

Modification of charge and spin textures by light chemical substitution in $\text{Eu}(\text{Al}_{1-x}\text{Ga}_x)_4$ ($x = 0.1$)

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We present the results of a resonant x-ray-diffraction experiment, resolving both charge and spin textures in the intermetallic topological magnet $\text{Eu}(\text{Al}_{1-x}\text{Ga}_x)_4$ ($x = 0.1$). Below ≈ 75 K the system develops a charge-density wave (CDW) with propagation vector $\mathbf{k}_{\text{CDW}} \approx (0, 0, 0.18)$. The CDW order parameter grows monotonically on cooling until ≈ 15 K, when a sudden decrease in the CDW amplitude occurs. Pairs of magnetic satellites of the $(0, 0, 8)$ Bragg reflection corresponding to two distinct domains, $\mathbf{k}_1 = (\pm\delta_m, 0, 0)$, $\mathbf{k}_2 = (0, \pm\delta_m, 0)$, $\delta_m = 0.2002(4)$ were studied at the Eu L_3 edge, appearing below $T_N = 14.8$ K. Our measurement of T_N is exactly coincident with the sudden drop in the CDW amplitude, which suggests strong coupling between the charge and spin orders, as observed in other compounds of the $\text{Eu}(\text{Al}_{1-x}\text{Ga}_x)_4$ series. Azimuthal measurements were consistent with a single helical spin arrangement having an elliptical envelope of $\mu_Y/\mu_Z = 1.19(6)$ for the $\mathbf{k}_2$ domain, and a tilted helical (helical) spin arrangement for the $\mathbf{k}_1$ domain, with $\mu_Y/\mu_Z = 1.14(4)$ and $\mu_X/\mu_Z = 0.20(2)$ that may be hidden for the $\mathbf{k}_2$ domain due to multiple subdomains. Temperature evolution of the magnetic satellite intensities in linear and circularly polarized light found the respective ratio to be invariant with temperature, suggesting a single helicoidal magnetic phase throughout the measured temperature range. This behavior is unlike the $x = 0$ material, in which a spin-density wave forms first, transitioning to a helical ground state on cooling through intermediate phases. Future theoretical work on the Eu electronic ground state, supported by related experiments, will help understand the effects of Ga substitution on the evolution of the magnetic structure.

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I. INTRODUCTION

Topological magnetic states such as skyrmion-type structures [1–3] have attracted considerable interest due to their exotic fundamental physics [4] and potential use in spintronic technology [5–8]. The precursor to topological magnetism is typically a helicoidal magnetic structure stabilized by the Dzyaloshinskii-Moriya interaction in a noncentrosymmetric material [9]. In the search for new topological magnetic phases, it is important to explore and understand the stability of these precursor phases, especially given the more recent discovery of topological magnetism in centrosymmetric materials [10,11].

A material family of particular interest is the $\text{Eu}(\text{Al}_{1-x}\text{Ga}_x)_4$, $x \in [0, 1]$. All members of this intermetallic solid solution adopt the same tetragonal crystal structure at high temperature, with centrosymmetric space group $I_{4/m}^1mm$ and approximate lattice parameters $a = b = 4.4$ Å, $c = 11.1$ Å [12]. Compounds with $x = 0, 0.1$ [13], and possibly 0.5 [14], have been shown to host skyrmion lattices in externally applied magnetic fields, with the $x = 0$ compound hosting a particularly rich magnetic phase diagram. The source of magnetism in these rare-earth compounds is the Eu^{2+} ion, with the spin-only ground state $^8S_{7/2}$ [15].

Previous reports showed [16,17] that EuAl_4 went through a series of four phase transitions on cooling in zero magnetic field [17]. A charge-density wave (CDW) developed below $T_{\text{CDW}} = 145$ K, with microscopic details yet to be determined. At the Néel temperature of $T_N = 15.4$ K, two incommensurate single- $\mathbf{k}$ transverse spin-density waves (SDWs) form orthogonal $\mathbf{k}$ domains. The propagation vectors are $\mathbf{k}_{\pm} = (\delta_1, \pm\delta_1, 0)$, $\delta_1 \approx 0.09$, respectively, with magnetic moments also lying in the crystallographic ab plane, ($\boldsymbol{\mu} \perp c$ and $\perp \mathbf{k}$). Below $T_2 = 13.2$ K a second magnetic phase

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transition occurs, characterized by a rotation of the SDWs now propagating along $\mathbf{k} = (\delta_2, 0, 0)$ and $\mathbf{k} = (0, \delta_2, 0)$, $\delta_2 \approx 0.18$, and with $\boldsymbol{\mu} \parallel c$. At $T_3 = 12.2$ K the collinear spin order collapses: moment magnitudes rotate in a plane perpendicular to their propagation vector, forming an elliptical helix. Two *helimagnetic* $\mathbf{k}$ domains develop across the sample with a common helicity and propagation vectors $\mathbf{k} = (\delta_3, 0, 0)$ and $\mathbf{k} = (0, \delta_3, 0)$, $\delta_3 \approx 0.17$, respectively. At $T_4 \approx 10$ K, the helimagnetic domains retain their structure; however, the chirality of the helices spontaneously reverses across the whole sample, and a sharp step occurs in the propagation vector amplitude to $\delta_4 \approx 0.19$. Furthermore, it was observed that the propagation vector of the CDW changes from incommensurate to commensurate at T_4 [16].

Such correlation between magnetic and charge modulations was also reported in EuAl_2Ga_2 [18], in which SDW order appears immediately below T_N , followed by a phase transition to a cycloidal ground state. This compound also showed evidence for skyrmion phases in applied magnetic field [14]. It is apparent, therefore, that similar phenomenological coupling occurs in both $x = 0$ and 0.5 compounds, despite drastically different magnetic phase diagrams. Hence, studies of the intermediate compositions may yield considerable insight into the physics of this complex material system.

