A new class of materials termed topological insulators have been intensively investigated due to their unique Dirac surface state carrying dissipationless edge spin currents. Recently, it has been theoretically proposed that the three dimensional analogue of this type of band structure, the Weyl Semimetal phase, is materialized in pyrochlore oxides with strong spin-orbit coupling, accompanied by all-in-all-out spin ordering. Here, we report on the fabrication and magnetotransport of Eu$_2$Ir$_2$O$_7$ single crystalline thin films. We reveal that one of the two degenerate all-in-all-out domain structures, which are connected by time-reversal operation, can be selectively formed by the polarity of the cooling magnetic field. Once formed, the domain is robust against an oppositely polarised magnetic field, as evidenced by an unusual odd parity magnetoresistance in magnetic domain walls of pyrochlore iridates. Our findings pave the way for exploring the predicted novel quantum transport phenomenon at the surfaces/interfaces or magnetic domain walls of pyrochlore iridates.
properties mediated by all-in-all-out spin ordering. We chose \( Ln = Eu \) because of the total magnetic moment \( J = 0 \) of \( Eu^+ \) for among \( Ln_2Ir_2O_7 \) series, this compound is a sole and ideal platform for studying carrier transport in the background of all-in-all-out spin structure composed of the Ir\(^{4+}\) moment in \( J_{\text{eff}} = 1/2 \) state\(^2\). The results indicate that the magnetic domain structure is detected via peculiar asymmetric term in the magnetoresistance (MR) and zero-field offset in Hall resistance. These observations provide controllability of the exotic electronic phase in this compound towards accessing WSM.

**Results**

**Sample fabrication and structural property.** In this study (111)-oriented \( Eu_2Ir_2O_7 \) thin films were epitaxially grown on Y-stabilized ZrO\(_2\) (YSZ) (111) single crystal substrates by pulsed laser deposition (see Methods and Supplementary Fig. S1). Transmission electron microscopy (Figs. 1(c) and 1(d)) confirms the formation of a single-crystal \( Eu_2Ir_2O_7 \) film free from crystalline domain boundaries. The macroscopic crystal structure was examined by X-ray diffraction (XRD), with the high quality of the films supported by the observation of a typical rocking curve of 0.09° for the \( Eu_2Ir_2O_7 \) (222) peak (Fig. 1(e) and Supplementary Fig. S2). In Fig. 1(f), the reciprocal space mapping is shown, indicating that the lattice of the film is elongated along [111] direction by 0.7% with respect to in-plane lattices. The surface morphology was measured by atomic force microscope, which showed root mean square roughness of ~1 nm before and after post-growth annealing (Supplementary Fig. S3). The film thickness is fixed at about 70 nm.

**Longitudinal resistivity.** Figure 2(a) displays the temperature dependence of the longitudinal resistivity \( (\rho_{xx}) \). For electrical measurements, the sample was defined into a Hall-bar geometry (inset of Fig. 2(b)) to reduce mixing of the longitudinal and Hall resistances. A metal-insulator transition (MIT) is observed around transition temperature \( (T_M) \) of 105 K, which is close to the reported value for bulk (120 K, Ref. 24). The absolute values of \( \rho_{xx} \) and the strength of the MIT sensitively depend on the growth conditions (data for typical films are shown in Fig. S4(a)), likely as a result of film-dependent \( Eu/Ir \) nonstoichiometry. Since analytical determination on the composition of thin films is challenging, we estimate the composition of films by comparing the resistivity ratio \( \rho_{xx}(2 \text{ K})/\rho_{xx}(300 \text{ K}) \) with those of previously reported polycrystalline bulk data\(^3\). The result indicates that the films are Ir-rich by 1–4% (Fig. S4(b)). The reduction of \( T_M \) compared with the bulk may also be explained by the cation nonstoichiometry. Irrespective of quantitative variations in \( \rho_{xx} \), we qualitatively obtain the same magnetotransport properties discussed later for all the thin films. Hence, we focus on the most conducting film (sample 3 in Supplementary Fig. S4) in the structural and transport data presented. As also noted in Ref. 25 for

![Figure 1 | Crystal and spin structures of the pyrochlore lattice.](image-url)

