Low-field magnetic anomalies in single crystals of the A-type square-lattice antiferromagnet EuGa$_4$

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(Dated: December 5, 2022)

The body-centered-tetragonal antiferromagnet EuGa$_4$ was recently identified as a Weyl nodal-line semimetal that exhibits the topological Hall effect below its reported antiferromagnetic (AFM) ordering temperature $T_N = 15$–16.5 K which we find to be $T_N = 16.4(2)$ K. The Eu$^{2+}$ ions are located at the corners and body center of the unit cell. EuGa$_4$ exhibits A-type antiferromagnetic order below $T_N$, where the Eu$^{2+}$ spin-$7/2$ moments are ferromagnetically aligned in the $ab$ plane with the Eu moments in adjacent Eu planes along the $c$ axis aligned antiferromagnetically. Low-field magnetization versus field $M(H_{ab})$ data at $T = 2$ K with the field aligned in the $ab$ plane are reported that exhibit anomalous positive curvature up to a critical field at which a second-order transition occurs with $H_{c1} \approx 0.85$ kOe for $H \parallel [1,1,0]$ and $\approx 4.8$ kOe for $H \parallel [1,0,0]$. For larger fields, the linear behavior $M_{ab} = \chi(T_N)H_{ab}$ is followed until the critical field $H_{c2}^\ast$ is reached at which all moments become aligned with the applied field. A theory is formulated for $T = 0$ K that fits the observed $M(H_{ab})$ behavior at $T = 2$ K well, where domains of A-type AFM order with fourfold rotational symmetry occur in the AFM state in zero field. The moments in the four domains reorient to become almost perpendicular to $H_{ab}$ at $H_{c1}$, followed by increasing canting of all moments toward the field with increasing field up to the critical field $H_{c2}^\ast$ which is reported to be 71 kOe, at which all moments become aligned parallel to the field. A first-order transition in $M(H_{ab})$ at $H_{ab} = H_{c1}$ is predicted by the theory for $T = 0$ K when $H_{ab}$ is at a small angle from the [1,0,0] or [1,1,0] symmetry-axis directions.

I. INTRODUCTION

Antiferromagnets are fundamentally interesting owing to their various spin arrangements as well as their technological applications in spintronics, spin valves, magnetotactical devices, and spin-wave-based information technologies [1–4]. Recently, many antiferromagnetic (AFM) compounds have also been discovered to host nontrivial topological electronic and spin states [5–8]. Understanding the magnetic interactions in these materials are important for their further development and discovery of new materials. The magnetic ordering in those materials are primarily determined by the interplay of exchange interaction, magnetic anisotropy energy, and any kind of disorder present in the system. In particular, magnetocrystalline and magnetic-dipole anisotropies play a crucial role in tuning the spin arrangements in different AFM materials.

Among these materials, Eu-based antiferromagnets have been of significant interest recently due to the complex interplay of magnetism and topological states [6–9–11]. EuX$_4$-type of materials (X = Al, Ga) constitute one such family which is generating significant interest due to the recent observation of the topological Hall effect (THE) and related phenomena in these materials [12–14]. They crystallize in the body-centered-tetragonal (bct) BaAl$_4$-type crystal structure (Fig. 1) with space group $I4/mmm$ [15], where the Eu atoms in each $ab$-plane layer form a square lattice and are known to exhibit a rich variety of magnetic and electronic properties. For example, EuAl$_4$ orders antiferromagnetically below $T_N = 15$ K along with a CDW transition at $T_{CDW} = 140$ K [16–20]. The CDW transition is suppressed to $T = 0$ K by the application of a pressure of 2.5 GPa [18]. The isoivalent analogue EuGa$_4$ also orders antiferromagnetically below $T_N \approx 16$ K and a CDW is only observed at $T_{CDW} = 105$ K under the application of a pressure $p = 0.75$ GPa [18–21]. A THE is also evidenced in EuAl$_4$ coexisting with CDW order [12]. Although magnetic spin reorientation and multiple metamagnetic transitions were observed earlier in EuGa$_2$Al$_2$ [19], a recent observation of the THE and incommensurate magnetic order suggest the presence of a field-induced skyrmion-like topological spin texture in this compound [14]. A lack of inversion symmetry in non-centrosymmetric materials with Dzyaloshinskii-Moriya (DM) interactions was initially thought to be the key ingredient for stabilizing this spin texture. However, observations of a skyrmionic phase in centrosymmetric materials [8–22–23] have challenged the understanding and mechanism of this spin-texture formation. Contemporary theoretical modeling suggests that the interplay of different spin interactions and anisotropy may play a crucial role in the formation of a topological spin texture in centrosymmetric materials [24]. Thus, to understand the mechanism of complex spin texture and its field-induced evolution, it is necessary to study the magnetic properties and anomalous behavior in antiferromagnets with small anisotropy.