It was reported recently that the lightly substituted $x = 0.1$ material stabilizes a square skyrmion lattice (sSKL) in applied magnetic field. The sSKL is identical to that observed in pure EuAl_4 , however located at a slightly different region in the magnetic-field—temperature-phase diagram (Fig. 1 in Ref. [13]). The distinctive feature of both compounds is that unlike in other materials [19], the skyrmion phases and the maximal topological Hall effect do not overlap on the H - T phase diagram, which is the usual empirical method of identifying skyrmion phases. It is also proposed that just like in the $x = 0$ compound, the ground state for $x = 0.1$ is best described by two types of helical domains, propagating along **a** and **b**, respectively, with the possibility of numerous phase transitions close to $T_N = 16$ K [12,13].

In this paper, we report a resonant x-ray-diffraction study on the $x = 0.1$ compound. We show that by 10% gallium substitution, the CDW transition temperature is suppressed by $\approx 50\%$, and the zero-field magnetic order changes drastically. A single incommensurate *general helicoidal* magnetic phase appears below $T_N = 14.8$ K with two $\mathbf{k}$ domains propagating along **a** and **b**. Analogous to the $x = 0$ and 0.5 compounds, strong coupling of the magnetic and charge modulation was observed in this material.

The paper is organized as follows. In Sec. II we describe the experimental setup and details of the measured crystal. In Sec. III a quantitative description of our observations is given. Section IV provides analysis and interpretation of these results. Finally, in Sec. V we draw conclusions.

II. EXPERIMENT

Single crystals of $\text{Eu}(\text{Al}_{0.9}\text{Ga}_{0.1})_4$ were synthesized by the self-flux technique described in Ref. [20]; crystal characterization can be found in Ref. [12]. A crystalline sample with approximate dimensions of $1.5 \text{ mm} \times 1.5 \text{ mm} \times 0.8 \text{ mm}$ was selected for resonant x-ray-diffraction experiments at the

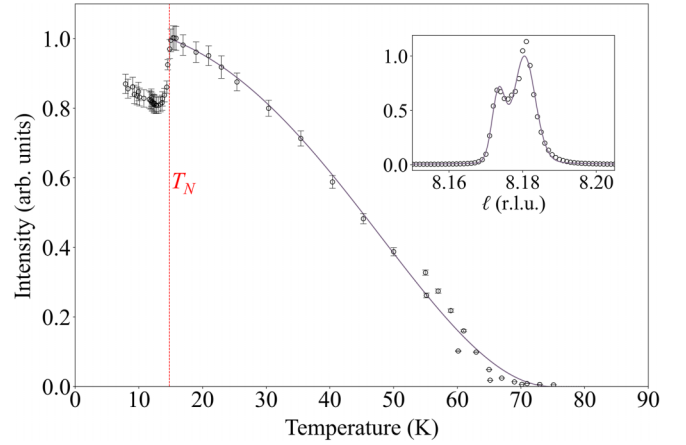


FIG. 1. Evolution of the $(1, 1, 8.18)$ CDW diffraction intensity as function of temperature. Inset: An example the CDW intensity measured along k at $T_{\text{base}} = 7.5$ K. The peak splitting was also observed for the $(1, 1, 8)$ structural peak, suggesting it originates in structural inhomogeneities.

I16 beamline at Diamond Light Source. The energy of the incoming x-rays was set just below the Eu^{2+} L_3 edge; the energy of the dipole-allowed, E_1 - E_1 , transition of an electron from the $2p_{3/2}$ state to the $5d_{3/2}$ and $5d_{5/2}$ states, at 6.977 keV. The polarization of the incident x-ray beam was switched between linear horizontal (σ), linear vertical (π), and circular using a 400- μm -thick diamond phase plate positioned upstream of the sample. The sample was mounted on the cold finger of a closed-cycle refrigerator with a base temperature of $T_{\text{base}} = 7.5$ K, aligned in reflection geometry with the $(0, 0, \ell)$ zone axis surface normal. Diffraction measurements were then performed about the $(0, 0, 8)$ and $(1, 1, 8)$ structural Bragg reflections.

III. RESULTS

Bragg diffraction intensities in the vicinity of the $(0, 0, 8)$ reciprocal-space position were measured at room temperature, and found to be consistent with the $I_{\text{m}}^4 mm$ parent crystal structure. On cooling below $T_{\text{CDW}} = 75$ K, weak diffraction satellites of the $(1, 1, 8)$ Bragg reflections were found with incommensurate propagation vector $\mathbf{k}_{\text{CDW}} \approx (0, 0, 0.18)$. We note that the CDW peak profile (inset to Fig. 1) is split by ≈ 0.01 reciprocal lattice unit (r.l.u.), similar to peak splitting observed in the main structural Bragg peaks, and is hence due to slight crystalline inhomogeneities that become apparent at such high instrumental resolution. These satellite intensities are consistent with the onset of a CDW, as reported for both $x = 0$ and 0.5 compounds [17,21,22]. The integrated intensity of the $(1, 1, 8.18)$ CDW reflection was measured as a function of temperature, as shown in Fig. 1. On cooling below T_{CDW} the intensity appears to grow monotonically, consistent with mean-field behavior. On further cooling a sudden drop in the intensity can be seen just below 15 K, coincident with the expected phase transition to long-range magnetic order [12], while no significant change in the CDW wave vector was observed. This suggests coupling between the two, as observed in other compounds of the series [21].

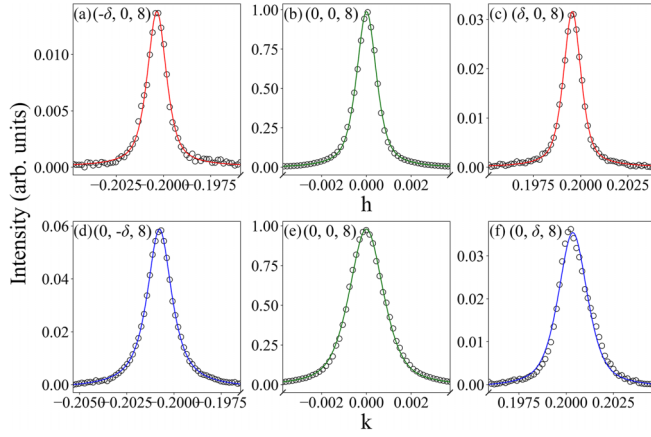


FIG. 2. The $(0, 0, 8)$ structural diffraction intensity (b), (e) (green) and its four magnetic satellites (a), (c) (red) and (d), (f) (blue), measured in the h - and k reciprocal-space directions. The respective magnetic wave vector was measured to be $\delta_h = \delta_k = 0.2002(4)$. Measurements were carried out at $T_{\text{base}} = 7.45 \pm 0.08$ K. The above color code is adopted throughout the paper unless stated otherwise, to differentiate the two $\mathbf{k}$ domains propagating along h and k , respectively. Note that the data were renormalized to the maximum intensity of the $(0, 0, 8)$ peak measured along k .

