Unusual Electrical and Magnetic Properties in Layered EuZn$_2$As$_2$

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Eu-based compounds often exhibit unusual magnetism, which is critical for nontrivial topological properties seen in materials such as EuCd$_2$As$_2$. The authors investigate the structure and physical properties of EuZn$_2$As$_2$ through measurements of the electrical resistivity, Hall effect, magnetization, and neutron diffraction. Their data show that EuZn$_2$As$_2$ orders antiferromagnetically with an A-type spin configuration below $T_N = 19$ K. Surprisingly, there is strong evidence for dominant ferromagnetic fluctuations above $T_N$, as reflected by positive Curie–Weiss temperature and extremely large negative magnetoence (MR) between $T_N$ and $T_H \approx 200$ K. Furthermore, the angle dependence of the MR$_{ab}$ indicates field-induced spin reorientation from the $ab$-plane to a direction $\approx 45^\circ$ from the $ab$ plane.

Compared to EuCd$_2$As$_2$, the doubled $T_N$ and $T_H$ make EuZn$_2$As$_2$ a better platform for exploring nontrivial magnetic and electronic properties in both magnetic fluctuation ($T_N < T < T_H$) and ordered ($T < T_N$) regimes.

1. Introduction

Since the discovery of topological states in semimetals$^{[1–6]}$ the search for new topological materials has been extremely active. Time-reversal symmetry ($T$) and crystal inversion symmetry ($P$), together with the Kramers theorem (each energy band is doubly degenerate for fermions), are crucial in understanding the formation of topological states in semimetals$^{[1]}$. When both $T$ and $P$ are preserved, a system may host Dirac fermions at the Dirac points (DP), at which bands are quadruply degenerate. Several Dirac semimetals have been identified, including Cd$_3$As$_2$,$^{[2]}$ Na$_3$Bi,$^{[3]}$ and ZrTe$_5$. When $T$ is broken but $P$ is preserved, one may expect two Weyl cones with opposite chirality in a Weyl semimetal. Considering the opposite situation, that is, $T$ is preserved but $P$ is broken, the number of Weyl cones is a multiple of four, removing the chirality.$^{[1]}$ Weyl states have been observed in semimetals such as MoTe$_2$, TaAs,$^{[7]}$ WTe$_2$, and TaIrTe$_4$. When $T$ and $P$ are both broken, a system is a semimetal, in which Dirac points (DP) are preserved, one may expect two Weyl cones with opposite chirality in a Weyl semimetal.

Recently, materials with the type-IV magnetic space group have also been studied.$^{[11]}$ The type-IV magnetic space group is defined by $G + T(e)[\tau]G$, where $G$ is the ordinary nonmagnetic space group, $e$ is the identity operation, and $[\tau]G$ represents the translation operation between spin-up and spin-down sublattices in a magnetic space group.$^{[12]}$ In such a magnetic space group, there is a nonsymmetric time-reversal $T'$ symmetry, which is related to $T$ through the translation operation $\tau (T' = \tau T)$. In the centrosymmetric type-IV magnetic space group, the symmetry of the product $PT'$ is preserved; therefore, Kramers degeneracy is protected, and Dirac points can exist. EuCd$_2$As$_2$ has been theoretically predicted to be an antiferromagnetic (AFM) Dirac semimetal, in which DPs are protected by the $PT'$ symmetry.$^{[13]}$ EuCd$_2$As$_2$ forms a threefold symmetry in the $ab$-plane which is broken due to the spin configuration. According to theoretical calculations, this leads to a gap between Dirac cones.$^{[14,15]}$ However, strong ferromagnetic...
Table 1. Single crystal crystallographic data and structure refinement for EuZn$_2$As$_2$.