(a) The Ir sublattice of \( Eu_2Ir_2O_7 \). Kagome and triangular lattices formed by Ir are located at orange and yellow planes, respectively. (b) Two distinct all-in-all-out spin structures, named as A-domain and B-domain, in the pyrochlore lattice. When four spins at tetrahedral vertices are consolidated at the centre of cube, they represent a magnetic octupole indicated by blue and red spheres. (c) Phase contrast image of high-resolution TEM of a \( Eu_2Ir_2O_7 \) (111) film on a YSZ (111) substrate. (d) Atomically resolved HAADF-STEM image at the \( Eu_2Ir_2O_7/YSZ \) interface. Triangular cross-sectional lattices composed of Ir and Zr are schematically shown. (e) \( \theta-2\theta \) scan of X-ray diffraction. Peaks from the substrate are marked with asterisks. (f) The reciprocal space mapping around the YSZ (331) peak. The peak position of bulk \( Eu_2Ir_2O_7 \) is indicated by an open triangle. The solid line indicates that \( Eu_2Ir_2O_7 \) is coherently grown on the substrate and expanded along [111] direction by 0.7%, which is illustrated by the pair of tetrahedra. An ideal pyrochlore lattice with cubic symmetry obeys the theoretical curve indicated by the dashed line.
behaviour remains controversial, we simply refer to this transition as the MIT in polycrystalline bulk samples, the temperature dependence of Hall coefficient \( R_H \) estimated by Hall measurement (see Methods) does not show significant anomaly across \( T_M \), in contrast to the sharp kink at MIT in \( \rho_{xx} \) as shown in Fig. 2(b). Although the origin of this behaviour remains controversial, we simply refer to this transition as the MIT.

**Magnetoresistance.** Magnetotransport experiments provide versatile information reflecting the orbital motion and spin states of electrons, as exemplified by the spin liquid recently detected by the zero-field anomalous Hall effect in Pr\(_2\)Ir\(_2\)O\(_7\) (Ref. 28). Figure 3(a) shows the out-of-plane MR at 2 K. The MR for zero-field cooling (left panel) shows a symmetric shape with respect to zero field with a roughly 0.5% decrease in resistance at high magnetic field - a typical response in magnetic materials. The sample cooled with an applied magnetic field, however, unexpectedly exhibits an asymmetric term in MR in addition to the symmetric term (right panel). Although the symmetric term is found to be independent of cooling field (Fig. S5), the sign of the asymmetric term is changed by inverting the polarity of the cooling field. We emphasize that this asymmetric term is an intrinsic property of magnetic materials. The sample cooled with an applied magnetic field, however, unexpectedly exhibits an asymmetric term in MR in addition to the symmetric term (Supplementary Fig. S6).

This observation appears peculiar as MR usually does not include an asymmetric term and is not dependent on the cooling field. Although magnetic metals or semiconductors show field asymmetric MR around zero field with a hysteretic behaviour due to a finite coercive field, it is symmetric when spins are flipped at high magnetic field. The MR observed in the data presented here can phenomenologically be understood by taking into account the all-in-all-out spin structure, based on the double exchange model as in the case of perovskite manganites. Here, the angle between the neighbouring spins dramatically affects the hopping probability, giving rise to MR\(^a\). For example, if A-domain is stabilized with cooling under +9 T, the spins on basal planes are canted towards the in-plane direction with positive magnetic field resulting in an increase in \( \rho_{xx} \) (top right inset). Conversely, a negative magnetic field tends to align those spins, giving rise to a lower \( \rho_{xx} \) (bottom left inset). The opposite response arises in the case of B-domain with −9 T field cooling. We have additionally checked the angle dependence of MR, which would reflect the crystallographic symmetry (Supplementary Figs. S9(a) and S9(b)). It turns out that the out-of-plane anisotropy is rather strong probably due to the structural shape anisotropy of the film and/or lattice distortion by epitaxial strain as is often the case of magnetic thin films (Supplementary Figs. S9(c) and S9(d)). Thus we utilize other methods for further verification as described in the following section.

**Temperature and cooling field dependence of the linear component in \( \rho_{xx} \).** To further explore this idea, the same measurements are systematically performed as a function of temperature and cooling field (see Methods). Here, we define a parameter \( x \) to describe the sign and the magnitude of the asymmetric term as \( \rho_{xx}(B) - \rho_{xx}(0)/\rho_{xx}(0) = \text{(even term)} + 2B \), since the odd terms equal to or higher than the third order are not significant (less than 1% of the linear term by fitting at 2 K). Figure 3(b) shows \( x \) as a function of cooling field at 2 K, indicating that \( x \) saturates above the cooling field of ±3 T. This bistability suggests that A-domain or B-domain may be selectively stabilized by applying a cooling field above ±3 T. The temperature dependence of \( x \) shown in Fig. 3(c) is also consistent with the above scenario as \( x \) disappears above \( T_M \). Although small hysteresis is observed around 60 K, the asymmetric term is clearly observed even in such a temperature region (Supplementary Fig. S7), indicating that one magnetic domain is dominant over the other.