EuGa$_4$ exhibits giant magnetoresistance (MR) and THE with a possibility of magnetic skyrmions [13–25]. Recently, the observation of large transverse MR in this semimetal is explained due to the presence of Weyl nodal-line (NL) states and magnetic-field-induced Landau quantization [25]. As reported earlier, EuGa$_4$ ex-
hibits collinear A-type antiferromagnetic (AFM) order below \( T_N \approx 16 \) K, where the Eu atoms are ferromagnetically aligned along the \( ab \)-planes and adjacent FM planes along the \( c \)-axis are aligned antiferromagnetically \([17, 18, 21, 25, 27, 28]\). Although a noncollinear magnetic structure is favorable for skyrmion-like texture formation, the possibility of this texture in collinear EuGa\(_4\) is quite intriguing where anisotropy can play an important role.

Previous magnetic studies on EuGa\(_4\) mostly focused on the magnetic ground state and the high-magnetic-field behavior, while the low-field behavior and the effect of anisotropy was hardly explored. However, magnetization \( M \) versus applied magnetic field \( H \) isotherm measurements of a crystal with the field along the \( [1,0,0] \) direction at \( T = 2 \) K revealed positive curvature up to a field \( H_d = 5 \) kOe, above which \( M(H) \) was linear up to the critical field \( H_{[1,0,0]} \approx 71 \) kOe at which all moments become parallel to the field, whereas for the \( c \)-axis field, \( M(H) \) was linear over the whole field range where \( H_{[0,0,1]} \approx 72 \) kOe (nearly isotropic) \([21]\). The authors suggested that this behavior was somehow associated with AFM domains that evolved into a single domain at \( H_d \), and found that \( H_d \) decreased smoothly to zero on heating to \( T_N \).

Here, we report studies of the magnetic-field evolution of the AFM ground-state spin texture at \( T = 2 \) K in detail emphasizing the low-field region. We found that although the \( c \)-axis magnetization increases linearly with the applied field \( H \), as expected and found for a A-type AFM, a nonlinear \( M(H) \) response at low fields was observed for the \( [1,0,0] \) field direction as previously found in Ref. \([21]\). Interestingly, we found that the low-field \( (ab) \)-plane nonlinearity differs significantly for fields in the \( [1,0,0] \) and \( [1,1,0] \) directions. On the basis of our temperature- and magnetic-field-dependent magnetic measurements complemented with theoretical analyses, we conclude that the ground-state A-type AFM structure consists of four AFM domains having fourfold rotational symmetry associated with the fourfold \( ab \)-plane magnetic anisotropy of the ferromagnetic \( ab \)-plane layers.

We propose a theory in which, with increasing field in the \( ab \) plane, the moments in each domain initially cant become nearly perpendicular to the field at a critical field \( H_{c1} \) (\( H_d \) above) with no change in the physical domain boundaries. Then with a further increase of the magnitude of the field all moments progressively cant towards the field giving rise to the observed linear \( M(H) \) behavior up to the critical field \( H_{cb} \) noted above. Our fits describe the experimental \( M(H) \) isotherms at \( T = 2 \) K for \( H \parallel [1,0,0] \) and \( H \parallel [1,1,0] \) with \( H \leq H_{c1} \) rather well, where the \( H_{c1} \) values for the two field directions are quite different.

The experimental details are given in Sec. II. The experimental results are presented in Sec. III, including magnetic susceptibility \( \chi(T) \) data in Sec. IIIA and magnetization versus field \( M(H) \) isotherms in Sec. IIIB. Theoretical fits to the experimental \( M(H) \) data at \( T = 2 \) K are given in Sec. IV.

II. EXPERIMENTAL DETAILS

EuGa\(_4\) single crystals were grown using an EuGa\(_9\) self-flux. The high purity elements (Eu metal from Ames Laboratory and 99.99999%-pure Ga from Alfa Aesar) were loaded in an alumina crucible and sealed in a silica tube. The ampule was then heated to \( 750 \) °C at a rate of \( 100 \) °C/h and held for 12 h. Then it was slowly cooled to \( 400 \) °C at a rate of \( 2 \) °C/h. The crystals were obtained after removing the flux using a centrifuge. The sample homogeneity and chemical composition were confirmed using a JEOL scanning electron microscope (SEM) equipped with an EDS (energy-dispersive x-ray spectroscopy) analyzer. The EDS measurements yielded a composition EuGa\(_{4.04(2)}\), close to the stoichiometric composition. Magnetic measurements were carried out using a Magnetic-Properties-Measurement System (MPMS) from Quantum Design, Inc.

We use cgs magnetic units throughout, where \( 1 \) T = \( 10^4 \) Oe.

