Sign-tunable anisotropic magnetoresistance and electrically detectable dual magnetic phases in a helical antiferromagnet

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Abstract
Emerging from competing exchange interactions, the helimagnetic order describes a noncollinear spin texture of antiferromagnets. Although collinear antiferromagnets act as the elemental building blocks of antiferromagnetic (AFM) spintronics, until now, the potential of implementing spintronic functionality in noncollinear antiferromagnets has not been clarified. Here, we propose an AFM helimagnet of EuCo$_2$As$_2$ as a novel single-phase spintronic material that exhibits a remarkable sign reversal of anisotropic magnetoresistance (AMR). The contrast in the AMR arises from two electrically distinctive magnetic phases with spin reorientation that is driven by the magnetic field prevailing in the easy plane, which converts the AMR from positive to negative. Furthermore, based on an easy-plane anisotropic spin model, we theoretically identified various AFM memory states associated with the evolution of the spin structure under magnetic fields. The results revealed the potential of noncollinear antiferromagnets for application in the development of spintronic devices.

Introduction
The advancements in the research of antiferromagnetic (AFM) spintronics have opened new avenues for spin-based devices. Generally, collinear antiferromagnets are utilized as the elementary basis for spintronic functionalities. However, recent examinations of intriguing and unexpected physical phenomena in noncollinear antiferromagnets have expanded the scope of potential materials for exploiting AFM spintronics. In particular, a large anomalous Hall effect was observed in noncollinear antiferromagnets, despite the vanishingly small magnitude of magnetization. The Hall effect originates from the Berry curvature associated with topologically nontrivial spin textures. Although its experimental observation is challenging, recent magnetic imaging techniques such as single spin relaxometry and scanning thermal gradient microscopy can be used for imaging noncollinear AFM textures and domain structures.

The ability to control and detect AFM memory states is imperative for AFM spintronics, and accordingly, magnetocrystalline anisotropy has been exploited as a generic foundation for manipulating AFM states. As controlled anisotropy provides an exceptional opportunity for extensive spintronic applications, anisotropic magnetoresistance (AMR) has been adopted to detect the various resistive states associated with crystal axes. Nonetheless, in several cases, an intricate stacking geometry with additional reference layers is required to achieve unified spintronic functionality.

Helimagnets exhibit a prototypical noncollinear spin structure in which the spin direction is spatially rotated in the plane, whereas the rotation axis is parallel to the propagation direction. In principle, helimagnets are noncollinear antiferromagnets that correspond to the zero...
net moment inherent in rotating spins. Thus, helimagnets offer the same advantages as antiferromagnets, such as the absence of a stray field and the features of intrinsically fast spin dynamics.\textsuperscript{27–29} Despite these merits, a crucial problem with noncollinear AFM spintronics is the establishment of controllable factors and mechanisms for the anisotropy of noncollinear spin configurations. Specifically, the lack of a comprehensive understanding of AMR affects its control and application in spintronic devices. In this research, we demonstrate that the helix-to-fan transition induces a sign reversal of the AMR in a helical antiferromagnet of EuCo\textsubscript{2}As\textsubscript{2} (ECA). Moreover, we verified the electrically discernible dual magnetic phases producing the alterable sign of the AFM memory state through both experimental and theoretical methods. In addition, we identified diverse AFM memory states relevant to the development of the spin structure under magnetic fields, which facilitates AFM spintronics based on noncollinear antiferromagnets.