**Hall measurements.** So far we have demonstrated that the MR contains a linear term reflecting the formation of a domain structure depending on the cooling field. We now show that the Hall resistance additionally captures the lattice distortion through the anomalous terms. \( \rho_{yx} \) is generally expressed as

\[
\rho_{yx} = R_H B + R_A M + \rho_{xy}^T, \tag{1}
\]

with \( R_H \) the ordinary Hall coefficient, \( R_A \) the Hall coefficient proportional to the macroscopic magnetization \( M \) as a result of spin-orbit interaction, and \( \rho_{xy}^T \) the topological term induced by spin chirality \( \chi = S_z (S_y \times S_x) \) (Ref. 30). Finite \( M \) and \( \chi \) may remain in the presence of uniaxial lattice distortion induced by epitaxial strain, while they should be cancelled out in the case of ideal all-in-all-out spin structure with cubic symmetry. Figure 4(a) shows the raw data of the Hall measurement at 2 K for the cooling magnetic fields of ±9 T. In addition to the ordinary B-linear component common in both measurements, the Hall resistance clearly exhibits a non-zero value at 0 T, as measured by the difference in \( \rho_{yx} \) between the opposite cooling magnetic fields (Δ\( \rho_{yx} \)). The overall linear term is reasonably assigned to the ordinary Hall effect (the first term in Eq. (1), since no anomaly is present across \( T_M \) as shown in Fig. 2(b). We can therefore ascribe Δ\( \rho_{yx} \) to the sum of the second and third terms in Eq. (1) as Δ\( \rho_{yx} \) disappears above \( T_M \) (Fig. 4(b)).

**Discussion**

As opposed to the standard field-symmetric MR typically observed in prevalent conductors, the magnetoresponse in the presence of all-in-all-out spin ordering is described in terms of a third rank tensor. Therefore, odd terms should characteristically be observed in response functions due to time-reversal symmetry breaking as proposed.
by Arima\textsuperscript{31}, which is a clear fingerprint of the all-in-all-out spin ordering. Here, conduction electrons must be sensitive to the local spin structure of the compound owing to spin-orbit coupling, known as the Kondo lattice model. Hence, our observation substantiates that MR reflects not only the magnetic spin structure but also the magnetic domain structure. From these considerations, the magnetic domain structure in Eu$_2$Ir$_2$O$_7$ is robust against a magnetic field of at least 9 T below $T_M$.

Figure 3 | Linear magnetoresistance mediated by all-in-all-out spin structure. (a) Magnetic field dependence of longitudinal resistivity $\rho_{xx}$ with $B//[111]$ at 2 K after zero field cooling (left panel) and $\pm 9$ T cooling (right panel). While the zero-field cooled data shows a symmetric response, $\rho_{xx}$ after field cooling includes the linear terms indicated by dashed lines. Magnetic responses of Ir$^{4+}$ magnetic moments corresponding to respective magnetic domain structures are schematically shown as insets, where spins are depicted as solid arrows and magnetic domains are symbolized by the pair of open and shaded triangles. The initial points and the direction of the magnetic field sweeps are indicated by open circles and open arrows, respectively, for each measurement. (b) Cooling field dependence of the linear magnetoresistance coefficient $\alpha$ at 2 K. $\alpha$ saturates above the cooling field of $\pm 3$ T, suggesting that magnetic domain be uniformly aligned to A-domain or B-domain. A multi-domain structure is formed by cooling in smaller magnetic fields. (c) Temperature dependence of $\alpha$ for representative cooling magnetic fields, showing that $\alpha$ appears only below the magnetic transition temperature $T_M$.

Figure 4 | Anomalies in Hall resistivity $\rho_{xy}$ originating from all-in-all-out spin structure. (a) Magnified view of magnetic field dependence of the Hall resistivity $\rho_{xy}$ with $B//[111]$ at 2 K after $\pm 9$ T field cooling. $\Delta \rho_{xy}$ is defined as the difference of $\rho_{xy}$ (0 T) between $+9$ T and $-9$ T field cooling. The scalar spin chirality induced by lattice distortion is schematically shown. The inset shows the data for the entire sweep range between $\pm 9$ T. (b) Temperature dependence of $\Delta \rho_{xy}$. The transition temperature $T_M$ is indicted by a dashed line.
\textit{Nd}_3\textit{Ir}_2\textit{O}_7 \text{ where A-domain and B-domain are thought to be switchable through the Ising character of Nd ions by sweeping magnetic field.} \text{ Therefore, it is plausible to attribute the robust domain structure to the } f = 0 \text{ character of Eu}^{2+}.

