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Lattice-commensurate skyrmion texture in a centrosymmetric breathing kagome magnet

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Max Hirschberger^{1,2}✉, Bertalan G. Szigeti³, Mamoun Hemmida³, Moritz M. Hirschmann², Sebastian Esser¹, Hiroyuki Ohsumi⁴, Yoshikazu Tanaka⁴, Leonie Spitz^{2,11}, Shang Gao^{2,12}, Kamil K. Kolincio^{2,5}, Hajime Sagayama⁶, Hironori Nakao⁶, Yuichi Yamasaki⁷, László Forró⁸, Hans-Albrecht Krug von Nidda³, Istvan Kezsmarki³, Taka-hisa Arima^{2,9} & Yoshinori Tokura^{1,2,10}

Skyrmion lattices (SkL) in centrosymmetric materials typically have a magnetic period on the nanometer-scale, so that the coupling between magnetic superstructures and the underlying crystal lattice cannot be neglected. We reveal the commensurate locking of a SkL to the atomic lattice in $\text{Gd}_3\text{Ru}_4\text{Al}_{12}$ via high-resolution resonant elastic x-ray scattering (REXS). Weak easy-plane magnetic anisotropy, demonstrated here by a combination of ferromagnetic resonance and REXS, penalizes placing a skyrmion core on a site of the atomic lattice. Under these conditions, a commensurate SkL, locked to the crystal lattice, is stable at finite temperatures – but gives way to a competing incommensurate ground state upon cooling. We discuss the role of Umklapp-terms in the Hamiltonian for the formation of this lattice-locked state, its magnetic space group, and the role of slight discommensurations, or (line) defects in the magnetic texture. We also contrast our findings with the case of SkLs in noncentrosymmetric material platforms.

Magnetic skyrmion lattices (SkLs) are periodic arrays of vortex-like spin structures. In SkLs, magnetic moments are twisted into a knot, covering all directions of a sphere as we traverse a single magnetic unit cell (UC) (Fig. 1a)^{1–3}. These vortices were first described as topological solitons using the concepts of field theory, and such continuum models are most suitable when the magnetic UC is at least two orders of magnitude larger than the underlying crystallographic UC^{3–5}. With a focus on frustrated, i.e. competing, interactions, recent theoretical work^{6–10} has proposed SkL formation in a high-symmetry context without spin-orbit driven Dzyaloshinskii-Moriya interactions, paving the way for the experimental observation of SkL phases with magnetic period on the nanometer-scale in centrosymmetric insulators and metals^{3,11–14}. These quasi-discrete SkLs have raised hopes of enhanced functional responses, especially those related to the interplay of

emergent electromagnetic fields with conduction electron (Bloch) waves, or with incident light waves^{3,15–20}.

Evidence for coupling between the atomic lattice and skyrmion textures with lattice spacing 2 – 3 nanometers has emerged in tetragonal magnets: Centrosymmetric alloys host square and rhombic skyrmion lattices^{3,21}, and non-centrosymmetric EuNiGe_3 exhibits a fascinating helicity reversal upon entering the SkL phase, where the magnetic texture breaks the sense of rotation prescribed by its polar structure^{13,14}. A key open challenge is the demonstration of a commensurate locking (C-locking) transition of the SkL's spin superstructure to the underlying lattice potential in such a centrosymmetric bulk material. This phenomenon is conceptually related to instabilities of the skyrmion vortex core anticipated for spin-1 systems²² and its observation would provide a bridge between – usually – large-scale SkL

¹Department of Applied Physics and Quantum-Phase Electronics Center, The University of Tokyo, Bunkyo-ku, Tokyo 113-8656, Japan. ²RIKEN Center for Emergent Matter Science (CEMS), Wako, Saitama 351-0198, Japan. ³Experimental Physics V, Center for Electronic Correlations and Magnetism, University of Augsburg, 86135 Augsburg, Germany. ⁴RIKEN SPring-8 Center, 1-1-1 Kouto, Sayo, Hyogo 679-5148, Japan. ⁵Faculty of Applied Physics and Mathematics, Gdańsk University of Technology, Narutowicza 11/12, 80-233 Gdańsk, Poland. ⁶Institute of Materials Structure Science, High Energy Accelerator Research Organization, Tsukuba, Ibaraki 305-0801, Japan. ⁷Research and Services Division of Materials Data and Integrated System (MaDIS), National Institute for Materials Science (NIMS), Tsukuba 305-0047, Japan. ⁸Stavropoulos Center for Complex Quantum Matter, Department of Physics and Astronomy, University of Notre Dame, Notre Dame, IN 46556, USA. ⁹Department of Advanced Materials Science, The University of Tokyo, Kashiwa 277-8561, Japan. ¹⁰Tokyo College, The University of Tokyo, Bunkyo-ku, Tokyo 113-8656, Japan. ¹¹Present address: Paul Scherrer Institut, Forschungsstrasse 111, 5232 Villigen PSI, Switzerland, ¹²Present address: Department of Physics, University of Science and Technology of China, Hefei 230026, China. ✉e-mail: hirschberger@ap.t.u-tokyo.ac.jp