Below $T_N = 14.8$ K, a further four weak satellites appeared at positions $(\pm\delta_m, 0, 8)$ and $(0, \pm\delta_m, 8)$, shown in Fig. 2. Fitting the data resolved incommensurate propagation vectors, with $\delta_m = 0.2002(4)$. The magnetic origin of these satellite reflections was confirmed via the presence of an energy resonance at 6.9715 keV, close to the Eu^{2+} L_3 edge; see Fig. 3. While the value of δ_m approximates the commensurate position of $1/5$, the incommensurate nature of the propagation vectors is supported by the observation that the center of the magnetic peaks had a slight temperature dependence; see Fig. 8 of Appendix B. Furthermore, measurement of the diffraction intensities using oppositely polarized circular light

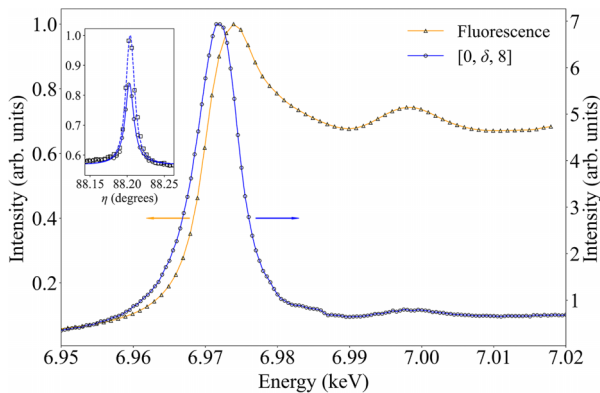


FIG. 3. The energy dependence of the $(0, \delta_m, 8)$ magnetic satellite intensity (blue), showing a large resonant enhancement at 6.9715 keV, just below the L_3 edge of the known magnetic ion Eu^{2+} at 6.977 keV. For reference, the x-ray fluorescence measured away from the diffraction condition is shown (orange). Inset: Diffraction intensities of opposite polarized circular light (dashed and solid lines, respectively). Both datasets were measured at T_{base} , and normalized to the maximum resonant intensity at $\psi = 250^\circ$ (see Fig. 7 for definition of azimuthal angle ψ).

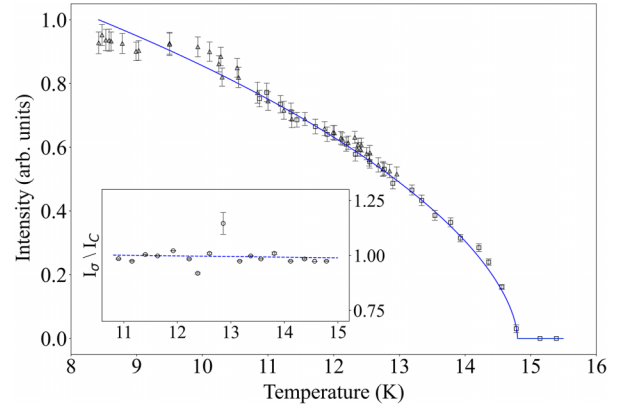


FIG. 4. The temperature evolution of the $(0, 0.2, 8)$ magnetic satellite measured on cooling. Long-range magnetic order develops at $T_N = 14.8$ K. Inset: Normalized ratio of intensities measured with linear (I_σ) and circularly polarized (I_c) incident light.

showed dichroic response, indicative of helicoidal magnetic order.

The intensity of the $\mathbf{k}_2 = (0, 0.2, 8)$ magnetic satellite was measured as a function of temperature (Fig. 4). The magnetic diffraction intensity was found to grow monotonically down to the lowest measured temperature, consistent with a single magnetic phase. To further confirm this result, the $\mathbf{k}_2$ magnetic diffraction intensity was measured using both linearly and circularly polarized incident x-rays, as a function of temperature. The ratio of these two intensities is strongly sensitive to transitions between SDW and helimagnetic structures with the same propagation vector, and also between helimagnetic structures of different chirality. The inset to Fig. 4 shows that this ratio is constant in the measured temperature range, hence confirming a single helimagnetic phase.

To resolve the magnetic structure, an *azimuthal measurement* (rotation of the sample about the scattering vector) was carried out of the $\mathbf{k}_1 = (-0.2, 0, 8)$ and $\mathbf{k}_2 = (0, 0.2, 8)$ magnetic satellites at T_{base} . The integrated intensities measured as a function of azimuthal angle, ψ , are shown in Fig. 5. The state of the incident x-ray beam was σ polarized, and the scattered beam was measured without polarization analysis having confirmed that the magnetic scattering resides fully in the rotated $\sigma - \pi'$ channel; see Fig. 7 for definition of polarization vectors. Limits in ψ were determined by the experimental geometry. As derived in Appendix A, we model the azimuthal dependence of the measured intensities using the following equations:

$$I(-\delta_m, 0, 8) = A_1 |\cos(\vartheta) \cos(\psi) \Omega_+ - \cos(\vartheta) \sin(\psi) \Omega_0 + \sin(\vartheta) \Omega_-|^2, \quad (1)$$

$$I(0, +\delta_m, 8) = A_2 |\sin(\vartheta) \cos(\psi) \Omega_+ - \sin(\vartheta) \sin(\psi) \Omega_0 - \cos(\vartheta) \Omega_-|^2, \quad (2)$$

where ϑ is half of the scattering angle, ψ is the azimuthal angle, and Ω_i are defined as

$$\Omega_+ = \sin(\chi) \langle \mu_Z \rangle + \cos(\chi) \langle \mu_X \rangle, \quad (3)$$

$$\Omega_0 = \langle \mu_Y \rangle, \quad (4)$$

$$\Omega_- = \cos(\chi) \langle \mu_Z \rangle - \sin(\chi) \langle \mu_X \rangle, \quad (5)$$