**Materials and methods**

**Sample preparation**

The ECA single crystals were grown following the flux method with Sn flux.\textsuperscript{30} Eu (99.9%, Alfa Aesar), Co (99.5%, Alfa Aesar), As (99.999%, Sigma Aldrich), and Sn (99.995%, Alfa Aesar) were mixed at a 1.05:2:2:15 molar ratio of Eu:Co:As:Sn. The mixture was placed in an alumina crucible sealed in an air-evacuated quartz tube. The quartz tube was placed in a high-temperature furnace at 1050°C for 20 h and thereafter gradually cooled to 600°C at a rate of 3.75°C/h. At 600°C, the incorporation of the remaining Sn flux in the crystals was prevented by using a centrifuge composed of stainless steel, while the quartz tube was cooled to room temperature. Ultimately, we obtained crystals with typical dimensions of 1.5 × 1.5 × 0.1 mm\textsuperscript{3}.

**Scanning transmission electron microscopy (STEM) measurement**

We prepared ECA samples with a cutting plane perpendicular to the \textit{a}-axis utilizing a dual-beam focused ion beam system (Helios 650, FEI). Along the cutting plane, the images presented a well-discernible atomic structure. To minimize any damage to the sample, the acceleration voltage conditions were gradually reduced from 30 to 2 keV. In addition, dark-field images were obtained using STEM (JEM-ARM200F, JEOL Ltd., Japan) at 200 keV with a Cs-corrector (CESCOR, CEOS GmbH, Germany) and a cold field-emission gun. The size of the electron probe was 83 pm, and the range of the high-angle annular dark-field detector angle was varied from 90 to 370 mrad.

**Magnetic and transport property measurements**

The dependence of magnetization on temperature and magnetic field was determined with the magnetic fields along the \textit{a}- and \textit{c}-axes using a vibrating sample magnetometer module in a physical property measurement system (PPMS, Quantum Design, Inc.). The electric transport measurements were performed using the conventional four-probe method in the PPMS. The AMR was measured by rotating the magnetic field in the \textit{ac} plane in a PPMS equipped with a single-axis rotator.

**Theoretical calculations**

The easy-plane anisotropic spin model can be expressed as

\[
\mathcal{H} = J_1 \sum_{i=1}^{5} \mathbf{S}_i \cdot \mathbf{S}_{i+1} + J_2 \sum_{i=1}^{5} \mathbf{S}_i \cdot \mathbf{S}_{i+2} - g\mu_B H \cdot \sum_{i=1}^{5} \mathbf{S}_i + K_\theta \cos^2 \theta_i - K_5 S(1 + \sin^2 \theta) \sum_{i=1}^{5} \cos 5\phi_i,
\]

where \(N\) denotes the number of Eu\textsuperscript{2+} moments in a single layer; \(J_1\) and \(J_2\) represent the AFM coupling strength between the Eu\textsuperscript{2+} moments of the two nearest layers and the next-nearest layers, respectively; \(S = 7/2\) for Eu\textsuperscript{2+} ions; \(g = 2\); and \(K_\theta\) denotes the magnetocrystalline anisotropy constant.\textsuperscript{31} We considered the helical spin structure of the ECA as commensurate \((k = 0.8)\) for the convenience of calculation and thus included only five layers with the periodic boundary condition. The first and second terms indicate the competing exchange interactions for an AFM helical state. The third term designates the Zeeman energy, where the magnetic field \(H\) is situated on the \textit{ac} plane, forming an angle \(\theta\) with the \textit{c}-axis. The fourth term denotes the easy-plane magnetocrystalline anisotropy energy that favors the planar spin orientation. The fifth term reflects that we are working with a three-dimensional system, considering the ferromagnetic interactions within a given layer as a mean-field term. For the helical AFM state, where \(\theta = \frac{2}{5}\pi\), i.e., \(J_2 = 0.31J_1\), we estimated the following parameters by fitting the theoretical results of anisotropic magnetization to the experimental data: \(g\mu_B H_m/J_1 S = 1.18\), \(K_\theta = 0.35J_1S^2\), and \(K_5 = 0.022J_1S\), where \(H_m\) indicates the occurrence of the helix-to-fan transition. Using \(H_m = 4.7\) T for the ECA at 2 K, \(J_1S^2\) and \(K_\theta\) can be evaluated as 5.45 × 10\textsuperscript{6} and 1.96 × 10\textsuperscript{6} J/m\textsuperscript{3}, respectively.

