Antiferromagnetic domains in a single crystal of the A-type spin-7/2 trigonal topological insulator $EuSn_2As_2$

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$EuSn_2As_2$ is a trigonal A-type antiferromagnetic topological insulator with the moments aligned in the ab plane and with a Néel temperature $T_N = 23.5$ K. Here we report that an $EuSn_2As_2$ crystal exhibits a broad peak at $H_{c1} = 1100$ Oe in the field derivative dM_{ab}/dH of the ab-plane magnetization $M_{ab}(H)$ at temperature $T = 2$ K, demonstrating the presence of trigonal antiferromagnetic domains. We model these $M_{ab}(H, T = 2\,\mathrm{K})$ data and obtain the trigonal anisotropy-energy coefficient K_3 that is 13.3 and 3.7 times larger than those we previously reported for single crystals of the trigonal compounds $EuMg_2Sb_2$ and $EuMg_2Bi_2$, respectively.

I. INTRODUCTION

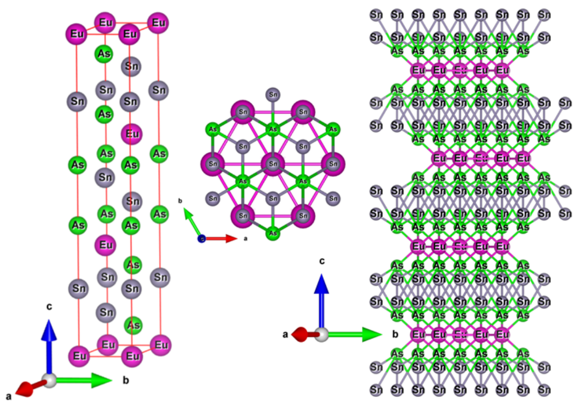

FIG. 1. Trigonal crystal structure of $EuSn_2As_2$ [2]. The Eu atoms are colored dark purple, the Sn atoms blue, and the As atoms green. The Eu atoms form a hexagonal (triangular) layer situated between two buckled SnAs honeycomb layers, with four Eu layers within each c-axis height. The Eu magnetic structure is A-type with the Eu^{2+} moments aligned in the ab plane [2]. The height of the magnetic unit cell is twice the c-axis lattice parameter.

$EuSn_2As_2$ is a topological insulator that has been reported to exhibit Dirac surface states at energies within the bulk band gap from angle-resolved photoemission spectroscopy (ARPES) measurements [1]. The compound crystallizes in the trigonal $NaSn_2As_2$-type structure with space group $R\bar{3}m$ shown in Fig. 1 [2]. It orders antiferromagnetically below its Néel temperature $T_N \approx 23.5$ K [2, 3]. Neutron-diffraction measurements [2] showed that the antiferromagnetic (AFM) structure is A-type, where the Eu^{+2} moments have spin $S = 7/2$, spectroscopic splitting factor $g = 2$, and ordered moment $\mu = gS\mu_B = 7\mu_B$, where μ_B is the Bohr magneton. The Eu moments are aligned ferromagnetically in an ab plane layer with the Eu moments in adjacent layers along the c axis aligned antiferromagnetically [2]. Other studies of the magnetic and thermodynamic properties of $EuSn_2As_2$ single crystals were also presented in Ref. [2].

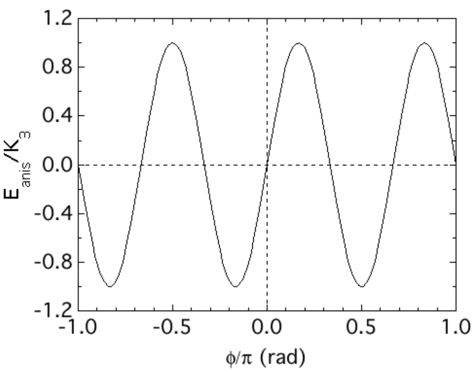

FIG. 2. Magnetic anisotropy free energy E_{anis} of ferromagnetic moments aligned at an angle ϕ with the positive x axis, normalized by the trigonal anisotropy constant K_3 in Eq. (1).