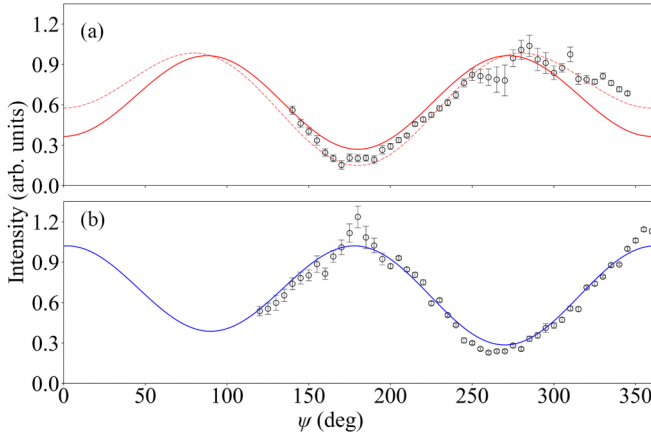


FIG. 5. The azimuthal dependence of the (a) $\mathbf{k}_1 = (-0.2, 0, 8)$ and (b) $\mathbf{k}_2 = (0, 0.2, 8)$ magnetic satellite intensities. Elliptical helix model fit is shown as solid red and blue lines. The helicoidal model fit is shown by the dashed line on (a). The zero point in azimuth corresponds to a sample orientation for which the $(1, 0, 0)$ reciprocal-space vector points away from the source, and is within the scattering plane.

with the χ angle, depending on the position of the magnetic satellite, calculated as

$$\cos(\chi) = \frac{1}{\sqrt{1 + \left(\frac{\delta c}{8a}\right)^2}}, \quad (6)$$

where a and c are the lattice parameters in r.l.u. The magnetic moment of the Eu^{2+} ion at the origin of the zeroth unit cell is expressed as $\boldsymbol{\mu} = (\mu_X, \mu_Y, \mu_Z)$. Note that the Cartesian coordinates $\{X, Y, Z\}$ are parallel to $\{\mathbf{a}, \mathbf{b}, \mathbf{c}\}$ for domain 1, and parallel to $\{\mathbf{b}, -\mathbf{a}, \mathbf{c}\}$ for domain 2; see the geometry described in detail in Appendix A in Fig. 7. $A_1, A_2, \mu_X, \mu_Y, \mu_Z, \varphi_X, \varphi_Y, \varphi_Z$ are unknown parameters, with the latter six represented as three complex Fourier modes $\langle \mu_j \rangle = \mu_j e^{i\varphi_j}$ to encapsulate the relative phase difference between their modulation. The model is properly constrained without loss of generality by setting $\mu_Z = 1$ and $\varphi_Z = 0$. Initial calculations showed that the modulation of μ_Y must be $\pi/2$ phase shifted with respect to that of μ_Z , while a small μ_X component is either in phase or antiphase with μ_Z . This is fully consistent with the circular dichroism reported in Fig. 3. The data were fit by an elliptical helix model, in which A_1 and A_2 were freely refined, μ_Y was refined but constrained to be the same for both $\mathbf{k}_1$ and $\mathbf{k}_2$ azimuthal datasets, and μ_X was set to zero for both datasets. The resulting fit is shown by the solid lines in Fig. 5, and the parameters are summarized in Table I. This elliptical helix model was in excellent agreement with the $\mathbf{k}_2$ azimuthal data, but failed to reproduce some features of the $\mathbf{k}_1$ data. The model was therefore refit with a small μ_X component freely refined for the $\mathbf{k}_1$ data. The introduction of this component leads to a helicoidal magnetic structure in the $\mathbf{k}_1$ domain, which can be thought of as a helix with the plane of rotation tilted about z towards the direction of propagation. In this case the fit was significantly improved ($\chi^2 = 1.56 \rightarrow \chi^2 = 0.88$), as shown by the dashed lines in Fig. 5. The respective parameters are also given in Table I.

TABLE I. Summary of refined magnetic structure parameters for the elliptical helix ($\chi^2 = 1.56$) and the helicoidal ($\chi^2 = 0.88$) models, as described in the main text.

	Magnitude	Relative phase ($\varphi - \varphi_Z$)
Elliptical helix		
μ_Y/μ_Z	1.19(6)	$+\frac{\pi}{2}$
A_1	1.5(1)	
A_2	1.6(1)	
Elliptical helix ($\mathbf{k}_1$) and elliptical helix ($\mathbf{k}_2$)		
μ_Y/μ_Z	1.14(4)	$+\frac{\pi}{2}$
$\mu_X^{(1)}/\mu_Z$	0.20(2)	0
A_1	1.64(7)	
A_2	1.70(7)	

IV. DISCUSSION

In good agreement with previous investigations [13], we found that the azimuthal dependence of our data was well described by a helix with an elliptical envelope (i.e., $\frac{\mu_Y}{\mu_Z} \neq 1$), which is also the ground state of EuAl_4 . However, the fit for the $\mathbf{k}_1$ domain was considerably improved by allowing a finite spin component parallel to the axis of modulation (see Table I), leading to a helicoidal spin structure, as shown in Fig. 6.

The apparent difference between $\mathbf{k}_1$ and $\mathbf{k}_2$ domains is surprising, and we suggest three possible interpretations. First, the magnetic structure of the $\mathbf{k}_2$ domain is indeed different from that of the $\mathbf{k}_1$ domain. The two domains span different regions of the sample and are associated with different directions of propagation. Hence, the observed difference could be due to subtle anisotropic inhomogeneities within the material. Indeed, Carneiro *et al.* [23] have performed measurements of other materials in this family and have identified different behavior of the two domains. Second, both $\mathbf{k}$ domains are helicoidal, but in our measurement the μ_X component of the $\mathbf{k}_2$ domain is hidden due to averaging over $\pm\mu_X$ subdomains (for this reason, the helicoidal structure could have been missed in previous studies). In this case, the refined value of $\frac{\mu_X}{\mu_Z}$ provides a lower limit on the relative value of μ_X . Third, the refined μ_X component could be an artifact of a systematic uncertainty due to beam movement between different populations of $\mathbf{k}_1$ and $\mathbf{k}_2$ domains in the azimuthal diffraction measurement. Finally, it should be emphasized that in our model we only consider $E1$ - $E1$ scattering events; however, it is possible that including higher-order terms could refine our conclusion.