**Results**

**Structure and properties of helical ECA antiferromagnets**

A two-dimensional (2D) layered ECA helimagnet forms a body-centered tetragonal structure (I4/mmm space group).\textsuperscript{30} The crystal exhibits a strong 2D nature that enables it to be mechanically exfoliated. By composition, it consists of two Co\textsubscript{2}As\textsubscript{2} layers on opposite sides of a unit cell, separated by a magnetic Eu layer, as depicted in Fig. 1a.\textsuperscript{32} As crystal quality is an essential aspect for investigating distinguished anisotropy, it was reviewed.
using the single-crystal X-ray diffraction technique. The analyses revealed the crystals as a single phase with high quality (refer to Supplementary S1). The magnetic moments of the Eu\(^{2+}\) ions \((S = 7/2\) and \(L = 0\)) were helically ordered, whereas those of the Co ions were paramagnetic and not relevant to the magnetic ordering\(^{30,33}\). The direction of the net moment in the Eu layer rotates in the \(ab\) plane and propagates along the \(c\)-axis (Fig. 1a). In general, the pitch of the helix was incommensurate with the lattice parameter, and thus, no two layers exhibited the same directions of the net moments\(^{34}\). In the ECA, a slightly incommensurate helimagnetic order with a propagation vector \(k = (0, 0, 0.79)\) has been observed via neutron diffraction\(^{32}\), which indicates that the AFM interaction between the adjacent layers is perturbed by an additional AFM interaction acting between the second-neighbor layers\(^{31}\). Accordingly, STEM was utilized to visualize the structural units of alternately arranged Eu and Co\(_2\)As\(_2\) layers (Fig. 1b). The STEM image with lower magnification indicates that all the layers were regularly aligned. The lattice constants were obtained using fast Fourier transformation (FFT) from the STEM data with \(a = 0.402\) nm and \(c = 1.150\) nm, similar to the results of previous reports\(^{30,32}\).

The helical AFM order emerges at \(T_N = 46\) K, as evident from the temperature \((T)\) dependence of the magnetic susceptibility defined by the magnetization \((M)\) divided by the magnetic field \((H)\), \(\chi = M/H\), measured at \(H = 0.1\) T (Fig. 1c). The \(\chi\) curves below \(T_N\) for \(H\) along the \(a\)- and \(c\)-axes \((H_a\) and \(H_c)\) indicate an anisotropic nature. The rapid decrease in \(\chi\) for \(H_a\) below \(T_N\) is consistent with the alignment of the magnetic moments of Eu ions along the \(ab\) plane, as previously verified via neutron diffraction and nuclear magnetic resonance experiments\(^{32,33}\). We observed metallic behavior with a distinct anomaly at \(T_N\), as shown by the variations in resistivity with \(T\) (refer to Supplementary Fig. S2). Therefore, an indirect Ruderman–Kittel–Kasuya–Yosida (RKKY) exchange interaction between the Eu\(^{2+}\) spins mediated by the spins of conduction electrons can be expected\(^{35}\). The phase diagram of the \(T\) and \(H_a\) dependences of the magnetic properties is illustrated in Fig. 1d, which clarifies the phase boundary between the helix and fan structures.