III. EXPERIMENTAL RESULTS

A. Magnetic Susceptibility

The \( \chi(T) \) data for EuGa\(_4\) obtained with in-plane (\( \chi_{ab}, H \parallel ab \)) and out-of-plane (\( \chi_{c}, H \parallel c \)) magnetic fields \( H = 0.1 \) kOe are shown in Fig. 2, where \( \chi_{ab} \) is measured for the two symmetry directions \( H \parallel [1,0,0] \) and \( H \parallel [1,1,0] \). The data in the figure indicates that \( \chi \) is nearly isotropic in the \( ab \) plane. As seen from the figure, EuGa\(_4\) undergoes an AFM transition at \( T_N = 16.4(2) \) K,
similar to values reported earlier [17,18,21,23,27,28]. The $\chi_c$ is found to be independent of $T$ for $T < T_N$, indicating that the moments are aligned perpendicular to the $c$ axis. This is consistent with the $\chi_{ab}$ data that decrease with decreasing $T$ with $\chi_{ab}(2 \text{ K})/\chi(T_N) \approx 0.5$. According to molecular-field-theory (MFT) [29,30] for a $c$-axis helix of identical crystallographically-equivalent Heisenberg spins, one has

$$\frac{\chi_{ab}(T = 0)}{\chi_{ab}(T_N)} = \frac{1}{2[1 + 2 \cos(kd) + 2 \cos^2(kd)]}, \quad (1)$$

where $k$ is the magnitude of the $c$-axis AFM propagation vector, $d$ is the distance along the $c$ axis between the FM layers of spins, and hence $kd$ is the turn angle between the adjacent layers of spins.

The ratio on the left side of Eq. (1) for $H \parallel [1,0,0]$ at $T = 2 \text{ K}$ was previously found to be $\approx 0.26/0.51 \approx 0.51$ [21]. According to Fig. 2 as noted above we find the similar value

$$\frac{\chi_{[1,0,0]}(T = 2 \text{ K})}{\chi(T_N)} \approx 0.24 \approx \frac{1}{2} \approx 0.48. \quad (2)$$

Using this value of $\chi_{ab}(2 \text{ K})/\chi_{ab}(T_N)$, Eq. (1) yields the turn angle between the moment directions in adjacent Eu layers to be $kd = 180^\circ$, indicating that the AFM structure is $A$-type, in agreement with the earlier neutron-diffraction solution of the magnetic structure of EuGa$_4$ [27].

The inverse molar magnetic susceptibility $1/\chi$ for $H = 0.1 \text{ T}$ is plotted versus $T$ for $H \parallel [1,0,0]$ in Fig. 3(a) and $H \parallel [0,0,1]$ in Fig. 3(b). Both plots are linear above $T_N$ and are described well by the inverse of the modified Curie-Weiss law

$$\chi(T) = \chi_0 + \frac{C}{T - \theta}, \quad (3)$$

where $\chi_0$ is the $T$-independent contribution, $C$ is the molar Curie constant and $\theta$ is the Weiss temperature. The fits yield the values of these variables in Table I. The magnitudes of the diamagnetic $\chi_0$ values are of the order expected for the diamagnetic core contributions but are very small relative to the $\chi$ values of the Eu$^{2+}$ moments. The listed effective moments $\mu_{\text{eff}}$ for the two field directions are close to the theoretical value of $7.94 \mu_B$/Eu$^{2+}$ for $g = 2$ and $S = 7/2$. The Weiss temperatures are positive, consistent with the $A$-type AFM structure in which FM planes of Eu spins are stacked antiferromagnetically along the $c$ axis. However, they are not close to the value of $T_N$, indicating that the AFM interactions between the Eu spins in adjacent layers perpendicular to the $c$ axis are also significant.

The magnetic-field dependences of $\chi(T)$ are shown in Fig. 4 for (a) $H \parallel c$, (b) $H \parallel [1,1,0]$ and (c) $H \parallel [1,0,0]$. No change in $\chi_c(T)$ is observed between $H = 0.1$ and 1 kOe. However, a significant variation of $\chi_{ab}(T)$ is observed in this field region. Interestingly, the field evolution of $\chi_{ab}(T)$ at low fields is quite different when the applied field is applied along the $ab$ plane $[1,0,0]$ and $[1,1,0]$ directions. The critical fields at which the moments become aligned with the applied field are at much higher fields $H_{ab}^c = 71 \text{ kOe}$ and $H_{c}^c = 72 \text{ kOe}$ for $H \parallel ab$ and $H \parallel c$, respectively [21], indicating a very small magnetic anisotropy between these two field directions as expected for Eu$^{2+}$ moments with $S = 7/2$ and $L = 0$.

