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Authors	S. Chakraverty,M. Saito,S. Tsukimoto,Y. Ikuhara,A. Ohtomo,M. Kawasaki
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Magnetic properties of Sr_2FeTaO_6 double perovskite epitaxially grown by pulsed-laser deposition

S. Chakraverty,[1,a)] M. Saito,[2] S. Tsukimoto,[2] Y. Ikuhara,[2,3,4] A. Ohtomo,[5]
and M. Kawasaki[1,2,6,7]
[1]*Correlated Electron Research Group (CERG) and Cross-Correlated Materials Research Group (CMRG), RIKEN Advanced Science Institute, Wako 351-0198, Japan*
[2]*WPI Advanced Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*
[3]*Institute of Engineering Innovation, The University of Tokyo, Tokyo 113-8656, Japan*
[4]*Nanostructures Laboratory, Japan Fine Ceramics Center, Nagoya 456-8587, Japan*
[5]*Department of Applied Chemistry, Tokyo Institute of Technology, Tokyo 152-8552, Japan*
[6]*Quantum-Phase Electronics Center and Department of Applied Physics, University of Tokyo, Tokyo 113-8656, Japan*
[7]*CREST, Japan Science and Technology Agency, Tokyo 102-0075, Japan*

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We have investigated the magnetic properties of *B*-site-ordered Sr_2FeTaO_6 double perovskite thin films grown on (111) $SrTiO_3$ substrates by using pulsed-laser deposition. High degree of ordering of Fe and Ta ions has been confirmed using x-ray diffraction and atomically resolved high-angle annular dark-field scanning transmission electron microscopy. We have found clear signatures of magnetic frustration in {111} planes of local spin moment arising from next-nearest-neighbor interaction in triangular FeO_6 sublattice below 50 K. It is also found that this behavior is suppressed by increasing disorder and eventually leads to antiferromagnetic state. © *2011 American Institute of Physics*. [doi:10.1063/1.3663214]

Double perovskites $A_2B'B''O_6$ are one of the most important classes of oxide materials, where the magnetic ground states can be designed and manipulated by choosing different combinations of B' and B'' cations. After the discovery of a high-Curie temperature half-metallic bulk ferromagnet Sr_2FeMoO_6,[1] synthesis of double perovskite has become an important subject to search potential materials for future spinelectronic applications.[2] It is of particular interest to study how magnetic ground state is modified by replacing Mo^{5+} with nonmagnetic transition metal ions such as Nb^{5+} and Ta^{5+}.[3] Regarding Sr_2FeTaO_6 (SFTO), only a disordered phase exists, exhibiting spinglass-like behavior below ∼23 K.[3,4] Such behavior was previously ascribed to next-nearest-neighbour (nnn) interaction between magnetic ions (i.e., Fe).[3] While those observations may also be attributed to the randomness in the magnetic interactions arising from the disordered *B*-site arrangements.

In order to verify the presence of nnn-interaction, it is essential to stabilize Sr_2FeTaO_6 in *B*-site-ordered form. Because of similar ionic radii of Fe^{3+} (0.645 Å) and Ta^{5+} (0.64 Å),[5] the ordered structure of this material refuses to grow in bulk form.[3,6] Whereas, substitution of Sr^{2+} by La^{3+} can produce an *B*-site-ordered structure.[7,8] One of the plausible reasons is that the formal valence difference between Fe^{3+} and Ta^{5+} may not be large enough to be ordered in stable state of Sr_2FeTaO_6. However, by reducing valence state of Fe through La^{3+} doping, the situation can be altered.[9] Hence, it is important to verify the possibility of *B*-site ordering in Sr_2FeTaO_6 without doping, if the reaction is forced to follow a path where the formal valence difference between Fe and Ta is more than two (i.e., Fe^{2+}/Fe^{3+} and Ta^{5+}) during the growth. Here we demonstrate that such an idea is feasible.

The SFTO films with a thickness of ∼50 nm were grown on an atomically flat surface of $SrTiO_3$ (STO) (111) single crystal by using a pulsed-laser deposition (PLD) system. KrF excimer laser pulses (248 nm, 4 Hz) were focused on a target (a SFTO ceramic tablet with disordered phase as confirmed by x-ray diffraction (XRD)) with a spot area (0.35×0.10 cm^2) at a fluence of 1.1 J/cm^2. The PLD growth was performed at various growth temperatures (T_g) and oxygen partial pressures (Po_2) as displayed in Fig. 1(a). To rule out the possibility of oxygen vacancies and mixed valence of Fe, we annealed the samples in air at 400 °C for 4 h.

The SFTO films deposited above $T_g = 900\,°C$ were found to crystallize with the (111) oriented perovskite structure, and no reflection indicating secondary phase was observed from θ to 2θ XRD pattern carried out by using a four-circle goniometer (X′pert MRD, PANalytical) with Cu K_α radiation ($\lambda = 1.5418$ Å). We have found that in a narrow window of growth conditions [$Po_2 = 3 \times 10^{-8}$ Torr in the range of T_g from 960 to 1010 °C, see Fig. 1(a)] superlattice peaks appeared along the [111] direction [marked with arrows in Fig. 1(b)], confirming the formation of double perovskite. From this result, it is concluded that highly reducing condition is suitable to stabilize the *B*-site ordered phase of this material.