**Electrically distinguishable dual magnetic phases and magnetoresistance anisotropy**

In an antiferromagnet, an adequately strong \(H\) along a magnetic easy-axis often generates a magnetic phase transition through spin reorientation, such as a spin-flop or spin-flip transition\(^{30,36}\). Phase conversion occurs with marked anomalies in the physical properties and stabilizes the flopped or flipped state. We observed a similar magnetic phase transition in the helimagnetic ECA. At 2 K, \(M_a\) (\(M\) along the \(a\)-axis) abruptly increases at \(H_m = 4.7\) T,
thereby indicating a helix-to-fan transition\textsuperscript{30}. The similarity of this transition to spin-flops is signified by the extrapolation of the linear slope above $H_m$ that merges at the origin (Fig. 2a). $H_m$ was determined from the peak in the derivative of $M_a$. As displayed in the inset of Fig. 2a, slight magnetic hysteresis is observed to arise from the first-order characteristic of this transition (refer to Supplementary Information S2). Across the phase transition, a fan structure emerges in which the net magnetic moments between the most proximate layers at zero magnetic fields greater than $H_c$ (Fig. 2c). In contrast, $M_c$ ($M$ along the $c$-axis) at 2 K displays a linear increase associated with the gradual canting of the net moments (Fig. 2c). The slope of $M_a$ is smaller than that of $M_c$ below $H_m$ and becomes larger in magnetic fields greater than $H_m$. Therefore, the value of $M_a$ exceeds $M_c$ at $H_m$.

An easy-plane anisotropic spin model was adopted to investigate the evolution of the helix-to-fan phase. The model Hamiltonian comprises the competing exchange interactions, Zeeman energy, magnetocrystalline anisotropy, and mean-field terms (refer to Methods and Supplementary Information S2 for details). Additionally, magnetocrystalline anisotropy terms (refer to Methods and Supplementary Information S2 for details). Additionally, the commensurate helical spin structure was considered for convenience of calculation. The relative angle of the two moments between the most proximate layers at zero $H$ was $\phi = \frac{4}{5} \pi$, corresponding to $j_2 = 0.31j_1$, because the relative angle of the two spins for a helical order can be expressed as $\phi = \cos^{-1}\left(-\frac{1}{4}\right)$\textsuperscript{31,34}. As the planar spin rotation is explicitly disintegrated with the application of $H_a$, the order parameter of the helical state can be expressed as $\sum_{i=1}^{5} \cos 5\phi_i$. Thus, the ferromagnetic interactions within a given layer can be treated by adding a mean-field term. The estimated $M_a$ and $M_c$ values obtained from the experimental data are represented as dotted curves in Fig. 2a, c. The well-matched fitting in the absence of a fourfold rotational magnetocrystalline anisotropy in the $ab$ plane implies the formation of helical spins that are independent of the planar crystalline axes in the ECA. Moreover, the theoretical estimation directly generated the spin configurations pertaining to the helix and fan phases, as illustrated in Fig. 2e, f. In the helix phase, the orientations of the net moments continually turned in the $H$ direction as $H_a$ increased. Greater than $H_m$, spin reorientation occurred in such a manner that the moments acting farther from the $H$ direction tended to be perpendicular. A further increase in $H_a$ generated an additional canting of the magnetic moments.

The influence of anisotropic $M$ on transport was examined based on the magnetoresistance, $MR = \frac{R(H) - R(0)}{R(0)}$, for both the $a$- and $c$-axes ($MR_a$ and $MR_c$). The increase in $M_a$ with increasing $H$ below $H_m$ (Fig. 2a)
indicates a partial and gradual alignment of the magnetic moments in the $H$ direction. The average net magnetic moment along the $H$ direction disrupts the equally distributed angles between the moments in the layers (Fig. 2e). The spin configuration away from the helix state increases the resistance, i.e., a positive $MR_a$ (Fig. 2b). With a further increase in $H$, $MR_a$ exhibits an abrupt reduction with the maximum slope at $H_m$ and becomes negative. The spin-reorientation-driven switching from positive to negative $MR_a$ is consistent with the crossing behavior of $M_a$. In the high-$H$ regime, $MR_a$ decreases faster with the additional canting of the moments toward the flipped state (Fig. 2f). In contrast, $MR_c$ decreases with increasing $H$ (Fig. 2d). This intimate correlation between the $M$ and MR plots suggests that the magnetic order governs the magnetotransport and its anisotropy. As $T$ increases, $H_m$ decreases, and the shape of the anomaly is broadened, as displayed in the anisotropic $M$ and MR plots at various $T$ values in Supplementary Fig. S6.