The helical and helicoidal magnetic structures have distinct symmetries. The elliptical helix transforms as the direct sum $m\Sigma_3 \oplus m\Sigma_4$, where $m\Sigma_i$ are irreducible representations of the parent space group associated with the $\Sigma = (k_x, 0, 0)$ line of symmetry. The respective magnetic space group is $I222.1'(0, 0, g)00ss$, with basis $\{(0, 1, 0, 0), (0, 0, -1, 0), (-1, 0, 0, 0), (0, 0, 0, 1)\}$ relative to the parent tetragonal lattice. The helicoidal structure transforms as $m\Sigma_2 \oplus m\Sigma_3 \oplus m\Sigma_4$, with the respective magnetic space group $B2.1'(a, b, 0)0s$, with basis $\{(1, 0, -1, 0), (0, 0, 1, 0), (0, -1, 0, 0), (0, 0, 0, 1)\}$. As one

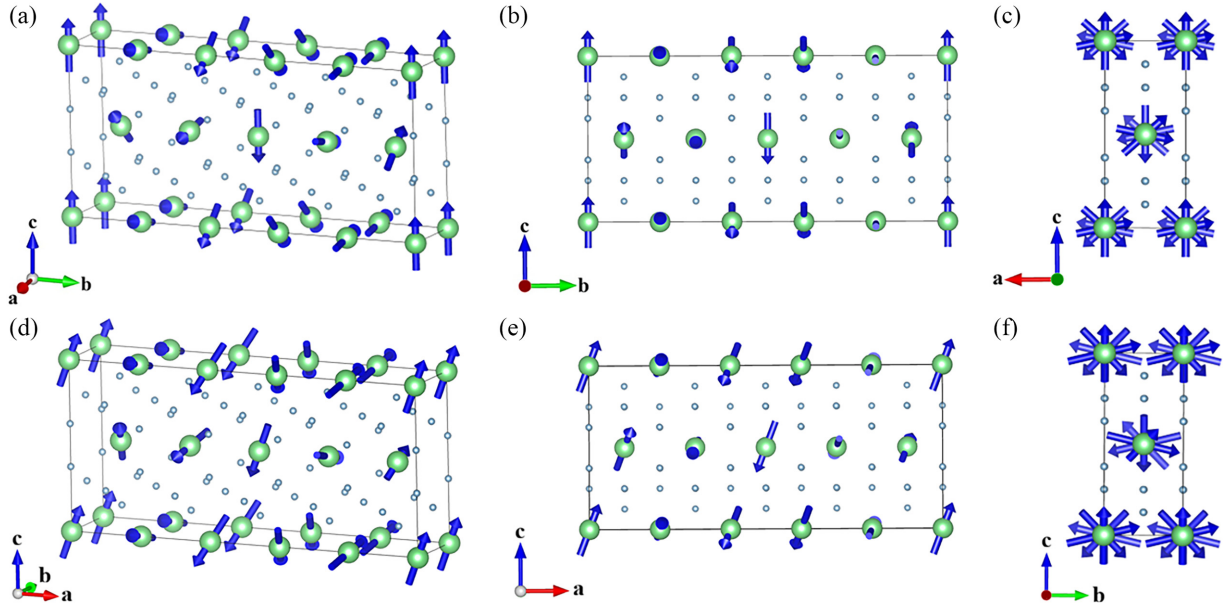


FIG. 6. Schematic diagram of the ground-state magnetic structure of the domain modulated along **b**, corresponding to (a)–(c) helical structure, and along **a**, corresponding to (d)–(f) tilted helicoidal structure with nonzero longitudinal component. Large green spheres represent the magnetic Eu^{2+} ions, while small pale-blue spheres represent the Al or Ga ions.

might anticipate, the helicoidal structure has lower monoclinic symmetry compared to the orthorhombic magnetic space group of the helix. In the case of second-order phase transitions, lower-symmetry solutions are disfavored. However, in both cases the transition at T_N must be first order, and hence one cannot preclude on symmetry grounds alone the possibility of a lower-symmetry structure stabilized by higher-order interactions.

The ratio of σ and circularly polarized intensities (Fig. 4, inset) was measured as a function of temperature and at an azimuthal angle of $\psi = 220^\circ$. In this geometry the above ratio is sensitive to both spontaneous changes in chirality, and phase transitions from achiral collinear spin-density waves to chiral helicoids [see Eq. (A23) of Appendix A]. The ratio of intensities is invariant with temperature to good approximation, indicating that, unlike pure EuAl_4 , the 10% Ga substitution results in the helicoidal magnetic structure appearing immediately below T_N (up to the possibility of very narrow phases in the vicinity of the Néel temperature, below our temperature resolution and intensity noise floor). Furthermore, the constant ratio also confirms that no chirality reversal occurred in the measured temperature range.

V. CONCLUSION

We have demonstrated that $\text{Eu}(\text{Al}_{1-x}\text{Ga}_x)_4$ ($x = 0.1$) hosts a charge-density wave on cooling below 75 K, with significant coupling to magnetism when long-range magnetic order appears at $T_N = 14.8$ K. Two **k** domains are formed, propagating along **a** and **b**, respectively. Azimuthal measurements confirmed that the ground-state magnetic structure is an elliptical helix to good approximation, while we find possible evidence for a tilted helix (helicoid) in the **k**₁ domain. This tilt may be unique to this domain, or could exist more broadly in this family of materials but lie hidden due to averaging

over subdomains. The presence of strong circular dichroism indicated that a predominantly single chiral domain was observed within the x-ray-beam footprint, which persisted at all temperatures below T_N . A complete characterization of the domain structure is left for future experiments, which may also establish whether chiral symmetry is broken in this material above T_N , and hence further elucidate the nature of magneto-structural coupling apparent in this system. Finally, we note that understanding the microscopic mechanisms by which 10% Ga substitution leads to such dramatic changes in the phase diagram is worthy of further study.

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DATA AVAILABILITY

Data that support the findings of this paper are available on reasonable request from the corresponding authors.