| Formula       | EuZn$_2$As$_2$ |
|---------------|----------------|
| F. W. (g mol$^{-1}$) | 432.54 |
| Space group, Z | P-3m1 (#164) |
| a [Å]         | 4.2093(1) |
| b [Å]         | 4.2093(1) |
| c [Å]         | 7.175(3) |
| V [Å$^3$]     | 110.09(6) |
| Absorption correction | Numerical |
| Extinction coefficient | 0.076(4) |
| $\Theta$ range [°] | 2.839–33.143 |
| hkl ranges    | $-6 \leq h \leq 6$
|               | $-6 \leq k \leq 6$
|               | $-11 \leq l \leq 10$ |
| No. reflections, $R_{\text{int}}$ | 1575, 0.0433 |
| No. independent reflections | 195 |
| No. parameters | 7 |
| $R_1$, w$R_2$, [all f] | 0.0258, 0.0378 |
| Goodness of fit | 1.084 |
| Largest diff. peak and hole [e$^{-} \text{Å}^{-1}$] | $-1.477, 2.013$ |

(FM) fluctuations above $T_N$ break the $T$ symmetry, giving rise to a Weyl state at high temperatures ($T > T_N$). The FM correlations in EuCd$_2$As$_2$ have been observed directly by resonant X-ray magnetic scattering. Interestingly, only a single pair of Weyl nodes is found in the condition of field-induced full spin alignment along the c-axis. Thus, such a system provides a rare case for studying the transition between the Dirac and Weyl states by tuning temperature or field.

To further study the influence of magnetism on the topology of the electronic band structure, we investigate the physical properties of EuZn$_2$As$_2$, a sister compound of EuCd$_2$As$_2$. The replacement of Cd by Zn is expected to weaken the spin-orbit coupling, thus influencing the gap size between Dirac cones. In addition, compared to EuCd$_2$As$_2$, we find that the AFM ordering temperature for EuZn$_2$As$_2$, $T_N = 19$ K, is doubled, offering a much wider temperature range for studying potential topological properties in a long-range AFM ordered state. By analyzing magnetization and magneto-transport data, we also find strong evidence for FM fluctuations over a much wider temperature range than that in EuCd$_2$As$_2$. Such information is key toward the understanding of magnetism-driven topological properties of related compounds.

### 2. Results and Discussion

The single crystal X-ray diffraction refinement confirms that our crystals form a trigonal structure with the formula EuZn$_2$As$_2$. The space group is P-3m1 (No. 164) with the lattice parameters $a = b = 4.2093(1)$ Å and $c = 7.175(3)$ Å. Replacing Cd with smaller Zn, the lattice parameters in EuZn$_2$As$_2$ decrease more significantly along the $a$ and $b$ axes (≈5.5% reduction) than the $c$ axis (≈2.4% reduction). Detailed information including atomic positions and sites occupancies is summarized in Tables 1 and 2.

Figure 1(a) illustrates the crystal structure of EuZn$_2$As$_2$, where Zn (green) and As (grey) form a honeycomb network separated by Eu atoms (pink). Figure 1(b) shows the X-ray diffraction pattern of a flat surface of a EuZn$_2$As$_2$ single crystal at room temperature. All peaks can be indexed with the abovementioned structure from the (001) plane (i.e., the $ab$-plane). A weak peak near $2\theta \approx 30^\circ$ results from residual Sn on the surface. A picture of a EuZn$_2$As$_2$ single crystal is presented in the inset of Figure 1(b). The powder X-ray diffraction (PXRD) pattern obtained from ground crystals was analyzed using the LeBail method and the result is presented in Figure S1, Supporting Information.

Figure 2(a) shows the temperature dependence of the zero-field-cooled (ZFC) and field-cooled (FC) magnetic susceptibility ($\chi = M/H$) measured by applying a magnetic field of 0.1 T parallel to the $c$-axis ($\chi_c$) and to the $ab$-plane ($\chi_{ab}$), respectively. With decreasing temperature, both $\chi_{ab}$ and $\chi_c$ initially increase with little difference between them. Below $T_N = 19$ K, $\chi_{ab}$ tends to saturate but $\chi_c$ decreases. Such behavior suggests that the system forms an A-type AFM order, with FM alignment in the $ab$-plane but AFM coupling along the $c$-axis. The much higher $T_N$ suggests stronger magnetic interactions both within the $ab$ plane and along the $c$ axis in EuZn$_2$As$_2$ than those in EuCd$_2$As$_2$. (19) consistent with the lattice parameter changes mentioned above.