Compared in Fig. 1(b) are the XRD patterns of disordered and ordered phases of SFTO films prepared in $Po_2 = 1 \times 10^{-7}$ Torr at $T_g \sim 900\,°C$ (sample A) and in $Po_2 = 3 \times 10^{-8}$ Torr at $T_g \sim 1010\,°C$ (sample B), respectively. Taking pseudocubic lattice into account, the average lattice parameter (a_{av}) is deduced through $a_{av} = V_{av}^{1/3}$, where V_{av} is the volume of SFTO unit cell, which satisfies the condition $V_{av} = 6\sqrt{3} \times (d_{11-2})^2 \times d_{111}$, where d_{111} and d_{11-2} are

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[a)] Electronic mail: suvankar.chakraverty@riken.jp.

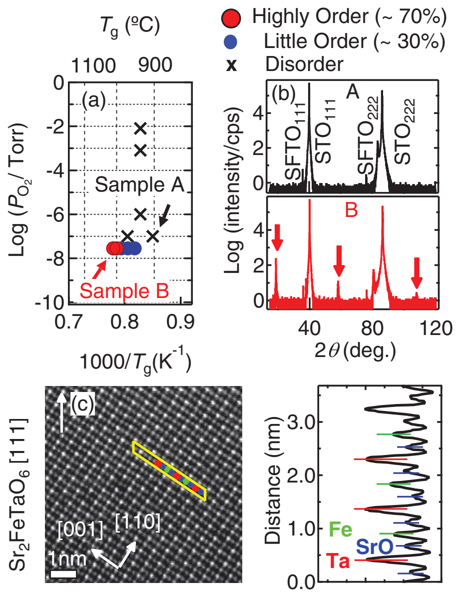

FIG. 1. (Color online) (a) Growth mode mapping representing Po_2 and T_g for the various samples grown with single phase perovskite structure. Symbols indicate degree of ordering at different growth conditions, larger filled circle, smaller filled circles, and crosses represent highly ordered, little ordered, and disordered phase, respectively. (b) $\theta-2\theta$ x-ray diffraction pattern of disordered (sample A, upper panel) and ordered (sample B, lower panel) films. (c) HAADF-STEM image of a highly ordered Sr_2FeTaO_6 film (sample B) grown along the [111] direction. Right panel shows the intensity profile along yellow column in (c).

estimated from $\theta-2\theta$ scan and (330) mapping, respectively. a_{av} for disordered and ordered samples are 3.96 Å and 4.05 Å, respectively.[10]

The crystal structure of sample B was further characterized using the atomically resolved high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) equipped with an aberration corrector and electron energy loss spectroscopy, operating at an acceleration voltage of 200 kV. High-degree of ordering in *B*-site ions was clearly visible from the intensity modulation difference due to periodic atomic arrangement [Fig. 1(c)]. The degree of ordering of sample B was estimated to be ~70% by statistical analysis of the STEM image.

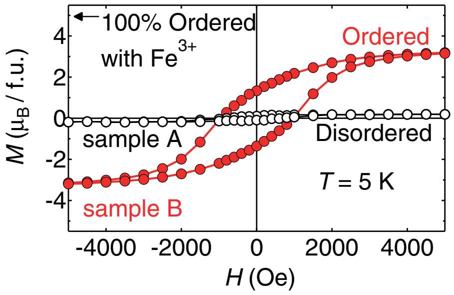

FIG. 2. (Color online) Magnetization as a function of applied magnetic field for sample A (open circle) and sample B (filled circle) at 5 K. Expected value of saturation magnetization of a fully ordered SFTO with Fe^{3+} state is also indicated.