To theoretically probe the magnetotransport property, we propose that the interlayer hopping amplitude can be expressed by $t_{i,i+1} = t_0 + t_s \langle \langle \hat{n}_i | \hat{n}_{i+1} \rangle \rangle$ ($i = 1–5$) with the periodic boundary condition, where $t_0$ and $t_s$ indicate the spin-independent and spin-dependent parts, respectively. Here, $\langle \langle \hat{n}_i | \hat{n}_{i+1} \rangle \rangle$ denotes the overlap integral between the two spinors. Each spinor aligns with the direction of the net magnetic moment in each layer. The overlapping integral is given by $\cos \gamma$, where $\gamma$ denotes the relative angle between the two spinors. For the MR calculations, the conductance ($\sigma$) of the system was assumed to be proportional to the geometric mean of multiple hopping amplitudes through the layers as $\sigma \propto \left( \prod_{i=1}^{6} t_{i,i+1} \right)^{1/6}$, where the geometric mean effectively presumes the average of the product $\prod_{i=1}^{6} t_{i,i+1}$. According to the definition of MR and $R = 1/\sigma$, MR is proportional to $\sigma(\sigma) - \sigma(H)$. Upon setting $t_0/t_s = 0.2$, the highly anisotropic trend between $MR_a$ and $MR_c$ is moderated by the purely spintronic consideration of $\sigma$ (Fig. 3a, b). This is because of the $L = 0$ condition for the ground state of the Eu$^{2+}$ ions. The MR response to an applied $H_a$ prominently distinguished two distinct magnetic phases. In the helix phase, the increased $\gamma$ for certain adjacent layers at $H_a$ below $H_m$ contributes more to $\sigma$ as it reduces, thereby

![Fig. 3 Calculated magnetoresistance anisotropy.](image)
inducing positive MR$_a$. For small $H_a$, one can explicitly prove that $\sigma$ decreases with $H_a$. In this regime, an inter-layer hopping amplitude can be approximately expressed as $t_{i+1} = \bar{t} e^{-\gamma_1/2 - \beta z_1/4}$, where $\gamma_1 = \varphi_{i+1} - \varphi_i - \pi/3$ represents the small deviation of the relative spin orientation $\varphi_{i+1} - \varphi_i$ from the average value, $\bar{t} = t_0 + t_0 \cos \frac{2\pi}{3} \alpha = t_0 \sin \frac{2\pi \theta}{3}$, and $\beta = t_0 (t_0 \cos \frac{2\pi}{3}) / 2 \pi^2$. As $\sum_{i=1}^5 \gamma_1 = 0$, $\sigma \propto (\prod_{i=1}^5 t_{i+1})^{1/2} \bar{t} e^{-\beta \sum_{i=1}^5 \gamma_1 / 20} < \bar{t}$. Therefore, $\sigma$ reduces for small $H_a$. In the fan phase, $\gamma$ continuously decreases with a further increase in $H_a$ by approaching the completely spin-aligned state along the magnetic field direction, which enhances $\sigma$ and diminishes MR$_a$. Various AFM memory states relevant to the various spin configurations under an external $H_a$ were theoretically identified, as depicted in Fig. 3a.