APPENDIX A: AZIMUTHAL SCANS

In this appendix, we derive the equations describing the azimuthal dependence on the magnetic scattering intensity [including Eqs. (1) and (2)], following the method outlined in Ref. [24].

1. Laboratory frame of reference

We define a Cartesian "laboratory" frame of reference, $\{x, y, z\}$, such that $\hat{z}$ is normal to the scattering plane

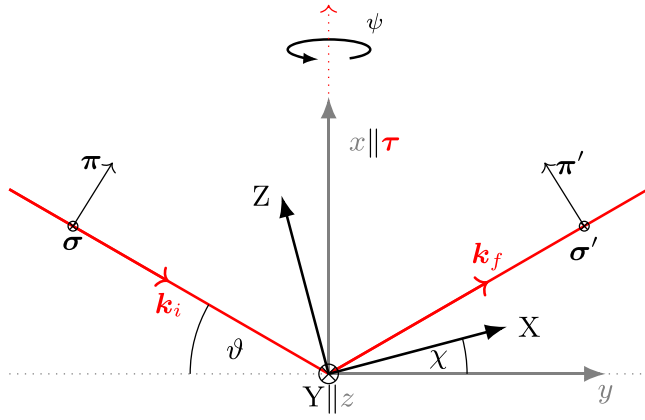


FIG. 7. Scattering geometry used in the azimuthal calculations. $\{X, Y, Z\}$ form a real space Cartesian coordinate system (black); the *crystal frame*. By definition, the axes $\{X, Y, Z\}$ are parallel to the crystallographic $\mathbf{a}, \mathbf{b}, \mathbf{c}$ directions in domain 1, respectively. In domain 2, $\{X, Y, Z\}$ are parallel to $\mathbf{b}, -\mathbf{a}, \mathbf{c}$, respectively. $\{x, y, z\}$ define the *laboratory frame*, which is also a real-space coordinate system (gray). Red lines represent the direction of incoming ($\mathbf{k}_i$) and scattered ($\mathbf{k}_f$) x-rays. Unprimed and primed σ and π define the respective polarization directions. The angle χ is computed using Eq. (A1). We define our coordinate system such that in scattering condition for the $(\delta, 0, 8)$ satellite, $\hat{\mathbf{x}}$ coincides with $\boldsymbol{\tau}$, the scattering vector. Rotations are defined to be right-handed. Rotations about $\boldsymbol{\tau}$ are described by the azimuthal angle ψ .

containing the incident and scattered x-ray wave vectors $\mathbf{k}_i$ and $\mathbf{k}_f$. The $\{x, y, 0\}$ plane rotates about $\hat{\mathbf{z}}$ such that $\hat{\mathbf{x}}$ is always bisecting $\mathbf{k}_i$ and $\mathbf{k}_f$, and hence always parallel to the scattering vector $\boldsymbol{\tau} = \mathbf{k}_f - \mathbf{k}_i$. Rotations are taken to be right-handed. The polarization of the incident and scattered light is labeled in the usual way, with σ perpendicular to the scattering plane and π lies within it.

2. Crystal frame of reference

We define a Cartesian ‘crystal’ frame of reference $\{X, Y, Z\}$. For domain 1, the Cartesian basis vectors are parallel to the tetragonal $\mathbf{a}, \mathbf{b}$, and $\mathbf{c}$ axes; for domain 2, the Cartesian basis vectors are parallel to $\mathbf{b}, -\mathbf{a}$, and $\mathbf{c}$, respectively. First, we calculate the azimuthal dependence of the $\boldsymbol{\tau} = (\pm\delta, 0, 8)$ magnetic reflections. The geometry in which the $+\delta$ reflection is in the scattering condition at an azimuthal angle of $\psi = 0$ is shown in Fig. 7. Here, $\hat{\mathbf{Y}} \parallel \hat{\mathbf{z}}$, $\hat{\mathbf{Z}}$ is close to $\hat{\mathbf{x}}$, and $\hat{\mathbf{X}}$ is close to $\hat{\mathbf{y}}$. We note that these latter directions are parallel to each other for $\boldsymbol{\tau} = (0, 0, 8)$, but the small incommensurate component $\pm\delta$ requires a rotation of $\mp\chi$ about $\hat{\mathbf{z}}$, where

$$\cos(\chi) = \frac{\boldsymbol{\tau} \cdot \mathbf{c}}{|\boldsymbol{\tau}|c} = \frac{1}{\sqrt{1 + \left(\frac{\delta c}{8a}\right)^2}}. \quad (\text{A1})$$

3. Unit-cell structure factor

Throughout this derivation we will adopt the conventions of Ref. [25]. Magnetic moment components are described as the expectation value of their respective polarization operator $\hat{T}_Q^{(K)}$, which is a tensor operator of rank K defined in the

spherical basis. Here, K and Q label, respectively, the total angular momentum and its projection onto the quantization axis. In the following calculations we consider scattering from dipoles only, i.e., $K = 1$. For clarity, we therefore drop the K index in the following and one should understand T to be a tensor of rank 1.

First, we define a covariant spherical tensor unit-cell structure factor [24]:

$$\mathcal{F}_q = \sum_j e^{i\boldsymbol{\tau} \cdot \mathbf{r}_j} \langle T_q \rangle^j, \quad (\text{A2})$$

where $\langle T_q \rangle$ is the expectation value of the covariant spherical tensor T_q associated with the j th atom located at position $\mathbf{r}_j$. The spherical basis of the tensors will be defined with respect to coordinates $\{Z, X, Y\}$ such that at $\psi = 0$ the tensor basis differs from the laboratory basis simply by rotation $\mp\chi$. Thus, in the relabeled basis, we have (see Eq. (11) in Sec. 1.2 in Ref. [25])

$$\begin{aligned} T_X^1 &= \frac{i}{\sqrt{2}}(T_{-1} + T_1) \\ T_Y^1 &= T_0 \\ T_Z^1 &= \frac{1}{\sqrt{2}}(T_{-1} - T_1). \end{aligned} \quad (\text{A3})$$