The $\chi_{ab}(T)$ in Fig. 4(b) for $T \leq T_N$ strongly increases between applied fields $H = 0.1$ and 1 kOe applied along the $[1,1,0]$ direction and at higher fields it becomes independent of $T$. On the other hand, only a gradual increase in $\chi_{ab}(T)$ with increasing $H$ is observed for $H \parallel [1,0,0]$ in Fig. 4(c) in the field range 0.1 kOe $\leq H \lesssim 6$ kOe. Moreover, a $T$-independent region of $M(H)$ is observed for $H = 1 \text{ kOe}$ for $H \parallel [1,0,0]$ and the temperature range of that plateau increases with increasing $H$. Finally, $\chi(T)$ for both $H \parallel [1,0,0]$ and $H \parallel [1,1,0]$ in the AFM state below $T_N$ becomes independent of $T$ for $H = 10 \text{ kOe}$. We show in Sec. [XV] below that the different low-field $M(H)$ behavior of $\chi_{ab}(H)$ for $H \parallel [1,0,0]$ and $H \parallel [1,1,0]$ in EuGa$_4$ is due to AFM domain formation arising from the fourfold tetragonal $c$-axis rotational symmetry. Similar effects were found previously in trigonal Eu-based compounds with threefold rotational symmetry.

TABLE I. The fitted parameters to the inverse susceptibility data in Fig. [III] including the $T$-independent contribution to the susceptibility $\chi_0$, molar Curie constant $C_0$ for $a = ab$, $c$ directions, effective moment per Eu spin $\mu_{\text{eff}} \approx \sqrt{8C_0}$ and the Weiss temperature $\theta_o$.

| Field direction | $\chi_0$ ($10^{-5} \text{ cm}^3/\text{mol}$) | $C_0$ ($\text{cm}^3K/\text{mol}$) | $\mu_{\text{eff}}$ ($\mu_B$) | $\theta_o$ (K) |
|-----------------|---------------------------------|----------------------------|-----------------------------|---------------|
| $H \parallel ab$ | -1.7(5)                         | 7.76(1)                    | 7.88(1)                     | 2.27(6)       |
| $H \parallel c$  | -3.5(3)                         | 7.86(1)                    | 7.93(1)                     | 0.5(1)        |
FIG. 3. (a) Out-of-plane Magnetic susceptibility $\chi_c(T)$ ($H \parallel c$) of EuGa$_4$ for different applied magnetic fields. In-plane magnetic susceptibility for different magnetic fields when (b) $H \parallel ab \parallel [1,1,0]$ and (c) $H \parallel ab \parallel [1,0,0]$. The field responses in these two in-plane symmetry directions are significantly different.

B. Magnetization Isotherms

In order to provide further insight into the field-dependent evolution of the magnetic behavior at $T < T_N$, $M(H)$ isotherm data were obtained that emphasize the low-field region of interest. As can be seen in Fig. 5, the $M(H)$ behavior measured at $T = 2$ K for $H \parallel c$ is linear. In accordance with the magnetic susceptibility measurements, a clear nonlinear response in $M(H)$ is observed for both $H \parallel [1,0,0]$ and $H \parallel [1,1,0]$. This is clearly reflected in the $dM/dH$ data, where $dM/dH$ initially increases rapidly with increasing $H$ and exhibits peaks at the critical fields $H_{c1,[1,0,0]} \approx 4.8$ kOe and $H_{c1,[110]} \approx 0.85$ kOe, followed eventually by an $H$-independent behavior for $H > H_{c1}$. The difference in the low-field $M(H)$ behavior for different in-plane symmetry directions can be explained by the rotation of the moments in $ab$-plane AFM domains as discussed in detail below.

The $M(H)$ data measured at different temperatures for $H \parallel [1,1,0]$ are shown in Fig. 6(a) and the corresponding $dM/dH$ versus $H$ data are plotted in Fig. 6(c). As seen in the latter figure, $H_{c1}$ slightly shifts to...
lower fields with increasing temperature below \( T_N \), with \( H_{c_2, [1,1,0]} = 0.85 \) kOe at \( T = 2 \) K decreasing to 0.6 kOe at \( T = 14 \) K. The \( M(H) \) behavior is linear for \( T > T_N \). The \( T \) dependences of \( M(H) \) and \( dM/dH \) for \( H \parallel [1,0,0] \) are shown in Figs. 6(b) and 6(d), respectively. Here, the nonlinearity in \( M(H) \) at \( T = 2 \) K persists up to \( H \approx 0.8 \) T, which is much larger than that observed for \( H \parallel [1,1,0] \). The \( dM/dH \) for this field direction shows a maximum at \( H_{c_1,[1,0,0]} = 4.8 \) kOe at \( T = 2 \) K. This critical field is significantly reduced to \( H_{c_1,[1,0,0]} = 0.85(5) \) kOe at \( T = 14 \) K. The striking difference observed in the \( M(H) \) and corresponding \( dM/dH \) behavior between the \( H \parallel [1,1,0] \) and \( H \parallel [1,0,0] \) directions indicates the presence of significant in-plane magnetic anisotropy. This anisotropy is associated with the magnetic-field-induced moment reorientation in the AFM domains discussed below in Sec. IV.