To understand magnetic interactions in EuZn$_2$As$_2$, we fit $\chi_{ab}$ and $\chi_c$ between 100 and 300 K using the Curie–Weiss formula $\chi(T) = \chi_0 + C/(T - \theta)$, where $\chi_0$ is a constant, $\theta$ is the Curie–Weiss temperature, and $C$ is the Curie constant with $C = \mu_0^2 N_A k_B \chi_0$ ($N_A$ is Avogadro constant and $k_B$ is Boltzmann constant). As shown in the inset of Figure 2(a), the formula fits the data well (the solid lines are fitting curves). The parameters obtained are $\chi_{ab} = -6.50 \times 10^{-3}$ cm$^3$ mol$^{-1}$, $\theta_{ab} = 15.1$ K, and $\mu_{ab}^\text{eff} = 8.42$ μB, and $\chi_c = -2.38 \times 10^{-3}$ cm$^3$ mol$^{-1}$, $\theta_c = 17.0$ K, and $\mu_{c}^\text{eff} = 8.26$ μB. The positive values of $\theta_{ab}$ and $\theta_c$ imply dominant FM interactions between Eu ions, with the effective magnetic moments close to $\pm \mu_0 S$ for Eu$^{2+}$ (the solid lines are fitting curves).

Note that increased magnetic field pushes $T_N$ to lower temperatures (Figure 2(d)). As shown in Figure 2(e), the field dependence of $\chi_{ab}$ and $\chi_c$ suggests that increased magnetic field pushes $T_N$ to lower temperatures (Figure 2(d)).
Figure 1. a) The crystal structure of EuZn$_2$As$_2$. The pink, green, and grey balls represent Eu, As, and Zn atoms, respectively. b) The XRD pattern of EuZn$_2$As$_2$ single crystal. Inset: picture of EuZn$_2$As$_2$ single crystal.

Figure 2. a) The temperature dependence of the magnetic susceptibility measured along $ab$-plane ($\chi_{ab}$) and $c$-axis ($\chi_c$). Inset: the inverse magnetic susceptibility measured along $ab$-plane and $c$-axis. b), c) $\chi_{ab}$ and $\chi_c$ measured at several magnetic fields as a function of temperature, respectively. d) $H$–$T$ phase diagram constructed using $\chi_{ab}$ and $\chi_c$ in b) and c). e) Magnetic hysteresis loop measured along the $ab$-plane ($M_{ab}$) and $c$-axis ($M_c$) at $T = 2$ K. f) The temperature dependence of specific heat. Inset: the specific heat measured with $H = 0$ and 9 T. The solid line represents the background specific heat consisting of contributions from electrons and phonons at $H = 0$.

the magnetization has no hysteresis in either $M_{ab}$ or $M_c$ at 2 K. Instead, both $M_{ab}(H)$ and $M_c(H)$ vary linearly with field before reaching saturation. The observed saturation moment is close to the theoretical value for Eu$^{2+}$ ($\mu_{sat} = gJ = 7\mu_B$, where $J$ is the total angular momentum).\(^{20}\)

To confirm the nature of the phase transition at $T_N$, we have measured the temperature and field dependence of the specific heat, $C_p$. Figure 2f shows the temperature dependence of $C_p$ at $\mu_0H = 0$ and 9 T. Note that there is a lambda-shaped anomaly in $C_p(H = 0)$ at $T_N = 19$ K, indicating a second-order phase transition. To estimate the entropy associated with the phase transition, we subtract the electronic and phonon specific heat by fitting data between 1.8 and 4 K and between 25 and 30 K using $C_p = \gamma T + \beta T^3$ ($\gamma$ and $\beta$ are constants), as plotted in the inset of Figure 2f. The entropy change $\Delta S \approx 11.2$ J mol$^{-1}$ K$^{-1}$, about 65% of the theoretically expected value for an eightfold
The refined magnetic moment is 7.33(7). Figure 3c. The solid curve is the fit of parameter. The temperature dependence of the rocking curve scan at (0 0 ½) as the order parameter. The temperature dependence of I is shown in Figure 3c. The solid curve is the fit of I(T) to \( I = A(1 - T/T_N)^\beta \) + B, with \( T_N \approx 19.4 \) K, \( A = 3119, \beta = 0.23, \) and \( B = 1394. \) The critical exponent \( \beta \) obtained corresponds to a 2D magnetic system, consistent with the layered structure of EuZn2As2. Figure 3d shows the calculated structure factor square \( (F^2_{\text{calc}}) \) versus the observed one \( (F^2_{\text{obs}}) \). The linear behavior indicates excellent structure refinement. Any deviation is likely to be due to the errors resulting from the absorption correction process.