In our material we have just two rare-earth ions Eu^{2+} in the conventional I -centred unit cell, and a single rare-earth ion in the primitive unit cell. Given that the magnetic structure preserves I centering (the spin-density wave on sites related by translation $[1/2, 1/2, 1/2]$ differs by a phase of $\pi\delta$), it is sufficient to consider only the atom in the primitive unit cell at the origin. Hence,

$$\mathcal{F}_q = \langle T_q \rangle. \quad (\text{A4})$$

4. Rotations

Next, we perform two rotations of the unit-cell structure factor. These rotations will be performed in terms of the appropriate Wigner D matrices $D_{m'm}^{(K)}(\alpha, \beta, \gamma)$ for $K = 1$, with Euler angles α, β, γ in the extrinsic z - y - z convention. First, the crystal is rotated by $-\chi$ about $\hat{\mathbf{z}}$, and then by the azimuthal angle ψ about $\hat{\mathbf{x}}$. Alternatively, in the passive picture of the Euler angles, the laboratory frame is rotated about $\hat{\mathbf{z}}$ by χ , then about $\hat{\mathbf{x}}$ by $-\psi$. We write the rotated structure factor, now defined in the laboratory frame of reference, as

$$\tilde{\mathcal{F}}_Q = \sum_q \sum_{q'} \langle T_q \rangle D_{qq'}(\chi) D_{q'Q}(\psi). \quad (\text{A5})$$

A rotation by χ about $\hat{\mathbf{z}}$ is defined as

$$D_{qq'}(\chi) = e^{-iq'\chi} \delta_{qq'}. \quad (\text{A6})$$

The azimuthal rotation is more complex. The rotation about $\hat{\mathbf{x}}$ in the extrinsic Euler z - y - z convention can be achieved by (1) rotation by $-\frac{\pi}{2}$ about $\hat{\mathbf{z}}$, then (2) rotation by $-\psi$ about $\hat{\mathbf{y}}$, then (3) rotation by $\frac{\pi}{2}$ about $\hat{\mathbf{z}}$. In this case

$$D_{q'Q}(-\psi) = e^{-i\frac{\pi}{2}q'} d_{q'Q}(-\psi) e^{i\frac{\pi}{2}Q}, \quad (\text{A7})$$

where $d_{q'Q}(-\psi)$ is the Wigner small d-matrix. (A7) can be evaluated, giving

$$\begin{pmatrix} \frac{1}{2}(1 + \cos(\psi)) & \frac{-i}{\sqrt{2}}\sin(\psi) & -\frac{1}{2}(1 - \cos(\psi)) \\ \frac{-i}{\sqrt{2}}\sin(\psi) & \cos(\psi) & \frac{-i}{\sqrt{2}}\sin(\psi) \\ -\frac{1}{2}(1 - \cos(\psi)) & \frac{-i}{\sqrt{2}}\sin(\psi) & \frac{1}{2}(1 + \cos(\psi)) \end{pmatrix}. \quad (\text{A8})$$

5. X-ray polarization

Finally, to calculate the scattering amplitudes $F(\varepsilon, \varepsilon')$, we take the dot product with contravariant spherical tensor $J^Q(\hat{\varepsilon}, \hat{\varepsilon}')$ that depends upon the x-ray polarization unit vectors of both the incident ($\hat{\varepsilon}$) and scattered ($\hat{\varepsilon}'$) beams, defined in a spherical basis with respect to the laboratory frame of reference:

$$\begin{aligned} F(\varepsilon, \varepsilon') &= \sum_Q J^Q(\hat{\varepsilon}, \hat{\varepsilon}') \tilde{F}_Q \\ &= \sum_{qq'Q} J^Q(\hat{\varepsilon}, \hat{\varepsilon}') \langle T_q \rangle D_{qq'}(\chi) D_{q'Q}(-\psi). \end{aligned} \quad (\text{A9})$$

For E1-E1 scattering, one can show that (see p. 256 of Ref. [24])

$$\begin{aligned} J_1^1(\hat{\varepsilon}, \hat{\varepsilon}') &= \frac{1}{\sqrt{2}}(\varepsilon'_{-1}\varepsilon_0 - \varepsilon'_0\varepsilon_{-1}) \\ J_1^0(\hat{\varepsilon}, \hat{\varepsilon}') &= \frac{1}{\sqrt{2}}(\varepsilon'_1\varepsilon_{-1} - \varepsilon'_{-1}\varepsilon_1) \\ J_1^{-1}(\hat{\varepsilon}, \hat{\varepsilon}') &= \frac{1}{\sqrt{2}}(\varepsilon'_0\varepsilon_1 - \varepsilon'_1\varepsilon_0), \end{aligned} \quad (\text{A10})$$

where our $\hat{J}$ tensor relates to the original $X_Q^{(1)}$ defined in Ref. [24], p. 256, by $J_1^Q = (-1)^Q X_{-Q}^{(1)}$. With reference to the geometry of Fig. 7, we evaluate the vectors $\hat{\varepsilon}$ and $\hat{\varepsilon}'$ for the $\sigma(\sigma')$ and $\pi(\pi')$ polarization directions:

$$\begin{aligned} \hat{\varepsilon}_\sigma &= [0, 1, 0] \\ \hat{\varepsilon}'_\sigma &= [0, 1, 0] \\ \hat{\varepsilon}_\pi &= \frac{1}{\sqrt{2}}[-e^{i\vartheta}, 0, e^{-i\vartheta}] \\ \hat{\varepsilon}'_\pi &= \frac{1}{\sqrt{2}}[-e^{-i\vartheta}, 0, e^{i\vartheta}], \end{aligned} \quad (\text{A11})$$

where ϑ is half the Bragg angle. Hence,

$$\begin{aligned} J^1(\sigma, \sigma') &= J^0(\sigma, \sigma') = J^{-1}(\sigma, \sigma') = 0 \\ J^1(\sigma, \pi') &= -J^{-1}(\pi, \sigma') = \frac{1}{2}e^{i\vartheta} \\ J^{-1}(\sigma, \pi') &= -J^1(\pi, \sigma') = \frac{1}{2}e^{-i\vartheta} \\ J^0(\sigma, \pi') &= J^0(\pi, \sigma') = 0 \\ J^0(\pi, \pi') &= \frac{i}{\sqrt{2}}\sin(2\vartheta) \\ J^1(\pi, \pi') &= J^{-1}(\pi, \pi') = 0. \end{aligned} \quad (\text{A12})$$

