Controlling the all-in-all-out magnetic domains in pyrochlore iridate thin films and heterostructures

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Domain walls of ferromagnetic or ferroelectric materials have attracted little attention as active elements of devices, but rather are considered to form as metastable objects of ferroic domains. In the case of ferromagnetic metal, domain walls frequently act as scatterers of electrons, deteriorating the device operation. Here we present transport properties of pyrochlore iridate thin films and heterosturctures, where metallic conduction at the domain walls are theoretically proposed while the bulk is kept insulating in stark contrast to conventional ferromagnetic metal.

Pyrochlore materials, expressed as $A_2B_2O_7$ (*A*: rare-earth, *B*: transition metal), are composed of tetrahedral network of rare-earth and iridium sublattices, respectively as shown in Fig. 1. Characteristic of this compound is the all-in-all-out spin structure, where all the four spins at the vertices of a tetrahedron point inward or outward alternatingly due to the cooperation of strong spin-orbit interaction and spin frustration. Although the all-in-all-out spin structure is antiferromagnetic, there are clearly distinct two magnetic domains as shown in Fig. 1, which we call A domain and B domain for simplicity. While pyrochlore iridates are metallic in the paramagnetic phase above the Neel temperature T_N, the all-in-all-out spin structure is theoretically predicted to induce intriguing semimetallic or insulating phases below T_N,[1)] depending on the strength of electron correlation tuned by the choice of rare-earth ions; the smaller the rare-earth ions are, the stronger the electron correlation is. Additionally, in the insulating phase, conducting domain walls are theoretically predicted,[2)] followed by experimental observation by microwave impedance microscopy.[3)] Although the electronic phases of the pyrochlore iridates have recently been gradually clarified experimentally, thin films are still difficult to fabricate and controlling the domain walls is not easily accessible in heterostructures. In this study, we aim at fabricating pyrochlore iridate thin films and heterostructures to artificially control the all-in-all-out magnetic domains.

The pyrochlore iridate thin films are fabricated by pulsed laser deposition using Y-stabilized ZrO_2 (111) substrates. The oxygen partial pressure and substrate temperatures are varied to find optimum growth conditions but the epitaxial $Eu_2Ir_2O_7$ films are not obtained probably due to low formation energy of pyrochlore iridates. Instead, we anneal the thin films after depositing amorphous films, resulting in successful formation of $Eu_2Ir_2O_7$ thin films. This method, so-called solid-state epitaxy, is also found to be applicable to pyrochlore iridate thin films with other rare-earth ions. X-ray diffraction and transmission electron microscope also show single crystalline $Eu_2Ir_2O_7$ thin films are formed.[4)]

For controlling the all-in-all-out magnetic domain wall, we have fabricated $Eu_2Ir_2O_7/Tb_2Ir_2O_7$ heterosturucutres.[5)] Eu^{3+} is nonmagnetic, while Tb^{3+} has a large magnetic moment of $J = 6$. Thus, we expect that magnetic domains of $Eu_2Ir_2O_7$ is not sensitive to external magnetic field and the domains of $Tb_2Ir_2O_7$ may be switched by magnetic field. Figure 2 shows magnetoresistance (MR) of $Eu_2Ir_2O_7$ and $Tb_2Ir_2O_7$ thin films. In the case of $Eu_2Ir_2O_7$, MR does not show hysteresis, but

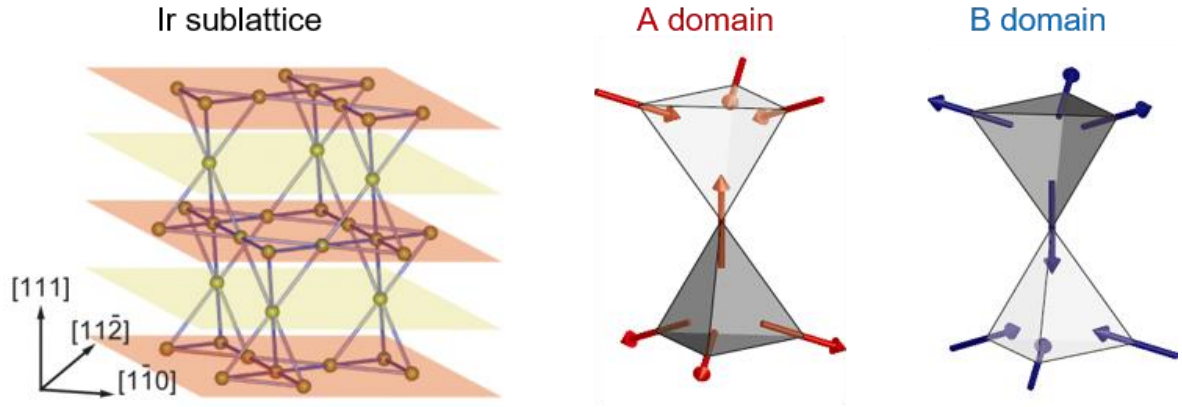

Fig. 1. Ir sublattice of pyrochlore iridate $A_2Ir_2O_7$ (*A*: rare-earth) and spin structures of two all-in-all-out magnetic domains (A domain and B domain).