With an A-type magnetic structure (Figure 3b), it is necessary to ask why \( \theta_s \), obtained at temperatures well above \( T_N \), is positive. For EuCd2As2, both electron-spin resonance and muon-spin relaxation measurements reveal strong FM fluctuations with long time and length scales, which persist up to \( \approx 100 \) K. Although this was not discussed in ref. [16], we note that the electrical resistivity of EuCd2As2 also begins to show a negative slope below 100 K. Bearing this in mind, we investigate the temperature and field dependence of both the \( ab \)-plane \( (\rho_{ab}, I \parallel ab) \) and \( c \)-axis \( (\rho_c, I \parallel c) \) resistivities. Figure 4a shows the temperature dependence of \( \rho_{ab} \) and \( \rho_c \) between 2 and 300 K for EuZn2As2. Several features are worth mentioning. First, while \( \rho_c > \rho_{ab} \) due to the layered structure of EuZn2As2, \( \rho_{ab} \) and \( \rho_c \) show similar temperature dependence over the entire temperature range. This implies that the scattering mechanism is more or less the same in both the \( ab \) plane and the \( c \) direction. Similar behavior has also been reported for EuCd2As2 [24]. Second, there is a sharp peak in both \( \rho_{ab} \) and \( \rho_c \), corresponding to the magnetic transition at \( T_N \). The peak in \( \rho_c \) is even sharper than that in \( \rho_{ab} \). Third, both \( \rho_{ab} \) and \( \rho_c \) initially vary linearly with temperature at high temperatures, deviating below \( T_N \approx 200 \) K and eventually acquiring negative slopes \( (d\rho_{ab}/dT < 0 \) and \( d\rho_c/dT < 0) \) below \( \approx 150 \) K. The sharp

**Figure 3.** a) The rocking curve scans at (0 0 ½) at 4 and 40 K, measured by neutrons. b) Magnetic structure of Eu sublattice obtained at \( T = 4 \) K. c) The peak intensity as a function of temperature with the empirical law fit (red solid line) discussed in the text. d) Calculated structure factor square \( (F^2_{\text{calc}}) \) versus the observed one \( (F^2_{\text{obs}}) \).
The magnetic field dependence of resistivity at 300 K, respectively. MR is defined by $MR = \rho(H)/\rho(0) \times 100\%$. Both MR$_{ab}$ and MR$_c$ below $T_N$ indicate that the resistivity in all directions is dominated by spin scattering above $T_N$. The departure from the high-temperature linear behavior marks the spin scattering contribution to the resistivity due to magnetic fluctuations below $T_N$.

To confirm the effect of magnetic fluctuations on $\rho_{ab}$ and $\rho_c$, the magnetic field dependence of $\rho_{ab}$ and $\rho_c$ at constant temperatures is investigated. Figure 4b,c shows the field dependence of the transverse ($H \perp I$) magnetoresistivity $\Delta \rho_{ab}$ ($I \parallel ab$, $H \parallel c$) and $\Delta \rho_c$ ($I \parallel c$, $H \parallel ab$) at various temperatures between 2 and 300 K, respectively. MR is defined by $MR = \Delta \rho(H)/\rho(0) \times 100\%$.