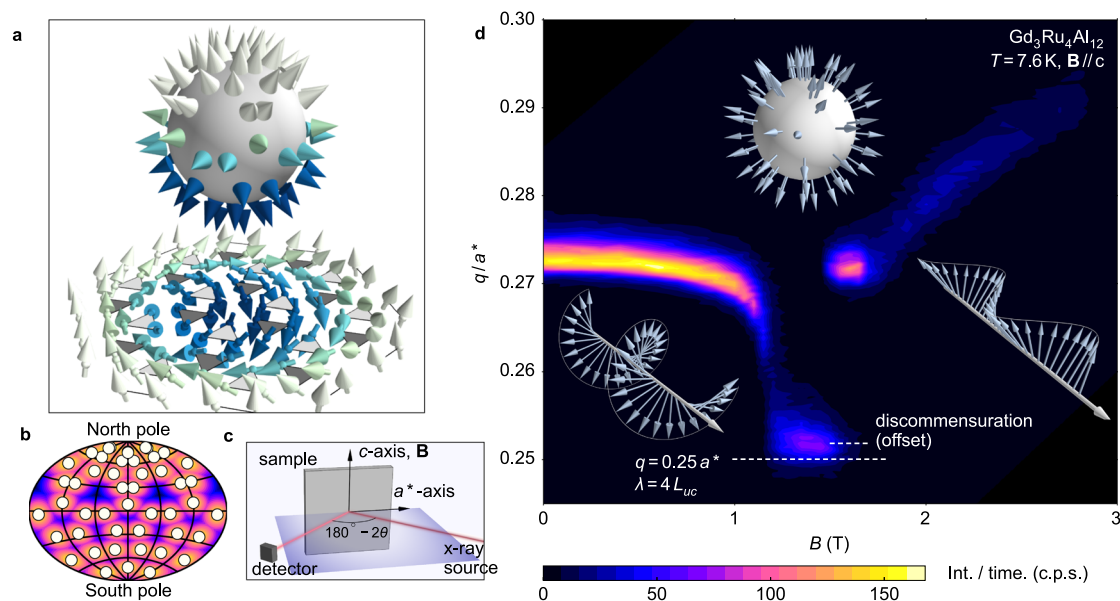


Fig. 1 | Commensurate locking of the skyrmion lattice phase in $Gd_3Ru_4Al_{12}$ (GRA). **a** Magnetic moments of a skyrmion winding a sphere (top), and corresponding two-dimensional magnetic moment texture in real space (bottom). Each arrow corresponds to a site of the magnetic sublattice (Supplementary Note 1). **b** Map projection showing a hemisphere of (a) unfolded, with white dots indicating directions of magnetic moments. The colour code indicates the distance of each point on the sphere from the nearest moment on the sphere, in radians. **c** Experimental geometry of resonant elastic x-ray scattering (REXS) at the Gd- L_2 absorption edge, where the pink line is the trajectory of the x-ray beam. **d** Scattering

intensity in REXS as a function of magnetic field (B) and momentum transfer q from $\mathbf{q} = (q, 0, 0)$; $q = 0.25$ corresponds to $\lambda = 2\pi/q = 4 L_{uc}$, four times the projection of the lattice constant parallel to \mathbf{q} (Supplementary Fig. 1). Incommensurate proper screw (IC-PS), commensurate skyrmion lattice (C-SkL), and IC fan phases are illustrated by insets. In C-SkL, the periodicity of the magnetic texture is locked to the crystal lattice up to a weak discommensuration (offset) of $\Delta q = 0.0018$ r.l.u. B denotes magnetic induction after demagnetization correction (Supplementary Fig. 4). Supplementary Fig. 6 and Supplementary Fig. 7 show raw data used to create this colormap and the results of Gaussian fitting to the data, respectively.

