Supplementary Materials for

Above-ordering-temperature large anomalous Hall effect in a triangular-lattice magnetic semiconductor

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S1  Structural characterization of EuAs films

Figure S1 illustrates epitaxial relation between EuAs and Al₂O₃. As shown in top views of their hexagonal crystal structures in Figs. S1A and S1B, lattice mismatch between them is expected to be only 1.1%. Reflection high-energy electron diffraction (RHEED) images taken for the EuAs film and the Al₂O₃ substrate in Figs. S1C and S1D confirm this relation. Clear streak patterns also indicate the two-dimensional growth of the EuAs film.

This epitaxial relation is more precisely examined by x-ray diffraction (XRD) measurements. Out-of-plane θ-2θ scan in Fig. S2A only shows reflections from the EuAs {001} lattice planes without any impurity peaks. A rocking curve of the (002) EuAs film peak in Fig. S2B is also very sharp, ensuring high quality of the obtained film. In a reciprocal space map in Fig. S2C, not the (226) but the (306) film peak is observed around the (1112) substrate one. This indicates that the a- and b-axes of the EuAs film are rotated by 30 degrees compared to the Al₂O₃ substrate ones, as shown in Figs. S1A and S1B, and that there are no crystal domains. In-plane φ-scan of the (306) peak in Fig. S2D also shows six-fold symmetry.

Figure S3A shows a cross-sectional transmission electron microscopy (TEM) image of the sample. EuAs is rather sensitive to air and its surface is readily oxidized. Actually, an oxidized layer different in appearance is formed on the surface and a magnified view of the inner layer, taken by high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM), reveals atomic arrangement consistent with the EuAs crystal structure, as shown in Fig. S3B. Electron energy loss spectroscopy (EELS) in Fig. S3C was also performed for measuring the Eu valence state in each layer. In comparison to EELS spectra previously reported for Eu²⁺ and Eu³⁺ oxides in Fig. S3D (44), the Eu valence in the inner EuAs layer is determined to be Eu²⁺ \((J = S = L = 0)\), also consistent with the observed magnetic properties. In contrast, valence in the oxidized surface layer is almost Eu³⁺ \((J = 0, S = L = 3)\), suggesting possible degradation to Eu₂O₃. This oxidized layer does not affect the transport and magnetization measurements, because it is insulating and nonmagnetic.

Atomic displacement corresponding to the Eu distorted triangular lattice can be directly vi-
sualized in a further magnified HAADF-STEM image and energy dispersive x-ray spectrometry (EDX) maps in Fig. S4. Reflecting the formation of $[\text{As-As}]^4^-$ dimers in the short As-As bonds as in SrAs (30, 31), Eu atoms are slightly displaced on the $a$-$b$ plane as also confirmed in Fig. 1A.
Fig. S1. Epitaxial relation between EuAs and Al₂O₃. Crystal structures of (A) EuAs film and (B) Al₂O₃ substrate, viewed along the out-of-plane c-axis. Each in-plane unit cell is indicated by the rhombus and the epitaxial relation is represented by the dashed hexagon. Typical RHEED images of (C) the EuAs film and (D) the Al₂O₃ substrate, taken after and before the film growth, respectively.
**Fig. S2. Detailed XRD characterization.** (A) XRD $\theta$-2$\theta$ scan of a single-crystalline EuAs film grown on the Al$_2$O$_3$ (0001) substrate. (B) Rocking curve of the (002) film peak. (C) Reciprocal space map around the (1112) substrate peak. A cross denotes film peak positions calculated from the EuAs lattice parameters. Observation of not (226) but (306) film peak indicates that the film is stacked on the substrate as shown in Figs. S1A and S1B. (D) In-plane $\phi$-scan of the (306) film peak.
**Fig. S3. EELS for determining the Eu valence state.** (A) Cross-sectional bright-field TEM image of the sample. (B) Atomically resolved image of the EuAs film, taken by HAADF-STEM for the dashed square area in (A). A EuAs unit cell is represented by the rectangle. (C) EELS spectra taken for the Eu N_{4,5}-edge at 5 points shown in (A). (D) EELS spectra of the Eu^{2+} state in EuO and the Eu^{3+} state in Eu_{2}O_{3}, reproduced from Ref. 44.
**Fig. S4. Direct observation of atomic arrangement.** (A) Higher resolution HAADF-STEM image of the EuAs film. A EuAs unit cell is represented by the rectangle. (B) EuAs crystal structure determined by x-ray diffraction measurements (30,31). There are short (blue) and long (red) As-As bonds along the c-axis. Corresponding element maps taken with EDX for (C) Eu L and (D) As K edges.
Analysis of anomalous Hall effect

Magnetization ($M$)-nonproportional anomalous Hall component $\sigma_{\text{AHE,nonM}}$ is quantitatively estimated by using the following equation

$$\sigma_{yx} = \sigma_{\text{OHE}} + \sigma_{\text{AHE,M}} + \sigma_{\text{AHE,nonM}}.$$  \hfill (S1)