At 300 K, both MR$_{ab}$ and MR$_c$ are small and positive, typical for a paramagnetic material. Quantitatively, both MR$_{ab}$ (300 K) and MR$_c$ (300 K) follow the $H^2$ behavior as expected. Upon cooling, both MR$_{ab}$ and MR$_c$ gradually decrease and become negative near 200 K. The field dependence of both MR$_{ab}$ and MR$_c$ also deviates from the $H^2$ behavior. With further cooling, their magnitudes continuously increase until $T_N$. The negative MR$_{ab}$ and MR$_c$ indicate the influence of FM fluctuations below $T_N$. At 200 K, consistent with what is seen in EuCd$_2$As$_2$,

Note that $T_N$ for EuZn$_2$As$_2$ is much higher than that in EuCd$_2$As$_2$. Both higher $T_N$ and $T_N$ offer wider temperature ranges for studying magnetism-related properties in EuZn$_2$As$_2$, than in EuCd$_2$As$_2$.

At $T_N$, the spin scattering is almost completely suppressed by the application of the magnetic field, so that MR$_{ab}$ and MR$_c$ reach $\approx -90\%$ at $H > H_{sat}$. Below $T_N$, the field dependence of both MR$_{ab}$ and MR$_c$ is nonmonotonic, with an initial increase followed by a decrease to negative values (Figure 4b,c). The initial positive MR$_c$ is attributable to AFM interaction in the $c$ direction (Figure 4a).

Positive MR$_{ab}$ should be related to the change in population of magnetic domains with the applied field. With increasing field (less than 1 Tesla at 5 K), both MR$_{ab}$ and MR$_c$ start to decrease, eventually becoming negative and saturated above $H_{sat}$. This implies continuous alignment toward the FM configuration. When $H$ reaches $H_{sat}$, all moments are aligned ferromagnetically. Figure 4d shows the field dependence of $\Delta \rho_{ab}$ at various angles $\phi$ ($\phi = H \cdot I$ -- defined in the inset, changing from transverse $[00\bar{1}]$ to longitudinal $[900]$) at $T = 0.6$ K. The low-field MR$_{ab}$ peak is gradually suppressed as $H$ turns away from the principal axes ($ab$- or $c$-axis). The inset of Figure 4d shows $\Delta \rho_{ab}(\phi)$ at $\mu_0H = 0.5$ and 6 T. Note that MR$_{ab}(\phi)$ reaches a minimum around $\phi = 45^\circ$, implying that there is field-induced spin reorientation. The preferable field-induced spin direction is $45^\circ$ between the $ab$-plane and $c$-axis, resulting in least spin scattering. This indicates that the magnetic configuration is extremely susceptible to the magnetic field.

With a strong magnetic fluctuation effect on the resistivity observed, it is interesting to consider the Hall response. Figure 4e shows the magnetic field dependence of the Hall resistivity ($\rho_{xy}$) at temperatures between $T_N$ and 300 K. Note that $\rho_{xy}$ increases linearly with the magnetic field at high temperatures, but deviation occurs when approaching $T_N$. For each temperature, we fit the data between 0 and 14 T to $\rho_{xy} = \rho_{0} + \rho_{0}R_{sh}H$, where $R_{sh}$ is a Hall coefficient. Figure 4f presents the temperature dependence of $R_{sh}$, which increases with decreasing temperature. The positive $R_{sh}$ suggests that holes are dominant carriers in EuZn$_2$As$_2$. Using the Drude relationship for a single band case with $R_{sh} = -1/ne$, the carrier concentration $n$ can be estimated. As shown in Figure 4f, $n \approx 4.6 \times 10^{20}$ cm$^{-3}$ at room temperature, consistent with the semimetallic scenario for EuZn$_2$As$_2$. It increases slightly with...
increasing temperature, which can be attributed to thermal effect if there is a small band gap as predicted in EuCd\(_2\)As\(_2\). It is our future work to elucidate the connection between magnetic fluctuations and possible topological phase transition in EuZn\(_2\)As\(_2\) as discussed in EuCd\(_2\)As\(_2\).[14–16]