spin textures in materials with broken inversion symmetry, and canted antiferromagnetism on the scale of a single unit cell. Indeed, theory predicts such C-locking based on Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions and magnetic anisotropy, when the length scale of magnetic textures approaches the size of a crystallographic UC²³. Among inversion breaking material platforms, the hcp-Fe/Ir(111) interface has been reported to exhibit C-locking using imaging techniques, although fcc-Fe/Ir(111) forms a lattice-incommensurate structure^{6,24–26}. However, such locking between the periodicity of a magnetic skyrmion lattice and the underlying crystal structure has never been observed in a bulk material.

Using precise resonant x-ray measurements, we report a commensurate skyrmion lattice (C-SkL) surrounded by incommensurate (IC) phases in bulk samples of the centrosymmetric intermetallic $Gd_3Ru_4Al_{12}$, locked to the distorted kagome network of magnetic gadolinium ions. We discuss this state based on (weak) single-ion anisotropy K_1 , as supported by electron-spin resonance experiments.

For large classical spins, calculations on both triangular and breathing kagome lattices show that, if the single-ion anisotropy is of easy-plane type (easy-axis type), a commensurate skyrmion vortex may gain energy by locating its core at an interstitial site (on a lattice site)^{23,27}; an incommensurate skyrmion lattice does not benefit from this type of energy gain (Supplementary Fig. 5). We illustrate this point in the lower part of Fig. 1a, depicting a realistic magnetic structure model for the C-SkL of $Gd_3Ru_4Al_{12}$, described by normalized vectors $\mathbf{n}(x, y)$, which is mapped onto a sphere using a stereographic projection, see Methods. Here, magnetic moments are conspicuously absent at the poles (Fig. 1a, upper). The sparsity of magnetic moments at the poles becomes more apparent when unfolding the sphere using a cartographic projection (Fig. 1b and Supplementary Note 1).

Results

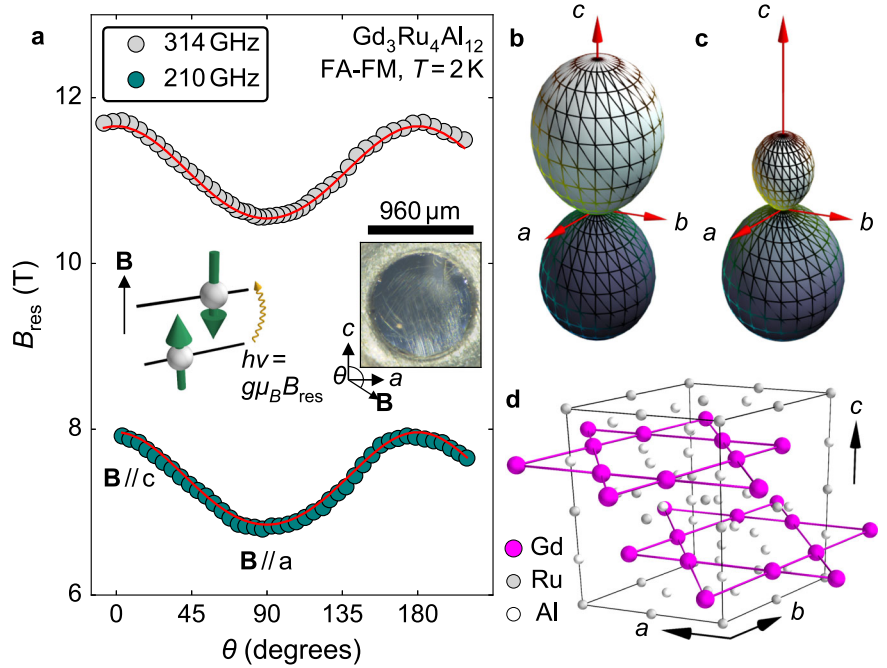
Observation of a commensurate skyrmion lattice (C-SkL)