We tested the reversibility of the nonlinear \( M(H) \) and \( dM/dH \) at low fields upon heating and cooling for \( H \parallel [1,0,0] \). The crystal was initially cooled to \( T = 2 \) K under a magnetic field \( H = 5 \) T. After \( T \) stabilization, \( M(H) \) was measured in the hysteresis \( H \) cycle 10 kOe \( \rightarrow 0 \) kOe \( \rightarrow 10 \) kOe, as shown in Fig. 7. No magnetic hysteresis was observed, indicating that the low-field-induced \( M(H) \) nonlinearity is reversible.

Similar \( M_{ab}(H) \) behavior was observed for the Eu-based trigonal compounds EuMgB\(_2\) and EuMg\(_2\)B\(_2\) [32, 33, 34, 35] and we successfully modeled those results [33] using an approach similar to that used below to model the low-field \( M(H) \) data for EuGa\(_4\).

### IV. FITS TO THE EXPERIMENTAL M(H) DATA

#### A. Theory

We write the fourfold rotational magnetic anisotropy energy \( E_{anis} \) for the ferromagnetic \( ab \)-plane layers in tetragonal EuGa\(_4\) versus the azimuthal angle \( \phi \) of the ferromagnetic moments in that layer as

\[
E_{anis} = K_4 \cos(4\phi),
\]

where \( K_4 > 0 \) is the fourth order anisotropy constant and \( \phi \) is the angle of the moments with respect to the \( x \) axis defined in Fig. 9 below. A plot of \( E_{anis}/K_4 \) versus \( \phi \) is shown in Fig. 8. The anisotropy-energy minima occur at \( \phi = \pm \pi/4 \) and \( \pm 3\pi/4 \) rad.

In order to model the anomalous low-field \( M_{ab}(H) \) behavior for EuGa\(_4\) in Figs. 5–7, we propose that the magnetic structure in \( H = 0 \) contains four equally-populated domains A, B, C, D of ferromagnetically-aligned moments in the A-type AFM structure illustrated in Fig. 9(a) as required by the tetragonal lattice symmetry. As shown in Fig. 9(a), each of the four domains contains moments that are ferromagnetically-aligned in every-other \( ab \)-plane and the moments in adjacent layers along the \( c \) axis are aligned at 180° with respect to the former moments, as required for an A-type AFM structure. We assume that within each physical AFM domain, the applied field \( H_x \) rotates the moments only in the \( ab \) plane and does not cause domain-wall motion. The former assumption is justified because in a body-centered-tetragonal lattice, the magnetic-dipole interaction favors ferromagnetic moment alignment in the \( ab \) plane rather than along the \( c \) axis [39]. As noted at the bottom of Fig. 9(a), the \( x \) direction of the applied field can be aligned along either the crystallographic \([1,0,0]\) or \([1,1,0]\) directions which are expected to have different anisotropy energies.

In \( H_x = 0 \), the angles of moments 1, 3, 5, and 7 with respect to the \( x \) axis in the respective ferromagnetically-aligned layer are in energy minima according to Fig. 8. Similarly, moments 2, 4, 6, and 8 in either of the two layers of the A-type AFM structure adjacent to the respective layers containing moments 1, 3, 5, and 7 are also in energy minima. On application of \( H_x \), due to the relationship of the directions of the moments in the different domains in Fig. 9 to each other, the magnitude of the change of the moment angle \( \Delta \phi \) is the same for the moments in each domain as shown in the figure. During this process, the moments in adjacent ferromagnetically-aligned layers retain their 180° alignment due to the AFM exchange interaction between moments in adjacent layers, apart from a very small canting towards the field which gives rise to the observed magnetization.

The moments in each domain eventually rotate to become perpendicular to \( H_x \) at a critical field \( H_{c_1} \) at which a maximum is observed in \( dM_{ab}/dH \) in Figs. 5–7. For \( H_x > H_{c_1} \), the moments in each domain increasingly cant towards the applied field direction as shown in Fig. 9(b). The magnetization saturates when all the moments become parallel to the applied field \( H_x \) at a critical field \( H_x \). As noted at the bottom of Fig. 9(a), it is possible to align \( H \) at an angle \( \phi_H \neq 0 \) with respect to the positive \( x \) axis. As illustrated later, at \( T = 0 \) K this is predicted to result in a first-order transition at \( H_{c_1} \).

For \( 0 \leq H_x \leq H_{c_1} \), from Fig. 9(a) the angles of the moments in each domain with respect to the positive \( x \) axis are respectively

\[
\begin{align*}
\phi_{12} & = \frac{\pi}{4} + \Delta \phi, \\
\phi_{34} & = \frac{3\pi}{4} - \Delta \phi, \\
\phi_{56} & = -\frac{3\pi}{4} + \Delta \phi, \\
\phi_{78} & = -\frac{\pi}{4} - \Delta \phi,
\end{align*}
\]

where \( 0 \leq \Delta \phi \leq \pi/4 \). The average anisotropy energy per domain for \( 0 \leq H_x \leq H_{c_1} \) obtained using Eqs. 4 and 5 is

\[
E_{anis,ave} = -K_4 \cos(4\Delta \phi).
\]