Here, $\sigma_{\text{OHE}}$, $\sigma_{\text{AHE,M}}$, and $\sigma_{\text{AHE,nonM}}$ are ordinary Hall conductivity, $M$-nonproportional anomalous Hall conductivity, and $M$-proportional anomalous Hall conductivity. $\sigma_{\text{OHE}}$ and $\sigma_{\text{AHE,M}}$ can be calculated by

$$\sigma_{\text{OHE}} = \frac{-1p\varepsilon B}{\rho_{xx}^2 + \left(\frac{1p\varepsilon B}{2}\right)^2}.$$  \hfill (S2)

and

$$\sigma_{\text{AHE,M}} = -A\alpha_{\text{xx}} M.$$  \hfill (S3)

Here $p$ is the carrier density with plus sign for hole, $A$ is a constant independent of magnetic field and temperature, and $\alpha$ is a scaling factor relating the longitudinal and anomalous Hall conductivities. In the analysis in the main text, $\sigma_{\text{AHE,M}}$ is calculated using $\alpha = 1.6$. While this value is experimentally reasonable for $\sigma_{xx} < 10^4 \ \Omega^{-1}\text{cm}^{-1}$ independent of conduction mechanisms (1), it has been also theoretically demonstrated that $\alpha$ is in the range of $1 < \alpha < 2$ for the incoherent conduction regime (1,45) and $1.33 < \alpha < 1.76$ for the hopping conduction one (28). We thus check the subtraction process using different scaling factors. As confirmed in Fig. S5, the variation in $\alpha$ does not have a significant effect on the estimation of the $M$-nonproportional anomalous Hall component. This is because $\sigma_{\text{AHE,M}}$ is much smaller than $\sigma_{\text{OHE}}$ and $\sigma_{\text{AHE,nonM}}$, ensuring that the large anomalous Hall angle obtained for EuAs is not an artifact caused in the subtraction process.

In addition, $\sigma_{\text{OHE}}$ is determined under the assumption that it is the only contribution to the remaining component at 56 T. Although $\sigma_{\text{AHE,nonM}}$ may still slightly contribute at the high field, this assumption gives the most conservative estimate of $\sigma_{\text{AHE,nonM}}$. Discrepancy of peak positions between $\rho_{\text{AHE,nonM}}$ and $\sigma_{\text{AHE,nonM}}$ is mainly due to negative magnetoresistance. As
\( \rho_{xx} \) monotonically decreases with increase of field, the peak shifts to a higher field in the conductivity plot. As shown in Fig. S6, we check temperature dependence of the carrier density \( p \) obtained from \( \sigma_{OHE} \) by Eq. S2. The estimated carrier density is nearly constant around \( p \approx 3 \times 10^{17} \text{ cm}^{-3} \).

We also examined other types of fittings which could be applied for explaining the nonmonotonic Hall signal observed in EuAs. We first checked whether it can be fitted by assuming two types of carriers with different carrier densities and mobilities. As shown in Fig. S7, a small hump observed at 150 K can be well fitted by the two-carrier model, but the fitting substantially deviates from the observed curves when the hump is enhanced at lower temperatures. Moreover, the mobility of one carrier estimated at low temperatures exceeds \( 10^3 \text{ cm}^2/\text{Vs} \), which is unphysically high for the hopping conduction regime. Therefore, it is reasonable to rule out the two-carrier conduction as an origin of the nonmonotonic Hall signal.

We also checked whether the nonmonotonic signal can be fitted without considering the \( M \)-nonproportional anomalous Hall component. As confirmed in Fig. S8A, the fitting composed only of \( \sigma_{OHE} \) and \( \sigma_{AHE,M} \) deviates from the observed curve especially at low magnetic fields, even although \( A \) and \( \alpha \) are treated as free parameters. Moreover, these fitting parameters unreasonably change with temperature, as shown in Figs. S8B-S8D. The carrier number is largely scattered and its sign is inverted three times, being independent of the magnetic phase transition at 23 K. \( A \) and \( \alpha \) also largely vary with temperature, while they should be nearly constant \((1, 28, 45)\). These fitting results also strongly support the existence of an \( M \)-nonproportional anomalous Hall component.
Fig. S5. Component separation using different scaling factors. Hall conductivity $\sigma_{yx}$ observed at 70 K and its separation to ordinary Hall conductivity $\sigma_{OHE}$, $M$-proportional anomalous Hall conductivity $\sigma_{AHE,M}$, and $M$-nonproportional anomalous Hall conductivity $\sigma_{AHE,nonM}$ in the cases of different scaling factors $\alpha = (A) 1.0$, (B) 1.6, and (C) 2.0 in Eq. S3.
Fig. S6. Temperature dependence of the estimated carrier density. Carrier density obtained from the ordinary component is almost constant with temperature.
Fig. S7. Two-carrier analysis of Hall resistivity. Two-carrier fitting of the Hall resistivity taken at (A) 150, (B) 90, (C) 70, and (D) 60 K. A dashed curve represents the fitting assuming two types of carriers with listed density and mobility.
Fig. S8. Fitting without considering \( M \)-nonproportional component. (A) Typical fitting of the Hall resistivity when only considering the ordinary and \( M \)-proportional anomalous components. (B) Temperature dependence of carrier density obtained by this fitting process. The carrier number is largely scattered and its sign is inverted three times. Temperature dependence of (C) coefficient \( A \) and (D) scaling factor \( \alpha \) in Eq. S3. Both should not greatly vary with temperature and the latter is expected to be in the range of \( 1 < \alpha < 2 \) (1, 28, 45).
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