Imaging and controlling all-in/all-out magnetic domains in pyrochlores

The pyrochlore lattice can be described as a cubic lattice of tetrahedra, joined by shared vertices (Fig. 1(a,b)). In the compounds $A_2B_2O_7$ of interest, magnetic ions B are located at these vertices and it is impossible to satisfy all their interactions if the exchange is antiferromagnetic. As a result, several magnetic states may co-exist at the same energy. One particular case is the so-called "all-in/all-out" magnetic structure: in the presence of uniaxial magnetic anisotropy along <111> all the magnetic moments on the vertices of a given tetrahedron will point either towards the center or away from it. Consequently, if one sets the orientation of a given magnetic moment, the orientation of all the others is decided. Therefore only two different states – corresponding to the two choices for the original orientation – can describe the magnetic structure: all-in/all-out (AIAO) or all-out/all-in (AOAI) as shown in Fig. 1 [1]. The AIAO and AOAI states are time-reversal symmetric of each other, which we may define as opposite *pseudo-orientation*. They can also be described as differently oriented zinc-blende crystals of magnetic hedgehog monopoles (Fig.1(c)) or face-centered crystals of magnetic octupoles. They are thus expected to host unconventional opposite magnetic behaviors [2], such as opposite linear magnetostriction, opposite linear magneto-capacitance or opposite quadratic terms in the magnetization. Suitable experimental means to image and control the AIAO and AOAI magnetic domains are therefore much needed.

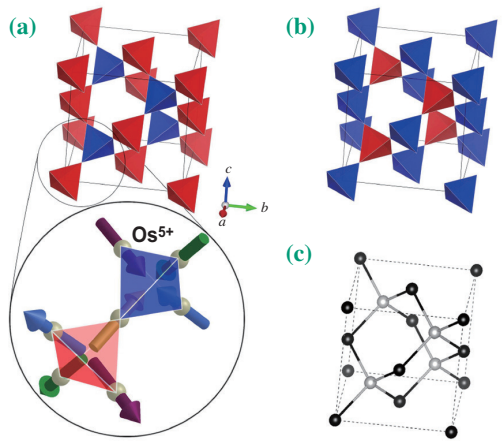

Fig. 1. **(a)** All-in/all-out and **(b)** all-out/all-in magnetic order on the pyrochlore lattice; both configurations are time-reversal symmetric of each other. All Os spins are located at the vertices of the tetrahedra and point either towards the center of the blue tetrahedra or away from the center of the red tetrahedra, as shown in the magnified region. **(c)** Equivalent zinc-blende lattice [1].

In this work, we experimentally investigated the compound $Cd_2Os_2O_7$: the AIAO ordering of the spins of the $5d^3$ Os^{5+} ions was previously reported below $T_N = 225$ K [3], along with a continuous metal-insulator transition [4]. In order to distinguish the AIAO order from the AOAI, i.e. to measure the *pseudo-orientation*, we performed polarized resonant X-ray microdiffraction at beamline **BL19LXU**. The structure factor of space-group forbidden 0 0 $4n+2$ Bragg reflections at the Os L_3 resonance (10.871 keV) is the sum of two components: a non-magnetic part due to the anisotropic tensor of susceptibility (ATS) and the screw axis along *c*, F_{ATS}; and a magnetic part due to the spin order, F_m . The diffracted intensity results from the interference of these two components. The key point is that the sign of F_m is opposite in AIAO and AOAI while that of F_{ATS} is constant. Since both F_{ATS} and F_m are polarization-dependent, we defined the contrast in diffraction for right-handed (I^+) and left-handed (I^-) circular polarization as the flipping ratio $FR = (I^+ - I^-)/(I^+ + I^-)$. We demonstrated that the sign of *FR* is opposite in opposite types of domains, which could be used to distinguish AIAO from AOAI. The *FR* was measured experimentally using a diamond phase plate upstream of the sample to control the handedness of the incident beam. A pair of Kirkpatrick-Baez mirrors was used to focus the X-ray beam to a 500 × 500 nm² spot on the sample, which was raster scanned.

The resulting maps measured over the (001) and {001} adjacent facets of the sample are shown in Fig. 2(a-f). Stripe-like patterns of opposite *FR* sign were observed, indicating the coexistence of AIAO and AOAI domains within the same crystal. The typical size of the magnetic domains was a few tens of microns. We note that these results are the first experimental imaging of AIAO and AOAI magnetic domains in the pyrochlore lattice. At the interface between opposite magnetic domains, magnetic domain walls (DW) could be observed. From the analysis of the DW orientations on the different facets, we could suggest two families of planes for the DW: {(113), (11$\bar{3}$), (1$\bar{1}$3), ($\bar{1}$13) and circular *hkl* permutations}, and {(011), (01$\bar{1}$) and circular *hkl* permutations}. The observation of DW is consistent with other experimental measurements, such as the finite conductivity at low temperature (attributed to transport along the DW) and the remnant magnetization below the antiferromagnetic ordering temperature (attributed to magnetic ordering of the frustrated spins in the DW).

