopy (STEM) has revealed the cubic structure and excellent Sr/Os ordering on the B'/Bsite (Fig. 1). The magnetic properties were measured by a superconducting quantum interference device (Fig. 2). The results indicate that, in 5*d* systems, extraordinary high  $T_C$  is possible in spite of the long distance (5.81Å) between 5*d* ions.



Figure 1 STEM image of the Sr<sub>3</sub>OsO<sub>6</sub> layer along the [110] direction. The inset show enlarged views together with energy dispersive X-ray spectroscopy (EDS)-STEM intensity profiles along the [001] direction.



Figure 2 Magnetization as a function of temperature with H = 2000 Oe for a Sr<sub>3</sub>OsO<sub>6</sub> film. Spontaneous magnetization as a function of temperature is also shown.

Density-functional-theory calculations also elucidate the ferromagnetic ground state of Sr<sub>3</sub>OsO<sub>6</sub>, where the large SOC of the Os<sup>6+</sup>  $5d^2$  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 5*d* electrons per Os<sup>6+</sup>, resulting in an insulating state.

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

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