The anomalies found in \(\rho_{xx}\) are consistent with the interpretation of MR data in which conducting carrier reflects the magnetic domain structure in another way, and additionally indicate that the lattice distortion actually induces macroscopic \(M\) and \(\gamma\). An attempt to directly measure \(M\) by magnetometer is not successful since \(M\) of this compound is still too small to compare with the signal from the substrate, yet \(\rho_{xx}\) is sufficiently sensitive to detect it without involving the substrate. In addition to \(\Delta\rho_{yx}\), there exists a low-field nonlinear component (\(\rho_{NL}\)), which is deduced by subtracting the linear term and the zero-field offset \((\Delta\rho_{xy})\) as shown in Fig. S8. Remarkably, \(\rho_{NL}\) shows sign inversion between the two domains. If we assume that \(M\) is proportional to \(B\) as in the case of the bulk \(\text{EuO}\), \(\rho_{NL}\) may be therefore attributed to \(\rho^{yp}\) possibly originating from spins at the surface or the interface. Confirming thickness dependence of magnetotransport properties would be useful comparison to clarify this issue. However, clear dependence is not obtained probably due to slight fluctuation of the stoichiometry from sample to sample as mentioned above, while the qualitative features of MR and Hall effect are maintained. Although this sign inversion in \(\rho_{NL}\) is an intriguing character of this system, the details remain to be investigated including the possibility of band crossing as in the case of EuTiO\(_3\) (Ref. 33) or SrRuO\(_3\) (Ref. 34).

In conclusion, we have fabricated single crystal Eu\(_3\)Ir\(_2\)O\(_7\) thin films via solid phase epitaxy and systematically explored the magnetotransport properties originating from the all-in-all-out spin order. The MR contains a linear field dependent term while the Hall resistance has zero field offset and anomalous bending, both of which inherently depend on the type of domain formed. Our findings indicate that the all-in-all-out spin structure is controllable by the polarity of the cooling field and once formed is robust below \(T_{\text{N2}}\). The ability to realize this spin structure in thin film form will become the cornerstone to realizing WSM phases via tailored pyrochlore iridate heterostructures and/or utilizing electrostatic gating.

\textbf{Methods}

The Eu\(_3\)Ir\(_2\)O\(_7\) (111) films were prepared on commercial YSZ (111) single crystal substrates by pulsed laser deposition. Phase-mixed ceramic targets fabricated by hot-press method at 950 °C under 25 MPa pressure were used with a prescribed ratio of Eu/Ir = 1/3. The films were deposited at a substrate temperature of 500 °C in an atmosphere of 100 mTorr Ar gas containing 1% O\(_2\). A KrF excimer laser (\(\lambda = 248 \text{ nm} \)) was used for ablating the target, with a fluence and frequency of 6 J/cm\(^2\) and 10 Hz. The films were in an amorphous phase after the deposition, with the pure Eu\(_3\)Ir\(_2\)O\(_7\) character of Eu\(_3\)Ir\(_2\)O\(_7\) starting to appear after annealing in an electrical muffle furnace at 1000 °C for 1.5 hours in air.

The Hall bar structure was processed by conventional photolithography and Ar ion milling. Ohmic Ni (10 nm)/Au (50 nm) contacts were deposited by an electron-beam evaporator. Magnetotransport measurements were performed in a liquid He cryostat equipped with a 9 T superconducting magnet (PPMS, Quantum Design Co.). The ordinary Hall coefficient \(R_H\) is calculated by linear term in Hall resistance above \(\pm 5 \text{ T}\) region. Temperature dependence measurements for \(R_H\) (Fig. 3(c)) were carried out by abling the target, with a fluxence and frequency of 6 J/cm\(^2\) and 10 Hz. The Hall resistance was annealed after the deposition, with the pure Eu\(_3\)Ir\(_2\)O\(_7\) phase appearing after annealing in an electrical muffle furnace at 1000 °C for 1.5 hours in air.

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Author contributions
T.C.F. performed the sample fabrication, measurements, and analysis. Y.K. assisted with the sample fabrication, measurements, and analysis. M.U., A.T. and T.A. assisted with the planning and analysis. M.K. directed the project.

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