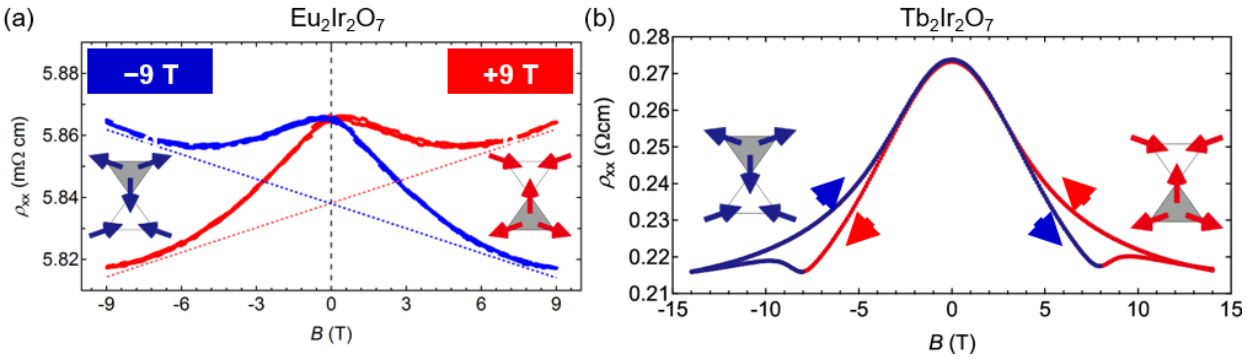

Fig. 2. Magnetoresistance of (a) $Eu_2Ir_2O_7$ and (b) $Tb_2Ir_2O_7$ thin films at 2 K. For $Eu_2Ir_2O_7$ thin film, magnetoresistance is measured after cooling the sample under + 9 T and −9 T, while that of $Tb_2Ir_2O_7$ thin film shows double hysteresis, indicating domain switching. The insets are expected domains of Ir spins.

is asymmetric with respect to magnetic field. The sign of the asymmetric part is inverted between positive and negative field cooling, which suggests that all-in-all-out magnetic domain is selectively stabilized by the polarity of the magnetic field. In contrast, MR possesses double hysteresis for the $Tb_2Ir_2O_7$ thin film, suggestive of magnetic domain switching.[6] The stabilization of the all-in-all-out domains can be visualized by scanning SQUID microscopy.[7] Although all-in-all-out spin structure in the cubic symmetry does not produce dipole moment, subtle distortion of the lattice can hold dipole moment, the sign of which depends on the all-in-all-out magnetic domain. Figure 3 shows the images of scanning SUQID microscope. Under zero-field cooling, magnetic domains are clearly observed. After warming up and cooling down the sample from 130 K under positive magnetic field, uniform shift of magnetic field is observed. This result clearly demonstrates the above assumption that the all-in-all-out magnetic domains can be selectively stabilized by cooling magnetic field.

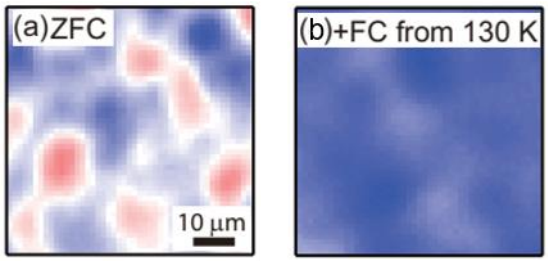

Fig. 3. Scanning SUQID images of a $Tb_2Ir_2O_7$ thin film at 4.7 K after (a) zero-field cooling and (b) cooling under positive magnetic field from 130 K.

Finally, we fabricated $Eu_2Ir_2O_7/Tb_2Ir_2O_7$ heterostructure and measured the magnetoconductance at the interface as shown in Fig. 4. The interface conductance exhibits hysteresis. Obviously, interface conductance is higher when the magnetic domains of $Eu_2Ir_2O_7$ and $Tb_2Ir_2O_7$ layers are opposite than when those magnetic domains are the same. This result indicates that the domain wall conduction is successfully controlled in the heterostructure with selective domain stabilization.

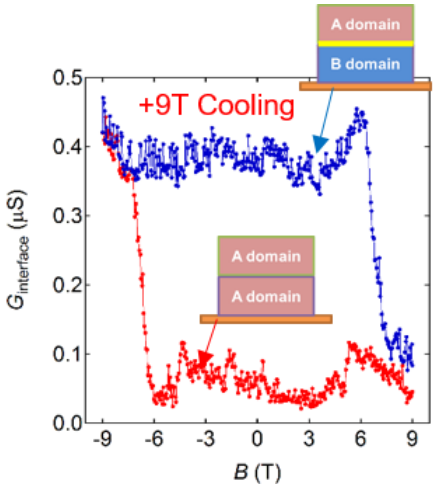

Fig. 4. Conductance at the $Eu_2Ir_2O_7/Tb_2Ir_2O_7$ heterointerface after cooling under +9 T.

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