The magnetization component \( \mu \parallel \) of a moment along the axis of a pair of collinear moments aligned antipar-
all in domain \( n \) with azimuthal angle \( \phi_n \) in Fig. 9(a) at \( T = 0 \) is zero, whereas the component \( \mu_\perp \) of the moment perpendicular to the moment is \( \mu_\perp = \chi_\perp H_\perp = \chi_\perp H_x \sin(\phi_n) \), where \( \chi_\perp \) is the magnetic susceptibility per moment in a domain when the field is perpendicular to it. The angle \( \phi_n \) is the angle of domain \( n \) in Fig. 9(a) with respect to the \( x \) axis given by Eqs. (5). The component of the applied field in the direction perpendicular to the moment is \( H_\perp = H_x \sin(\phi_n) \). Therefore the

FIG. 6. In-plane \( M(H) \) data measured at different temperatures for (a) \( H \parallel [1,1,0] \) and (b) \( H \parallel [1,0,0] \). The respective \( dM(H)/dH \) versus \( H \) data are shown in (c) and (d). The data for \( T = 18 \) K are about 2 K above \( T_N \).

FIG. 7. In-plane field-cooled \( M(H) \) behavior for \( H \parallel [1,0,0] \) in the hysteresis \( H \) cycle 10 kOe \( \rightarrow \) 0 \( \rightarrow \) 10 kOe. No hysteretic behavior is observed. The corresponding \( dM(H)/dH \) behavior is also shown in the right ordinate.

FIG. 8. Fourfold \( ab \)-plane rotational anisotropy energy normalized by the anisotropy constant \( K \), \( E_{\text{anis}}/K \), versus the \( ab \)-plane tilt angle \( \Delta\phi/\pi \) rad of a moment in tetragonal EuGa4.
Increasing $H$, $H_{c1} \leq H \leq H_c$

All Domains A, B, C, D

Increasing $H$, $H_{c1} \leq H \leq H_c$

(b)

All Domains A, B, C, D

Increasing $H$, $H_{c1} \leq H \leq H_c$

$m\times H$ || $[1,0,0]$ or $[1,1,0]$  $|\phi_H| < \pi/4$

$H_x, \phi_H$

$\chi_{\perp H} H_x^2 / K_4$

FIG. 10. The angle $\Delta \phi$ in Fig. 9(a) versus $\chi_{\perp H} H_x^2 / K_4$ for $\phi_H = 0$ rad as defined at the bottom of Fig. 9(a).

$m\times H$ || $[1,0,0]$ or $[1,1,0]$  $|\phi_H| < \pi/4$

$H_x, \phi_H$

$\chi_{\perp H} H_x^2 / K_4$

FIG. 9. (a) Schematic diagram of the nearly-locked moment orientations in the $ab$-plane of adjacent antiparallel layers of moments along the $c$ axis in the four collinear A-type AFM domains A, B, C, and D and their magnetic field evolution with increasing $x$-axis field $H$ at low fields $H \leq H_{c1}$ shown by arrows. (b) For $H_x > H_{c1}$, all pairs of moments in adjacent ferromagnetically-aligned layers increasingly cant by the same amount towards the increasing field as shown, until at the critical field $H_c$ all moments are aligned with the field.

magnetic energy of a moment in domain $n$ in the regime $0 \leq H_x \leq H_{c1}$ is given by

$$E_{\text{mag } n} = \mu_n H_x$$

$$= - \left[ \chi_{\perp H_x} \sin(\phi_n) \right] \left[ H_x \sin(\phi_n) \right]$$

$$= - \chi_{\perp} H_x^2 \sin^2(\phi_n).$$

Summing $E_{\text{mag } n}$ over the angles of the moments in the four domains in Eq. (3) and dividing by four gives the average magnetic energy per moment for $0 \leq H_x \leq H_{c1}$. Then using Eq. (8) and normalizing the total energy per moment by $K_4$ gives the total average energy per moment as

$$E_{\text{ave}} / K_4 = (E_{\text{anis ave}} + E_{\text{mag ave}}) / K_4$$

$$= - \cos(4\Delta \phi) - \frac{\chi_{\perp} H_x^2}{2} \left[ 1 + \sin(2\Delta \phi) \right].$$

Setting the derivative of $E_{\text{ave}} / K_4$ with respect to $\Delta \phi$ in Eq. (7b) equal to zero gives $\Delta \phi$ versus $\chi_{\perp} H_x^2 / K_4$ as plotted in Fig. 10. The maximum value $\Delta \phi = \pi/4$ is the value at which all moments become perpendicular to the applied field $H_x = H_{c1}$, apart from a small canting of all moments towards the field that gives rise to the measured magnetization along the $x$ axis. At larger fields up to the critical field $H_c$, the individual moments cant towards the field and molecular-field theory predicts $\chi = \chi_{\perp} H_x = \chi(T_N) H_x$ until the critical field $H_c$ is reached at which all the moments are aligned with the applied field.