We use elastic x-ray scattering in resonance with the $L_{2,3}$ absorption edge of gadolinium, setting the sample in reflection geometry to

precisely detect the magnetic period of each magnetic phase (Fig. 1c and Methods). Reporting data from a synchrotron light source, Fig. 1d depicts the core observation of this work: At moderate temperatures $T = 7 - 8$ K, a magnetic field \mathbf{B} applied along the c -axis drives the incommensurate proper screw ground state (IC-PS) into a commensurate skyrmion lattice phase (C-SkL), and again to incommensurate fan-like order (IC-Fan). The C-locking of the SkL was not observed in a previous study¹¹. The present and previous results are compared and summarized in Supplementary Fig. 4. Let a^* and c^* be the reciprocal space lattice constants for our target material $Gd_3Ru_4Al_{12}$ in the hexagonal $P6_3/mmc$ space group, where magnetic gadolinium ions form a kagome (star of David) lattice with a breathing distortion, corresponding to alternating bond distances. In zero magnetic field, the magnetic modulation wavevector is $\mathbf{q} = (q, 0, 0)$ with $q \approx 0.272 a^*$ or wavelength $\lambda \approx 3.7 L_{uc}$, where L_{uc} is the dimension of the crystallographic UC projected parallel to \mathbf{q} (Supplementary Fig. 1). The wavevector's length q decreases with B in IC-PS, approaching a discontinuous jump at the first-order transition¹¹ towards $q = 0.25 a^*$ in C-SkL. The magnetic period λ in C-SkL is very close to $4 L_{uc}$, with a slight offset indicated by two dashed horizontal lines in Fig. 1d. The role of this slight offset, or discommensuration λ_{disc} , is discussed below. To further support the C-locking at $q = 0.25 a^*$, we demonstrate in Supplementary Fig. 7 that the peak profiles are within less than one standard deviation from the commensurate value, and in Supplementary Fig. 10 that q has weak temperature dependence in C-SkL as compared to IC-PS.

In the regime labeled as C-SkL in Fig. 1, previous real-space imaging experiments have observed vortex structures¹¹, and precise Hall effect measurements demonstrate that the noncoplanar magnetic state that is easily destroyed by a slight in-plane magnetic field²⁸. However, neither this prior work nor the present REXS technique are able to determine whether the skyrmion core is located on a crystallographic lattice site, or on an interstitial site. This question of the relative phase shift between magnetic

Fig. 2 | Magnetic anisotropy in field-polarized ferromagnetic (FA-FM), incommensurate proper screw (IC-PS), and skyrmion lattice (C-SkL) phases of $Gd_3Ru_4Al_{12}$. **a** Resonance field $B_{res} = \mu_0 H_{res}$ in ferromagnetic resonance (FMR) experiments in FA-FM at 210 and 314 GHz (grey and green markers). Red lines: Fit to the data according to Smit-Beliers-Suhl, corresponding to weak in-plane anisotropy, see Methods. Left inset: principle of FMR, where transitions between up and down moments are induced by microwave radiation in a cavity. Right inset: geometry of magnetic field \mathbf{B} with respect to the principal axes of a disk-shaped single crystal. **b, c** Anisotropic energy landscape $F_{anis} = K_1 \cos^2(\theta) - E_Z \cos(\theta)$ in zero field (Zeeman term $E_Z = 0$) and finite magnetic field ($E_Z/K_1 = 0.3$), as a function of the direction of the bulk magnetization vector \mathbf{M} . **d** Crystal structure of $Gd_3Ru_4Al_{12}$ with magnetic breathing kagome sublattice of gadolinium, shown in magenta.



texture and crystal lattice can be addressed by measurements of single-ion anisotropy, combined with theoretical modeling: We turn to the ferromagnetic resonance (FMR) technique in the following section.

Single-ion anisotropy in $Gd_3Ru_4Al_{12}$

We prepared a cylindrical disk-shaped sample for ferromagnetic resonance (FMR) experiments, spanned by the crystallographic a and c directions (Fig. 2a, right inset). This highly symmetric geometry allows for simple data analysis when rotating the magnetic field in the plane of the disk, see Methods. In the experiment, we drive the crystal into the field-aligned ferromagnetic (FA-FM) state with a large magnetic field, irradiate it with microwaves of frequency $\nu = 210$ or 314 GHz, and observe a change of its reflectivity when the microwaves excite a resonance between moment-up and -down states (Fig. 2a, left inset).