The related topological compounds $EuMg_2Sb_2$ and $EuMg_2Bi_2$ crystallize in the trigonal $CaAl_2Si_2$-type crystal structure with space group $P\bar{3}m1$ and order antiferromagnetically at $T_N = 8.2$ and 6.7 K, respectively. Neutron-diffraction measurements demonstrated that both compounds exhibit A-type AFM order with the Eu moments aligned in the ab plane [4, 5]. Remarkably, crystals of both compounds showed a distinct positive curvature in $M(H_{ab})$ isotherms at low fields $0 \leq H_{ab} \lesssim H_{c1}$ at $T = 1.8$ K, where $H_{c1} \approx 220$ and ≈ 465 Oe for $EuMg_2Sb_2$ and and $EuMg_2Bi_2$, respectively [6]. At higher fields up to the respective saturation critical fields which were $H_c^c = 3.4$ T for $EuMg_2Sb_2$ at 1.8 K [4] and 4.0 T at 2 K for $EuMg_2Bi_2$ [7], it was found that $M_{ab} = \chi(T_N)H_{ab}$ as expected from molecular-field theory [8] since the moments were initially nearly perpendicular to $\mathbf{H}_{ab}$ at the respective critical field H_{c1}.

These observations motivated us to consider the presence of equally-populated trigonal AFM domains below the respective T_N of $EuMg_2Bi_2$ and $EuMg_2Sb_2$. These

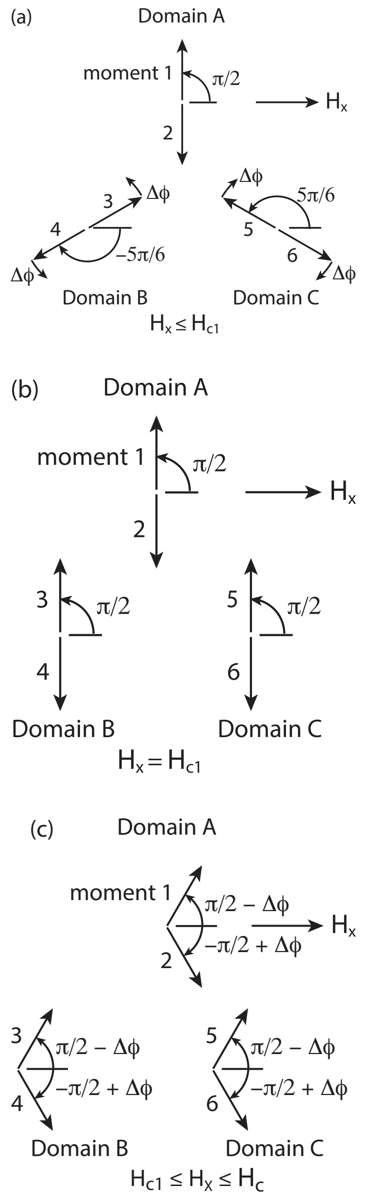

FIG. 3. (a) Domain model for trigonal A-type antiferromagnets in $H_x = 0$ [6]. Moments 1, 4, and 6 are the magnetic moments in a central layer in zero field according to Fig. 2, whereas moments 2, 3, and 5 represent moments in adjacent layers in the collinear A-type AFM structure. The arrows and $\Delta\phi$ indicate the direction of rotation of the moments in a domain for $H_x \le H_{c1}$. (b) Orientation of the moments when $H = H_{c1}$, apart from a slight canting toward H_x that gives rise to the measured magnetization. (c) Rotation of the moments in each domain towards H_x for $H_{c1} < H_x < H_{ab}^{c}$, where H_{ab}^{c} is the ab-plane saturation (critical) field.

domains result from the existence of a trigonal anisotropy free energy of the form

$$F(\phi) = K_3 \cos(3\phi), \qquad (1)$$

where K_3 is the anisotropy energy amplitude and ϕ is the angle between moments in an A-type AFM domain and the x axis, which is assumed to be the axis in which a magnetic field H_x is applied. A plot of $F(\phi)$ is shown in Fig. 2, where three minima in $F(\phi)$ are at $\phi/\pi = -5/6$, $-1/6$, and $1/2$.