3. Summary
We have successfully grown single crystalline EuZn\(_2\)As\(_2\), which forms a trigonal structure. The electrical resistivity, magnetization, and neutron diffraction investigation indicate that EuZn\(_2\)As\(_2\) orders antiferromagnetically below \(T_N = 19\) K with an A-type spin configuration. Similar to EuCd\(_2\)As\(_2\), there are strong FM fluctuations that give rise to profound spin scattering between \(T_N\) and \(T_{\text{fl}} \approx 200\) K in EuZn\(_2\)As\(_2\). Our MR\(_{ab}\) measurements with variable applied field direction indicate that there is field-induced reorientation of the spins to \(\approx 45^{\circ}\) between the ab-plane and c-axis. Compared to EuCd\(_2\)As\(_2\), the doubled \(T_N\) and \(T_{\text{fl}}\) make EuZn\(_2\)As\(_2\) a better platform for exploring topological properties in both magnetic fluctuation (\(T_N < T < T_{\text{fl}}\)) and ordered (\(T < T_N\)) regimes. It is especially interesting to find out 1) if the reduced spin-orbit coupling in EuZn\(_2\)As\(_2\) will reduce or close the gap between Dirac cones predicted in EuCd\(_2\)As\(_2\), 2) if the canted spin structure induced by magnetic field impacts the topological states, and 3) where the Dirac/Weyl points in EuZn\(_2\)As\(_2\) are located relative to the Fermi energy, and whether or non trivial electronic bands cross the Fermi level.

4. Experimental Section
The single crystals of EuZn\(_2\)As\(_2\) were grown via the flux method using Sn. The elemental Eu (99.99% pieces, Alfa Aesar), Zn (99.8% granules, Alfa Aesar), As (99.99% powder, Alfa Aesar), and Sn (99.9% granules, Alfa Aesar) were placed into an alumina crucible with a molar ratio Eu:Zn:As:Sn = 1:2:2:20, and sealed in an evacuated quartz tube. The sample was heated up to 600 °C at a rate of 60 °C h\(^{-1}\), and kept at this temperature for 5 h. This was followed warming to 1000 °C and tempering for 10 h. The sample was then slowly cooled (\(-3\) °C h\(^{-1}\)) down to 600 °C with a further centrifuge in order to remove Sn flux. The resulting single crystals had a typical size of 4 mm × 2 mm × 1 mm and were stable in air.

The crystal structure was determined by single crystal X-ray diffraction using a Bruker Apex II single X-ray diffractometer equipped with Mo radiation (\(\lambda_{\text{Mo}} = 0.71073\) Å), and by PXRD by means of a Rigaku Miniflex 600 diffractometer with Cu K\(_{\alpha}\) radiation (\(\lambda = 1.5406\) Å). The crystal structure was solved with the full-matrix least-squares method using the SHELXTL package.[26] The PXRD pattern was analyzed using the Fullprof refinement suite software.[10] The crystal structure was drawn by means of VESTA software.[27]

The magnetic properties were measured in a Quantum Design Magnetic Properties Measurement System (7 T). The electrical resistivity, Hall effect, and heat capacity were measured using a Quantum Design Physical Property Measurement System (14 T). The standard four-probe technique was used to measure the electrical resistivity and Hall effect, and the relaxation method was used for the heat capacity. For both the electrical resistivity and Hall effect measurements, four thin Pt wires were attached to a sample using silver epoxy. The contact resistance was 10–50 Ω. To eliminate signal from possible contact misalignment, both magnetoresistance (MR) and Hall resistivity (\(\rho_H\)) were obtained by sweeping the magnetic field between \(-14\) and \(+14\) T, that is, MR = [MR(\(+H\)) + MR(\(-H\))]\(/2\) and \(\rho_H = [\rho_H(\+H) - \rho_H(-H)]/2\). The angle dependence of the MR was measured at the pulsed-field facility of National High Magnetic Field Laboratory (NHMFL, Los Alamos).

To determine the magnetic structure of EuZn\(_2\)As\(_2\), single-crystal neutron diffraction experiment was performed on DEMAND (HB-3A) at the High Flux Isotope Reactor at Oak Ridge National Laboratory.[28] A wavelength of 1.008 Å from the bent Si-311 monochromator was used to reduce the heavy neutron absorption. Considering the large neutron absorption coefficient of Eu, PLATON software was employed to apply an absorption correction.[29,30] The magnetic and nuclear structures were both determined using Fullprof refinement Suite software.[10]

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Data Availability Statement
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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