In Fig. 2a, the anisotropic part of the free energy $F_{anis} = K_1 \cos^2(\theta)$ is deduced in FA-FM from a fit to the angular dependence of the FMR resonance field B_{res} ; we disregard anisotropy constants beyond the first order, see Methods. Using the saturation magnetization $M_S = 7 \mu_B / Gd^{3+}$, our fit yields easy-plane anisotropy with $K_1 = -0.13$ meV / Gd^{3+} . Therefore, the anisotropy field B_{ani} can be calculated as $\mu_0 |2K_1| / M_S = 0.74$ Tesla. As compared to magnetization measurements in Supplementary Fig. 11, the present FMR study yields enhanced precision; field-dependent FMR experiments separate g -factor anisotropy and exchange anisotropy from the single-ion term, see Methods. Figure 2b, c illustrates the resulting iso-energy surfaces $F_{anis}(\theta, \varphi)$ in zero (finite) magnetic field along the c -axis, respectively, where the spherical coordinates refer to the direction of the sample's bulk magnetization \mathbf{M} .

Anisotropy and anharmonic distortion of proper screw and skyrmion phases

Easy-plane anisotropy ($K_1 < 0$) can also be verified semi-quantitatively by resonant elastic x-ray scattering in the ordered phases with periodic long-range order. Figure 1c shows the geometry of our experiment with polarization analysis: the purple scattering plane is spanned by the wavevectors \mathbf{k}_i and \mathbf{k}_f of the incoming and outgoing x-ray beams, with beam polarization ε_i and ε_f , respectively. We choose the incoming beam polarization ε_i to lie within the scattering plane for all our experiments (π -polarization). While the data in Fig. 1d represents a sum of scattered

photons with all possible ε_f , we now add an analyser plate before the x-ray detector to separate two components of the scattered beam: $I_{\pi-\pi'}$ and $I_{\pi-\sigma'}$ with ε_f within or perpendicular to the scattering plane, respectively. From the scattered intensities at various magnetic reflections, we extract the ratio, see Methods,

$$R \sin^2(2\theta) = I_{\pi-\sigma'} \sin^2(2\theta) / I_{\pi-\pi'} = [\mathbf{k}_i \cdot \mathbf{m}_{ab}(\mathbf{q}) / m_c(\mathbf{q})]^2 \quad (1)$$

with \mathbf{m}_{ab} the component of the modulated magnetic moment in the scattering plane. The expected behavior of IC-PS (spin plane a - c) and IC cycloid (spin plane a^* - c) is indicated in Fig. 3b by black and green dashed lines, respectively; a line with positive slope indicates IC-PS character. Supplementary Fig. 3 shows representative raw data, as used to create this panel.

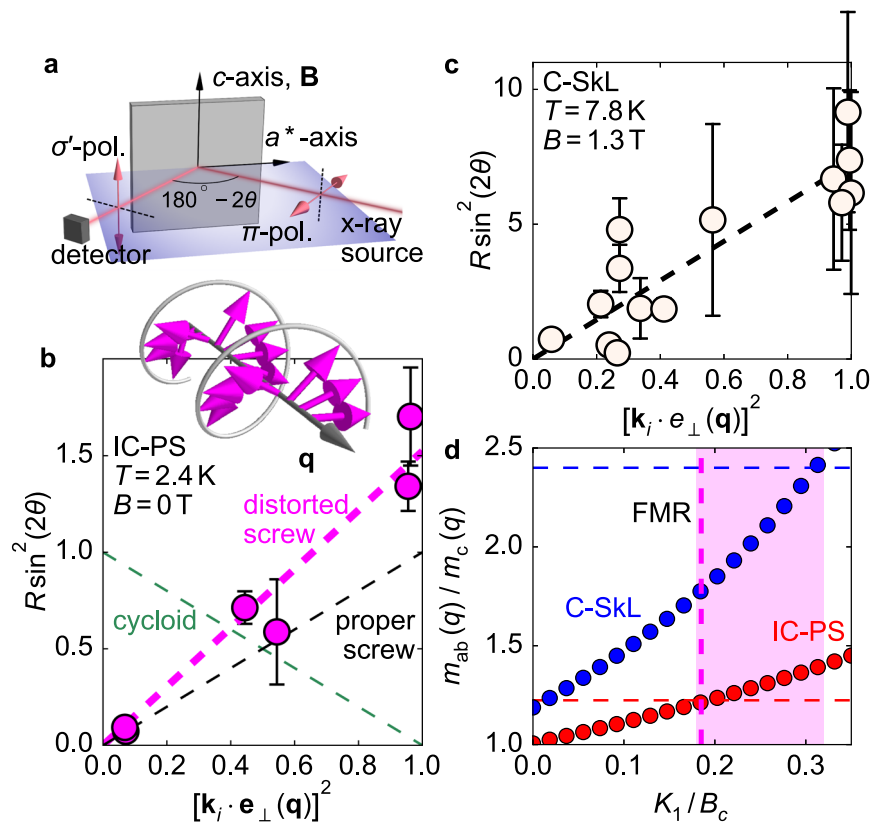
Beyond identifying the PS character of the spin modulation, this analysis allows us to estimate the effect of single-ion anisotropy in IC-PS. Specifically, we observe an elliptic distortion, i.e., a deviation from the proper screw model. Figure 3b, c displays maximum values of $R \sin^2(2\theta)$ around 1.5 (IC-PS) and 7 (C-SkL), so that Eq. (1) delivers $m_{ab}(\mathbf{q}) / m_c(\mathbf{q})$ around 1.2 (IC-PS) and 2.6 (C-SkL). Figure 3b, inset, illustrates the proposed anharmonic distortion in IC-PS, where magnetic moments prefer to tilt towards the basal plane to gain anisotropy energy. In Fig. 3d, a simulation of REXS anisotropy as a function of K_1 for a spin model in both IC-PS and C-SkL (Supplementary Note 1) shows that the experimentally observed ellipticity is consistent with the results of FMR in Fig. 2. Given the robust observation of $K_1 < 0$ via various experimental techniques, we conclude that a C-SkL in magnetic space group $P6_32'2'$ and with skyrmion core between lattice sites is stable in $Gd_3Ru_4Al_{12}$ (Supplementary Note 2).