The critical field $H_x = H_{c1}$ is the ab-plane field at which all moments in each domains become nearly perpendicular to H_{ab}, apart from a small canting $\lesssim 1^\circ$ toward the field that gives rise to the observed magnetization. The $M(0 \le H_{ab} \le H_{c1})$ data for both $EuMg_2Sb_2$ and $EuMg_2Bi_2$ were successfully fitted by a model in which the positive curvature in $M(H_{ab})$ below H_{c1} arose from the field response of the Eu spins in trigonal domains in Fig. 3 [6]. These fits allowed the respective ab-plane trigonal anisotropy parameters K_3 to be determined.

II. THEORY

Here we analyze the low-field $M(H_{ab}, T = 2\ \mathrm{K})$ data for $EuSn_2As_2$ [2] in Fig. 4(b) by our model [6] for the magnetic response of the moments in isolated trigonal domains at $T = 0$ K and derive therefrom the value of its trigonal anisotropy constant K_3. We will then compare this value with the values of K_3 previously obtained for trigonal domains in $EuMg_2Sb_2$ and $EuMg_2Bi_2$ [6].

With increasing ab-plane field H_x at $T = 2$ K, the three AFM domains initially at 120° to each other as shown in Fig. 3(a) begin to align nearly perpendicular to H_x, which according to Fig. 4(c) occurs at $H_x \approx 1100$ Oe. Then with a further increase in H_x the collinear spins in each domain progressively cant towards $H_x = H_{ab}$ until they reach FM alignment at the critical field $H_{ab}^{c} \approx 3.7$ T according to Fig. 4(a).

Following Ref. [6], the anisotropy energy (1) averaged over the three domains and then normalized by K_3 is

$$\frac{E_{\rm anis\ ave}}{K_3} = -\frac{1}{3}[1 + 2\cos(3\Delta\phi)], \qquad (2)$$

and the magnetic energy normalized by K_3 is

$$\frac{E_{\rm mag\,ave}}{K_3} = -\frac{\chi_\perp H_x^2}{3K_3}\left[1 + 2\sin^2\left(\frac{\pi}{6} + \Delta\phi\right)\right], \qquad (3)$$

where $\Delta\phi$ in Fig. 3(c) is the rotation angle of the moments due to a particular H_x and $\chi_\perp = \chi_c$ is the magnetic susceptibility perpendicular to the ab plane, which according to molecular-field theory [8] is $\chi_c = \chi(T_{\rm N})$. Then setting

$$h_x = \chi_\perp H_x^2 / K_3 \qquad (4)$$

gives

$$\frac{E_{\rm mag\,ave\,2}}{K_3} = -\frac{h_x}{3}\left[1 + 2\sin^2\left(\frac{\pi}{6} + \Delta\phi\right)\right]. \qquad (5)$$

Thus the total average energy $E_{\rm tot\,ave}$ normalized by K_3 is

$$\frac{E_{\rm tot\,ave}}{K_3} = \frac{E_{\rm anis\ ave}}{K_3} + \frac{E_{\rm mag\,ave\,2}}{K_3}. \qquad (6)$$