Finally we showed that we could control the

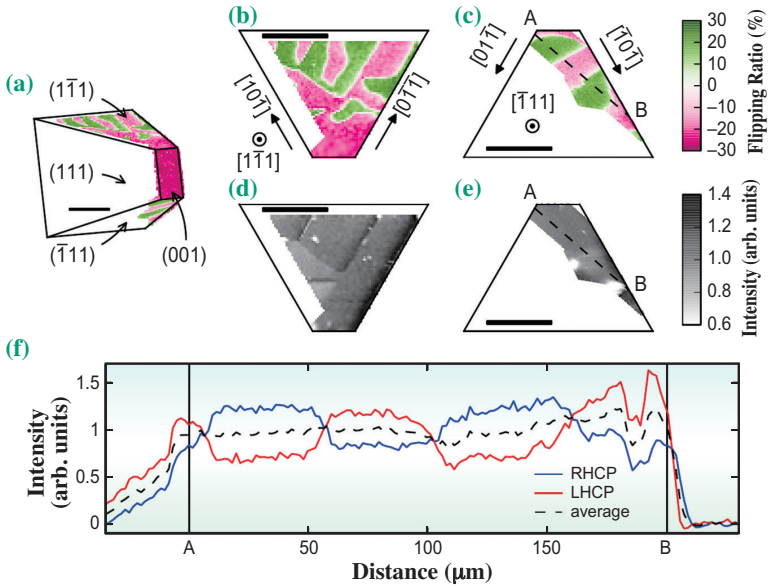

Fig. 2. **(a)** FR map at the 0 0 10 reflection, as measured experimentally by rastering the sample in a plane perpendicular to the scattering vector. The ideal crystal is outlined and the orientation of the facets is shown. Opposite signs of the FR indicate opposite pseudo-orientations of the magnetic domains (AIAO *vs* AOAI). **(b, c)** FR maps and **(d, e)** normalized intensity on the top (1 $\bar{1}$ 1) and bottom ($\bar{1}$ 1 1) facets after correction of the projection, respectively. The intensity is normalized to the average value on each facet to outline the deviation from the average structure. All scale bars are 100 mm across. **(f)** Line scan across the bottom ($\bar{1}$ 1 1) facet, indicated by the AB dashed line in **(c, e)**, for both right- and left-handed circular polarization and their average [1].

orientation of the AIAO and AOAI domains by using a magnetic field-cooling procedure and orienting the field along a <111> direction. We showed that using a double magnets setup we could reverse the domain distribution by reversing the direction of the magnetic field while cooling through T_N, as illustrated in Fig. 3. The possible origins of the coupling to the external field are the domain-dependent piezomagnetism and non-linear magnetization mentioned earlier, as well as the presence of uncompensated spins on the {111} facets.

Our results show that AIAO/AOAI magnetic domains can be measured in real space and their distribution controlled. This paves the way for future investigation of pyrochlore lattices with all-in/all-out magnetic ordering.

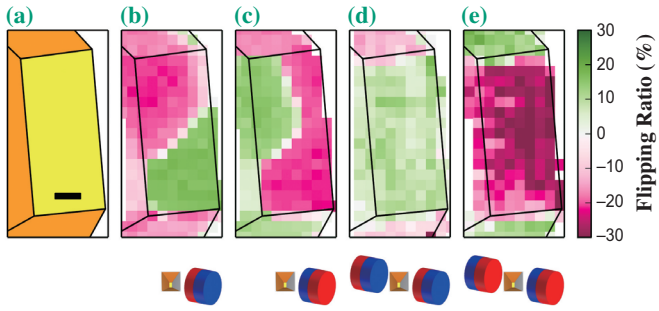

Fig. 3. **(a)** Sketch of the sample view in the geometry used, the central rectangle is the (0 0 1) facet. The scale bar is 25 mm across. **(b–e)** Maps of the FR across the sample at the 0 0 10 reflection at 100 K for different field cooling conditions, as sketched in the lower panels. The North pole of the cylindrical permanent magnets is indicated in red, the magnetic field at the surface of the magnets is 0.4 T. The surface of the magnet is also parallel to the ($\bar{1}$ $\bar{1}$ 1) facet, indicated in light grey in the sketch [1].

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