Competition of commensurate and incommensurate phases

Figure 4a shows a magnetic field scan of x-ray scattering intensity at the lowest accessible temperature, $T = 1.5$ K; see Supplementary Fig. 4 for detailed phase diagrams and comparison to prior work. In contrast to Fig. 1d, the C-SkL phase is now absent, being replaced by an incommensurate transverse conical (IC-TC) state¹¹.

We compare the experimental and theoretical results for $Gd_3Ru_4Al_{12}$ to earlier studies of magnetism in elemental rare earth metals. Various scenarios for the interplay of commensurate (C) and IC phases have been advanced^{29,30}: One typical observation in systems with strong single-ion

Fig. 3 | Magnetic anisotropy in ordered states of $Gd_3Ru_4Al_{12}$. **a** Experimental geometry of resonant elastic x-ray scattering (REXS) with (in contrast to Fig. 1c) polarization analysis. π and σ indicate x-rays (photons) polarized parallel and perpendicular to the scattering plane (purple), respectively. **b** Elliptic distortion of IC-PS determined by polarization analysis in REXS. $I_{\pi-\sigma}$, $I_{\pi-\pi}$, \mathbf{k}_i , and \mathbf{e}_\perp are integrated intensities in the polarization-flipping and polarization-conserving scattering channels, the incoming beam's wavevector, and the in-plane direction perpendicular to the propagation vector \mathbf{q} of IC-PS, as in Eq. (1). Green, black, and magenta dashed lines indicate model predictions for spherical cycloid, spherical proper screw, and elliptically distorted screw, the latter being shown in the inset cartoon. The value of $R\sin^2(2\theta)$ at the right boundary of the panel corresponds to the elliptic distortion $(m_{ab}/m_c)^2$ as defined in Eq. (1). **c** Analogous data for the commensurate skyrmion (C-SkL) state; see Supplementary Table 1 for more details. Note the different y -axis range as compared to (b). **d** Spin-model calculation of elliptic distortion in IC-PS and C-SkL as a function of anisotropy constant K_1 , normalized to the critical field $B_c = 4$ T for transition to the field-aligned ferromagnetic (FA-FM) state. Dashed horizontal lines, dashed vertical line, and highlighted area indicate the experimental values for the ellipticity, the value of K_1 from FMR, and the range of K_1 consistent with REXS experiments, respectively. Error bars in (b, c) are obtained from Gaussian fits to line profiles as in Supplementary Fig. 3, with quadratic error propagation for the intensity ratio.



anisotropy is the appearance of spin-slip structures in the IC order, and eventual *squaring up* at low temperatures. In this scenario, thermal fluctuations at higher T locally reduce the ordered magnetic moment, allowing for the formation of IC magnetism close to the Néel point, e.g. in elemental Holmium²⁹. Secondly, when single-ion anisotropy is much weaker than exchange interactions, C orders are favored at higher temperatures where short-range correlations dominate. However, they can give way to IC ground states as further-neighbor exchange gains importance upon cooling (e.g. ref. 31). Thirdly, in inversion breaking systems with strong Dzyaloshinskii-Moriya interactions, IC-C transitions appear at low temperature when solitonic distortions are introduced into an IC order via a magnetic field^{32–35}, especially if a large charge gap is opened due to nesting³⁵. The present study generalizes the second scenario to the case of complex, twisted magnetic textures, such as the C-SkL, with an important role for (weak) magnetic anisotropy.