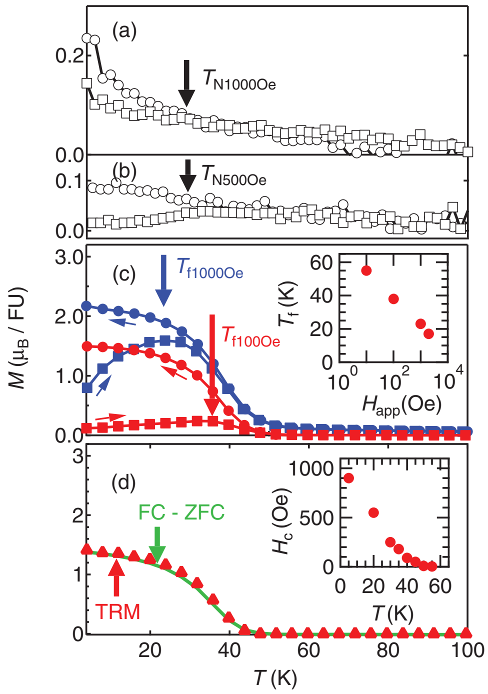

FIG. 3. (Color online) (a) and (b) FC (open circle) and ZFC (open square) magnetization for sample A measured in 1000 Oe and 500 Oe of applied magnetic field, respectively. The transition temperature (T_N) is marked by arrows. (c) FC (filled circle) and ZFC (filled square) magnetization for sample B measured in 1000 Oe (blue) and 100 Oe (red) of applied magnetic field, respectively. Inset shows the freezing temperature (T_f) of sample B as a function of different applied fields. (d) TRM (filled triangle) and difference of FC and ZFC magnetization (solid line) measured in 100 Oe of applied magnetic field. Inset shows the magnetic coercivity of sample B as a function of temperature.

The magnetic properties were measured using a quantum design magnetic properties measuring system in reciprocating sample option (RSO) mode. All the magnetic data presented hereafter include the correction of diamagnetic background of STO substrate. Both the samples showed well defined magnetic hysteresis loop at 5 K as shown in Fig. 2. Sample A showed a very low saturation magnetization of ~0.2 μ_B/f.u. (Bohr magnetron per formula unit) at 5 K. In contrast, sample B had a much higher saturation magnetization of ~3.2 μ_B/f.u. Field-cooled (FC) and zero-field-cooled (ZFC) magnetization measurements were carried out on both samples under various applied fields (H_{app}).[11] A bifurcation in FC and ZFC curves followed by a hump in ZFC curve had been observed in both the samples. In sample A, the position of the hump in ZFC and the bifurcation between FC and ZFC curves is independent of H_{app} as shown in Figs. 3(a) and 3(b). It is worth noting that the bifurcation between FC and ZFC became vanishingly small when the measurement was carried out under $H_{app} = 1000$ Oe, suggesting that this is an effect of coercivity and this hump is associated with the antiferromagnetic behaviour of the sample. The estimated position of this hump is ~25 K, which matches well with that reported for the bulk disordered sample.[3,6] In contrast,

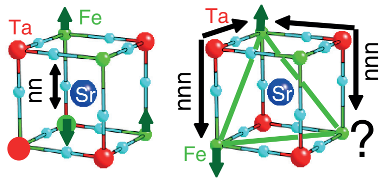

FIG. 4. (Color online) Schematics of crystalline and possible magnetic structures of disordered (left) and ordered (right) phase of Sr_2FeTaO_6 below their respective magnetic transition temperatures.

the scenario is quite different for sample B. It is worth noting that for sample B, the position of the hump (defined as the freezing temperature T_f) was shifting systematically towards higher temperature with decreasing H_{app} [see the inset of Fig. 3(c)] unlike sample A. The value of T_f at $H_{app} = 10$ Oe was around 55 K. Whereas the coercivity vanished at around 45 K [inset of Fig. 3(d)], indicating the possibility of frustrated magnetic state. [12] To confirm that, we carried out another measurement, in which the sample B was cooled to 5 K at a constant rate of 4 K/min with $H_{app} = 100$ Oe. Then, the magnetic field was withdrawn at 5 K, and the system was heated to room temperature at same constant rate while the magnetization was monitored. This protocol is formally known as thermoremanant magnetization (TRM).[12] Note that in Fig. 3(d) TRM almost exactly matches with the FC magnetization subtracted by the ZFC magnetization under $H_{app} = 100$ Oe. These magnetic measurements suggest the frustrated magnetic structure in sample B.

Tantalizing fact in our observation is that *B*-site ordering induces spin frustration in the system. Here we apply the following possible model to explain the present scenario. In disordered structure, there is a high probability that one Fe ion is occupied next to another one where local spin moment is aligned antiparallel via antiferromagnetic nearest-neighbour (nn) superexchange interaction as it is the case for most of perovskite (e.g., $LaFeO_3$).[13] In the ideally ordered structure, however, each Fe ion can interact with another one only through nnn superexchange interaction via Fe-O-Ta-O-Fe because of the periodic dilution of *B*-site with nonmagnetic Ta ions.[14] In such a triangular FeO_6 sublattice, magnetic frustration naturally arises from competing coupling strength of local spin moments. This scenario is schematically presented in Fig. 4.

In conclusion, we have demonstrated the nonequilibrium growth of the *B*-site ordered structure of Sr_2FeTaO_6 by using pulsed-laser deposition technique, which may pave a way to grow a large variety of new double perovskite oxides. The magnetic properties of ordered and disordered phases were studied under various magnetic protocols. Magnetic measurements clearly show the signatures of frustrated magnetic behavior associated with nnn interaction in the magnetic Fe^{3+} sublattice in structurally ordered phase. The magnetic property of the disorder phase is quite similar to the bulk phase of this material. This material may be considered as a prototype material where the proper choice of *B*-site ions may produce a new class of magneto-electrically coupled system for future generation spin-based devices.

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