Figure 10 shows the value of $\chi_{\perp} H_x^2 / K_4$ at the critical field $H_{c1}$ for which $\Delta \phi = \pi/4$ rad is given by

$$\frac{\chi_{\perp} H_x^2}{K_4} = 8,$$

where the factor of 8 appears to be exact. This equation gives the value of the anisotropy constant $K_4$ in terms of measurable quantities as

$$K_4 = \frac{\chi_{\perp} H_x^2}{8 N_A}.$$

Since $K$ is normalized to a single moment, whereas the measured $\chi_{\perp}$ is normalized to a mole of moments, this difference can be taken into account by writing Eq. (8b) as

$$K_4 = \frac{\chi_{\perp} H_x^2}{8 N_A},$$

where $N_A$ is Avogadro's number. For EuGa$_4$, the measured values are $\chi_{ab}(T_N) = 0.48$ cm$^3$/mol and $H_{c1} \approx 4.8$ kOe for $H_x || [1,0,0]$, yielding

$$K_4 = 1.43 \times 10^{-3} \text{ meV} / \text{Eu atom}.$$

This value is of order 100 times larger than the values of the threefold anisotropy constants $K_3 = 6.5 \times 10^{-5}$ and
\[ 1.8 \times 10^{-5} \text{meV/atom} \] obtained for trigonal EuMg_2Bi_2 and EuMg_2Sb_2, respectively [33].

**B. Fits of the M(H) data at T = 2 K by theory**

The magnetization \( M_x \) per mole of Eu moments versus magnetic field \( H_x \) is calculated from

\[
M_x(H) = \frac{x \perp H_x}{2} \left[ \sin^2 \left( \frac{\pi}{4} + \Delta \phi \right) + \sin^2 \left( \frac{3\pi}{4} - \Delta \phi \right) \right] = \frac{x \perp H_x}{2} \left[ 1 + \sin(2\Delta \phi) \right],
\]

where \( x \perp \) is equal to the molar magnetic susceptibility at \( T_N \) and \( \Delta \phi \) is given by the data in Fig. [10]

The experimental \( M_{ab}(H) \) data for \( H_x \parallel [1,0,0] \) and \( H_x \parallel [1,1,0] \) directions along with the calculated theoretical \( M_x(H) \) behavior using Eq. [10] are shown in Figs. [11(a) and 11(b)], respectively. The value of \( H_{c1} \) is seen to be quite different for the two field directions. The theory reproduces the experimental data rather well at low and high fields, but deviates somewhat from the data for \( H \approx H_{c1} \). The reason for this discrepancy is not clear at present but may be associated with the fact that the theoretical calculations are done for \( T = 0 \) K, whereas the observed \( M_x(H) \) data were obtained at \( T = 2 \) K. A larger discrepancy between the theoretical and experimental data taken at \( T = 1.8 \) K was also observed earlier for trigonal EuMg_2Bi_2 and EuMg_2Sb_2, where the measurement temperatures were \( T \approx 0.27 T_N \) and \( T \approx 0.23 T_N \), respectively. The discrepancy is smaller for EuGa_4 with measurement temperature \( T \approx 0.13 T_N \). This is expected since the \( T = 0 \) K theoretical predictions are expected to most accurately agree with the \( M(H) \) data when the data are measured at \( T \ll T_N \).

We have also calculated the \( M(H) \) behavior when \( H_x \) is not along the \( x \) axis parallel to the \([1,0,0]\) or \([1,1,0]\) direction, but is in a direction in the \( ab \) plane where \( H \) is at an arbitrary positive angle \( \phi_H < \pi/4 \) with respect to the +\( x \) axis as indicated at the bottom of Fig. [7(a)]. In this case there are effectively two domains A and B, because \( \phi_C = -\phi_1 \) and \( \phi_D = -\phi_2 \). We therefore minimize the energy only with respect to \( \phi_1 \) of Domain A and \( \phi_2 \) of Domain B. The angles of the two domains for \( 0 \leq H_x \leq H_{c1} \) with respect to \( \phi_H \) are

\[
\phi_1 - \phi_H = \frac{\pi}{4} + \Delta \phi_1 \quad (0 \leq \Delta \phi_1 \leq \pi/4 + \phi_H),
\]

\[
\phi_2 - \phi_H = \frac{\pi}{4} - \Delta \phi_2 \quad (0 \leq \Delta \phi_2 \leq \pi/4 - \phi_H).
\]

The average anisotropy energy associated with the two domains is

\[
E_{\text{anis ave}} = -\frac{K}{2} \left[ \cos(4\phi_1) + \cos(4\phi_2) \right].
\]

The average magnetic energy in the regime \( 0 \leq H_x \leq H_{c1} \) is given by

\[
E_{\text{mag ave}} = -\frac{x \perp H_x^2}{2} \left[ \sin^2(\phi_1 - \phi_H) + \sin^2(\phi_2 - \phi_H) \right] = -\frac{x \perp H_x^2}{4} \left[ 2 + \sin[2(\phi_1 - \phi_H)] \right]
\]

\[
+ \sin[2(\phi_2 - \phi_H)].
\]