We consider the observation of C-locking induced by magnetic fields, in our experiment, based on the single-ion contribution to the spin Hamiltonian (Supplementary Note 3)

$$\mathcal{H}_{\text{anis}} \propto -K_1 \sum_{\mathbf{G}} \sum_{\mathbf{q} \in \text{BZ}} S^z(\mathbf{q}) S^z(\mathbf{G} - \mathbf{q}) \left[\sum_{\mathbf{d} \in \text{u.c.}} e^{i\mathbf{G} \cdot \mathbf{d}} \right] \quad (2)$$

where $\mathbf{r} = \mathbf{R} + \mathbf{d}$ is the position of a magnetic ion, decomposed into a unit cell coordinate \mathbf{R} and an intra-cell coordinate; \mathbf{q} and \mathbf{G} are the momentum in the first Brillouin zone and a reciprocal lattice vector, respectively. Typically, a small number of Fourier modes $\mathbf{q} = \mathbf{q}_v$ and $\mathbf{G} = 0$ are selected in models of incommensurate helimagnetic ordering^{12,21,28}. Meanwhile, C-locking is favored by the Umklapp terms $\mathbf{G} \neq 0$ ^{36,37}, representing a coupling between the primary Fourier mode of a helimagnet and its higher harmonics. Specifically for the $q = 0.25 a^*$ C-SkL in $Gd_3Ru_4Al_{12}$, $S(3\mathbf{q}) \cdot S(\mathbf{q})$ and $S(2\mathbf{q})^2$ are the leading contributors, adding to $\mathbf{G} = (1, 0, 0)$ and coupling the helimagnetic

order to the lattice. Application of a magnetic field to IC-PS enhances the elliptic distortion as demonstrated in Fig. 3, amplifies anharmonicity, shifts \mathbf{q} away from the value preferred by the exchange interaction and towards commensurability, and ultimately induces C-locking between the spin texture and the underlying lattice. In Supplementary Note 3, we thus derive an expression for the energetic contribution that depends on the position of the skyrmion core. Nevertheless, a full numerical treatment of Eq. (2) with large numbers of Fourier modes, to capture changes in the optimal \mathbf{q} – as well as changes in the cycloidal / screw character of the modulation – as a function of magnetic field and temperature, remains a challenge at present (Supplementary Note 1).

To explain the effect of single-ion anisotropy more intuitively, we initialize C-SkL and C-PS on the lattice and allow the magnetic moments to relax in the combined potential of exchange interaction, magnetic anisotropy, and external field (white/green arrows in Fig. 4b, c). The red arrows show a significant distortion of the textures, especially around the south pole. This density of strongly distorted moments is much larger for the C-PS state with quasi one-dimensional spin texture, which has an extended south pole region. In particular, the south pole direction of the magnetic moment sphere in Fig. 1a corresponds to a point (to a line) in case of a SkL (of a PS). We also calculated the z -projection of magnetic moments for a spin model of C-PS, C-SkL, and other orders, using our experimental K_1 from FMR, thus confirming numerically the favorable pinning of the C-SkL due to stronger higher harmonics (Supplementary Fig. 5).

Discussion

Figure 1d reveals a discommensuration of the magnetic lattice, i.e., a slight offset from $q/a^* = 0.25$ that indicates occasional magnetic defects. The discommensuration effect is well understood in one-dimensional chain systems³⁸: For example, the introduction of small amounts of chemical disorder causes proliferation of discommensurations in spin-Peierls chains,