Spin-reorientation-driven reversal of anisotropic magnetoresistance

A peculiar spintronic characteristic of a noncollinear AFM ECA is presented by angle-dependent magnetotransport. The results of the AMR, defined as $\frac{R(\theta) - R(0)}{R(0)}$, are depicted in Fig. 4a, b. As indicated in the geometry of the AMR measurement in Fig. 4a, $H$ is continually rotated perpendicular to the current, excluding the extrinsic Lorentzian MR effect. At $H < H_{opt}$, $ab$-planar helix formation facilitates twofold rotational symmetry. The AMR is maximized at $\theta = 90^\circ$ and $270^\circ$ (Fig. 4a) owing to the positive value of MR$_a$, which gradually increases with $H$ (Fig. 2b). At $H$ values slightly exceeding $H_{opt}$, the AMR near $\theta = 90^\circ$ and $270^\circ$ starts to partially reverse, which is observed as a dip in Fig. 4a. A further increase in $H$ results in a complete reversal of the AMR. The AMR contour map (Fig. 4b) demonstrates the sign-tunable AMR upon crossing $H_{opt}$. The detailed influence of increasing $T$ on the AMR is plotted in the contour maps of Supplementary Fig. S7. The overall trend of AMR development under the applied $H$ was reproduced using theoretical calculations, as portrayed in Fig. 4c, d. The contrast emerging from the reversal behavior of the AMR effect reflects the intrinsic bulk properties and clarifies the different magnetotransport features between the helix and fan structures.
Discussion
In this study, we have demonstrated a mechanism to generate the sign-changing AMR phenomenon that originates from electrically distinctive dual magnetic phases separated by a magnetic phase transition. The magnetic transition emerges from the competition of diverse energy scales included in the model Hamiltonian. Specifically, the highly 2D characteristic of magnetocrystalline anisotropy is vital for determining the consistency between bulk measurements and theoretical calculations. In pursuance of the AMR effect predominantly caused by the magnetocrystalline anisotropy in AFM spintronics, the approach proposed in this study is applicable to a variety of antiferromagnets in which the distinct features of the anisotropic magnetotransport could be observed depending on the alteration of the magnetocrystalline anisotropy. The development of magnetic devices with the desired properties requires detectable macroscopic effects that are contingent on the variable magnetic states. In complex noncollinear antiferromagnets, more spintronic effects can exist. Therefore, the electrical access to various AFM memory states associated with the evolution of helical spin texture would provide an opportunity for realizing multilevel AFM memory devices.

Notably, the reversal phenomenon of AMR has seldom been reported. In a previous study on ferromagnetic La0.7Ca0.3MnO3 ultrathin films, a sign reversal of the AMR originated from the planar tensile strain that facilitated the rotation of the ferromagnetic easy axis, which is different from the present case.37 The sign-tunable AMR effect in a single-phase noncollinear antiferromagnet is unique owing to its intrinsic origin from the electrically discernible magnetic phases. Additionally, a variety of topological states mediated by spin-orbit interactions yield exotic topological magnetism.38–40 More recently, helical magnetism driven by Weyl-mediated RKKY interactions was observed in a Weyl semimetal, NdAl5Si41. The recognition of versatile AFM memory states in a helimagnet offers valuable guidelines for investigating the intimate interplay between the electronic and magnetic topological properties and thus for implementing topological AFM spintronics.

In summary, we propose a new single-crystalline spintronic material in which the sign-tunable AMR effect reflects completely intrinsic bulk properties. These results are beneficial for the development of AFM spintronics, which has been driven by novel materials. Theoretically, the highly anisotropic 2D spin characteristic was highlighted as a core factor for anisotropic magnetotransport in a natural noncollinear AFM ECA. The scheme followed in this study is a particular type of spin-reorientation-driven mechanism to study the intriguing anisotropic properties, which can motivate further investigations of noncollinear AFM for extensive spintronic applications.

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Author contributions
N.L. and Y.J.C. conceived the study. J.H.K. and JMH. synthesized the single crystals. J.H.K, H.J.S., J.M. H, K.W.J, and J.S.K. measured the physical properties of the crystals. J.H.K. and H.J.S. acquired the STEM images. M.K.K. and K.M. performed the theoretical calculations. J.H.K., H.J.S., M.K.K., KM., N.L., and Y.J.C. analyzed the data and prepared the manuscript. All authors have read and approved the final version of the manuscript.

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