Thus, the total average energy at \( T = 0 \) normalized by \( K_4 \) is

\[
E_{\text{ave}}/K_4 = (E_{\text{anis ave}} + E_{\text{mag ave}})/K_4
\]

\[
= -\frac{1}{2} \left[ \cos(4\phi_1) + \cos(4\phi_2) \right] - \frac{x \perp H_x^2}{4K_4} \left[ 2 + \sin[2(\phi_1 - \phi_H)] \right]
\]

\[
+ \sin[2(\phi_2 - \phi_H)].
\]

Here we minimize of \( E_{\text{ave}}/K_4 \) with respect to both \( \Delta \phi_1 \) and \( \Delta \phi_2 \) where \( \phi_1 \) and \( \phi_2 \) have maximum values of \( \pm \pi/4 + \phi_H \).
The angles $\Delta \phi_1$ and $\Delta \phi_2$ in Eqs. (11) versus $\chi \perp H^2_x/K$ for $\phi_H = 1/100$ rad. $\Delta \phi_1 = \pi/4$ for $\chi \perp H^2_x/K > 7.1$, whereas $\Delta \phi_2$ eventually asymptotes to $\pi/4$ rad.

and $\pi/4 - \phi_H$, respectively [see Fig. 9(a)]. As can be seen from the Fig. 12, a discontinuous first-order transition is observed for $\Delta \phi_1$ for $\phi_H > 0$ at $H = H_{c1,A}(\phi_H)$, where $\Delta \phi_1$ reaches $\pi/4 + \phi_H$ in order for the moments to be perpendicular to $H_x$ at $H_{c1,1}$. On the other hand, no such first-order transition is observed for $\Delta \phi_2$, where $\Delta \phi_2$ asymptotes continuously to $\pi/4 - \phi_H$ at larger $H$. The critical field $H_{c1,1}$ is found to decrease and the discontinuity of $\Delta \phi_1$ at $H_{c1,1}$ to increase with increasing $\phi_H$. These calculations reveal that when the applied field $H_x$ is not along a crystallographic $ab$-plane axis, the field responses of the moments in the two orthogonal domains A and B are quite different. Additional measurements along these lines would be of interest.

\section{V. CONCLUDING REMARKS}

The Eu square-lattice compound EuGa$ _4$ exhibits A-type antiferromagnetic order at a Néel temperature $T_N = 16.4(2)$ K with the moments aligned in the $ab$ plane. A magnetic-field-induced anomaly is observed at low fields in the $M_{ab}(H)$ isotherms at $T = 2$ K. We infer that the $H = 0$ A-type magnetic structure consists of four AFM domains associated with a fourfold in-plane magnetic anisotropy, where each domain consists of antiparallel moments in adjacent $ab$ planes along the $c$ axis. On application of an in-plane magnetic field $H_x$, the collinear moments in each of the AFM domains gradually orient to become perpendicular to $H_x$ at a critical field $H_{c1}$, yielding a nonlinear $M_{ab}(H)$ at $T = 2$ K. The $M_{ab}(H)$ behavior along the two $ab$-plane crystallographic symmetry directions $[1,0,0]$ and $[1,1,0]$ are quite different with respective critical fields $H_{c1,[1,0,0]} = 4.8$ kOe and $H_{c1,[110]} = 0.85$ kOe, respectively. The experimental $M_{ab}(H)$ data for $H \parallel [1,0,0]$ and $[1,1,0]$ were successfully modeled by a theory incorporating the fourfold tetragonal in-plane magnetic anisotropy and associated AFM domains. However, the calculations predict a first-order transition when the in-plane field $H_x$ is at a finite angle to one of the $[1,0,0]$ or $[0,1,0]$ directions. Since the theoretical calculations are done for $T = 0$ K whereas the experiments were performed at 2 K, it would be interesting to calculate the finite temperature effects on the $M(H)$ behavior. In addition, the effects of Eu-moment rotations for the small magnetic fields discussed here on the topological properties of EuGa$ _4$ would be very interesting to explore.

Similar moment-reorientation effects due to small fields aligned in the $ab$ plane have recently been observed in the trigonal A-type antiferromagnets EuMg$ _2$Bi$ _2$ and EuMg$ _2$Sb$ _2$ containing $S = 7/2$ Eu$ ^{2+}$ spins with the moments aligned in the $ab$ plane. It seems likely that $M(H)$ measurements for other layered Eu$ ^{2+}$ spin-7/2 compounds with A-type AFM order and moments aligned in the layer plane would also exhibit low-field effects similar to those described here and in Ref. [33].

\section{ACKNOWLEDGMENTS}

This research was supported by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering. Ames National Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. DE-AC02-07CH11358.
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