6. Scattering amplitudes

We are now in a position to calculate the scattering amplitudes $F(\varepsilon, \varepsilon')$ from dipoles in each polarization channel for the $(\pm\delta, 0, 8)$ satellites:

$$F(\sigma, \sigma') = 0, \quad (\text{A13})$$

$$\begin{aligned} F(\sigma, \pi') &= \frac{1}{2}(e^{i\vartheta}\tilde{F}_1 + e^{-i\vartheta}\tilde{F}_{-1}) \\ &= -\frac{i}{\sqrt{2}}(\cos(\vartheta)\cos(\psi)\Omega^c + \sin(\vartheta)\Omega^s \\ &\quad - \cos(\vartheta)\sin(\psi)\Omega^0), \end{aligned} \quad (\text{A14})$$

$$\begin{aligned} F(\pi, \sigma') &= \frac{1}{2}(e^{-i\vartheta}\tilde{F}^1 + e^{i\vartheta}\tilde{F}^{-1}) \\ &= \frac{i}{\sqrt{2}}(\cos(\vartheta)\cos(\psi)\Omega^c - \sin(\vartheta)\Omega^s \\ &\quad - \cos(\vartheta)\sin(\psi)\Omega^0), \end{aligned} \quad (\text{A15})$$

$$\begin{aligned} F(\pi, \pi') &= \frac{i}{\sqrt{2}}\sin(2\vartheta)\tilde{F}^0 \\ &= \frac{i}{\sqrt{2}}\sin(2\vartheta)(\sin(\psi)\Omega^c + \cos(\psi)\Omega^0), \end{aligned} \quad (\text{A16})$$

where

$$\begin{aligned} \Omega^c &= \mp \sin(\chi)\langle T_Z \rangle + \cos(\chi)\langle T_X \rangle \\ \Omega^0 &= \langle T_Y \rangle \\ \Omega^s &= \cos(\chi)\langle T_Z \rangle \pm \sin(\chi)\langle T_X \rangle \end{aligned} \quad (\text{A17})$$

when written in terms of dipole tensors in the $\{X, Y, Z\}$ basis.

7. Scattering intensities

For a general incident photon polarization, $\hat{\varepsilon} = \alpha\hat{\varepsilon}_\pi + \beta\hat{\varepsilon}_\sigma$, the intensity measured by a detector that is insensitive to the polarization of the scattered beam is written

$$I = |\alpha F(\pi, \pi') + \beta F(\sigma, \pi')|^2 + |\alpha F(\pi, \sigma') + \beta F(\sigma, \sigma')|^2. \quad (\text{A18})$$

For σ polarized incident light $\alpha = 0$ and $\beta = 1$, and given that $F(\sigma, \sigma') = 0$, the relevant diffraction intensity is

$$I_\sigma = |F(\sigma, \pi')|^2. \quad (\text{A19})$$

By evaluating Eq. (A19) for $(-\delta, 0, 8)$, one obtains Eq. (1). Then, to calculate the intensity for $(0, \delta, 8)$, one adds a $\frac{\pi}{2}$ offset to ψ in Eq. (1), giving Eq. (2).

For π polarized incident light $\alpha = 1$ and $\beta = 0$, and the measured intensity is

$$I_\pi = |F(\pi, \pi')|^2 + |F(\pi, \sigma')|^2. \quad (\text{A20})$$

For circularly polarized incident light of different handedness (which we label $+$ and $-$), $\alpha = \frac{1}{\sqrt{2}}$ and $\beta = \pm \frac{i}{\sqrt{2}}$. The total intensity $I_\pm$ of the two channels is then

$$I_\pm = \frac{1}{2}(I_\sigma + I_\pi \mp I_C), \quad (\text{A21})$$

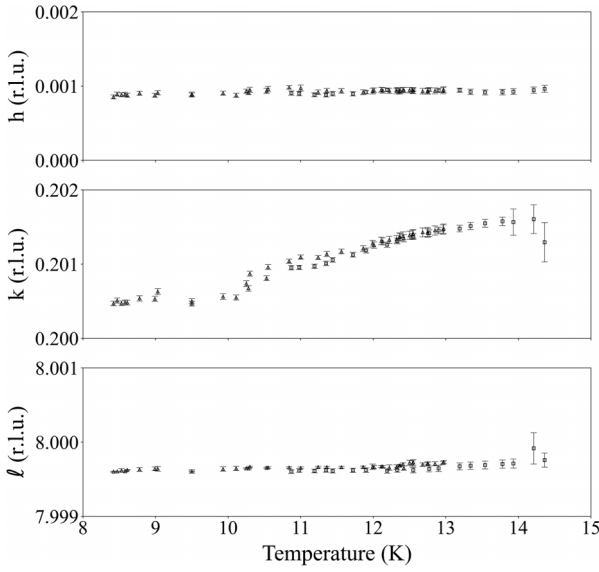


FIG. 8. Temperature dependence of the center of the (0, 0.2, 8) magnetic satellite.

where

$$I_C = \text{Im}\{F(\pi, \pi')F(\sigma, \pi')^*\}. \quad (\text{A22})$$

One can show that for a collinear SDW, $I_C = 0$, while for a magnetic helix $I_C \neq 0$, and its sign depends upon the chirality of the helix.

The ratio of $I_{\pm}$ to I_{σ} is given by

$$\frac{I_{\pm}}{I_{\sigma}} = \frac{1}{2} + \frac{I_{\pi} \mp I_C}{2I_{\sigma}}. \quad (\text{A23})$$

Assuming the incident beam always illuminates a single helical domain, a phase transition at which the chirality of the magnetic helix spontaneously reverses can be observed as a step change in the above ratio of magnitude I_C/I_{σ} . In general, a step change may also be observed for a transition from a collinear SDW to a chiral helix, which leads to a change in I_{σ} , I_{π} , and I_C .

APPENDIX B: TEMPERATURE DEPENDENCE OF SATELLITE CENTER

The temperature dependence of the (0, 0.2, 8) magnetic peak center is shown in Fig. 8. The position in h and ℓ remained constant on warming, while the peak center varies monotonically in k . This variation is consistent with an incommensurate magnetic wave vector.

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