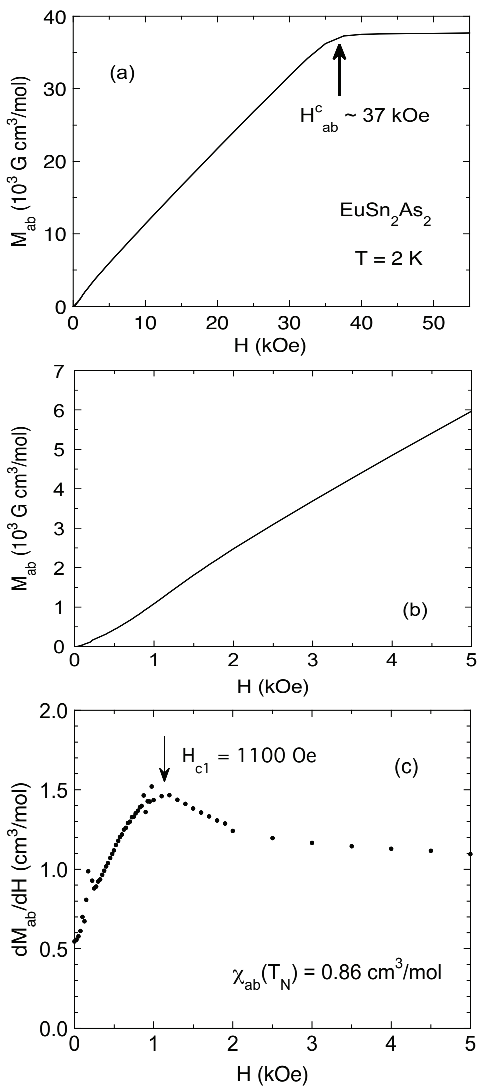

FIG. 4. (a) Magnetization versus x-axis field H_x up to 55 kOe. The critical field for the ab-plane field is $\approx$ 37 kOe. (b) Expansion of $M_{ab}(H)$ for $H \leq 5$ kOe. (c) Low-field slope dM_{ab}/dH, yielding $H_{\rm c1} \approx 1100$ Oe. The small peak at $H \approx 200$ Oe was also observed to occur for dM_{ab}/dH with $\mathbf{H} \parallel c$ [2] and was postulated to arise from the presence of magnetic defects.

Then using Eqs. (2) and (5) and minimizing $E_{\rm tot\,ave}/K_3$ in Eq. (6) with respect to $\Delta\phi$ gives a simple solution for h_x in terms of $\Delta\phi$ as

$$h_x(\phi) = 3\csc[(\pi + 6\Delta\phi)/3]\sin(3\Delta\phi). \tag{7}$$

From Figs. 3(a) and 3(b), we have $\Delta\phi = \pi/3$ when $H_x = H_{\rm c1}$, *i.e.*, when all moments are approximately perpendicular to H_x. Then Eqs. (4) and (7) and give

$$h_x(\pi/3) = \frac{9}{2} = \frac{\chi_\perp H_{\rm c1}^2}{K_3}. \tag{8}$$

This value is per Eu^{2+} ion, whereas the measured $\chi_\perp = \chi(T_{\rm N})$ is per mole of Eu ions, so Eq. (8) becomes

$$\frac{9}{2} = \frac{\chi_\perp H_{\rm c1}^2}{N_{\rm A} K_3}, \tag{9}$$

where $N_{\rm A}$ is Avogadro's number. Then using $H_{\rm c1} = 1100$ Oe from Fig. 4(c), the anisotropy-corrected $\chi_\perp = \chi(T_{\rm N}) = 0.86$ cm^3/mol [2], and solving for K_3 gives

$$\begin{aligned} K_3 &= \frac{\chi_\perp H_{\rm c1}^2}{(9/2)N_{\rm A}} \\ &= 2.4 \times 10^{-7}\ \text{eV/Eu}. \end{aligned} \tag{10}$$

This K_3 value is 13.3 and 3.7 times larger than those found for single crystals of the two previously-studied trigonal A-type antiferromagnets $EuMg_2Sb_2$ and $EuMg_2Bi_2$, respectively [6].

III. CONCLUDING REMARKS

The present work is part of an ongoing effort to characterize the ab-plane magnetic-field dependence of Eu^{2+} and Gd^{3+} spin $S = 7/2$ trigonal and tetragonal A-type antiferromagnets with the moments aligned in the ab plane. In addition to the present study of $EuSn_2As_2$, these compounds have so far included trigonal $EuMg_2Sb_2$ and $EuMg_2Bi_2$ [6] as noted in the text, and also trigonal $EuAl_2Ge_2$ [9] and tetragonal $EuGa_4$ [10]. In future work, it would be interesting to determine if the ab-plane magnetic-field-induced changes in the domain structure also affects the topological properties of relevant A-type antiferromagnets.

ACKNOWLEDGMENTS

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