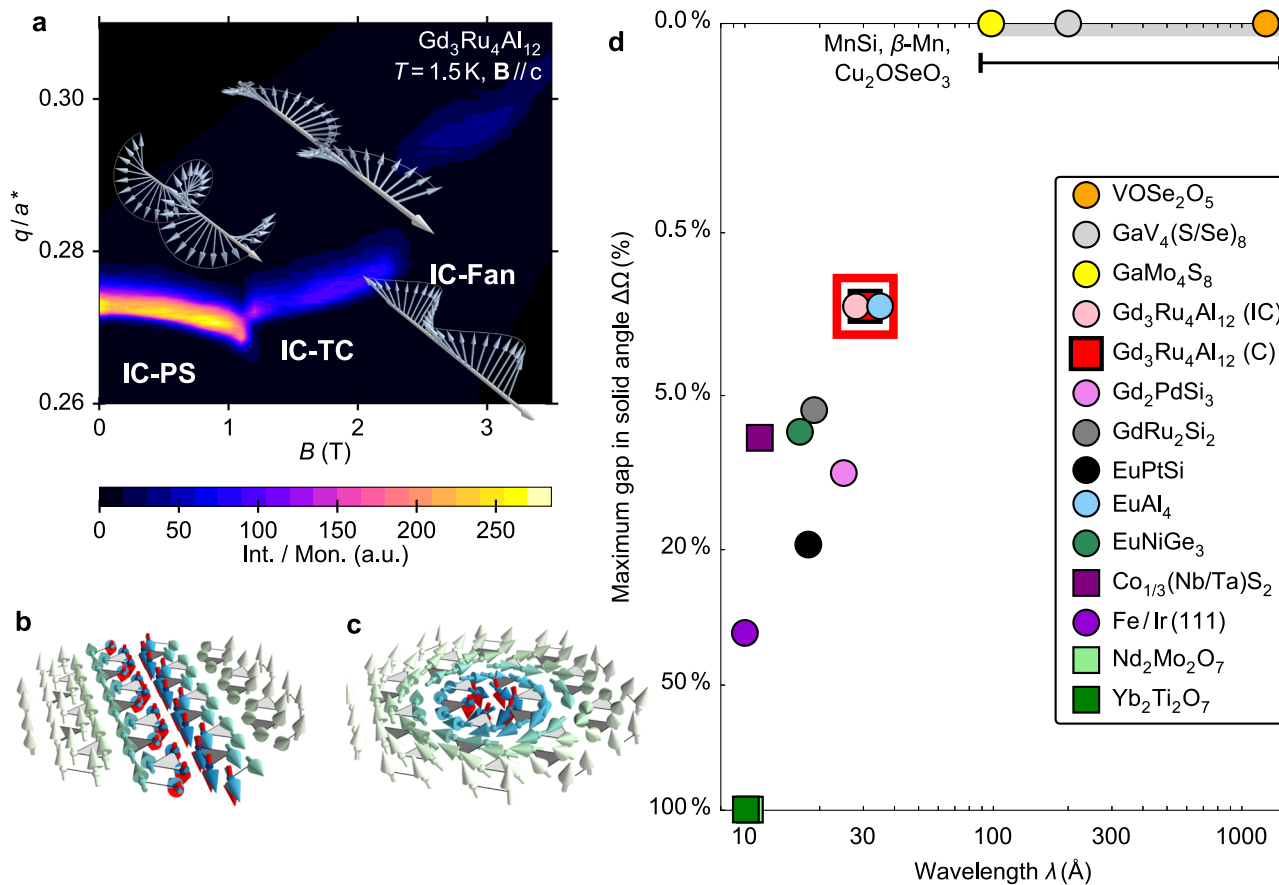


Fig. 4 | Stability of the commensurate skyrmion lattice (C-SkL) as compared to other magnetic phases. **a** Resonant x-ray scattering intensity, normalized to monitor counts, at base temperature ($T = 1.5$ K). Three incommensurate (IC) magnetic phases are shown: from left to right, proper screw (IC-PS), transverse conical (IC-TC), and fan-like (IC-Fan) as illustrated by insets. Quantitative analysis of Gaussian line-scan profiles is given in Supplementary Fig. 8. **b**, **c** Distortion of proper screw (left) and skyrmion textures in anisotropy potential and external magnetic field along the z -direction (Supplementary Note 1). The undistorted

moment directions (red) are superimposed on those moments that have been rotated by more than a critical angle. **d** Relationship of magnetic texture dimensions (λ) and coverage of directions on the sphere for various materials with noncoplanar textures and spin chirality^{3,11,13,14,44,45,51}. At the center of the plot, the commensurate C-SkL state in $Gd_3Ru_4Al_{12}$ is highlighted by a red box. On the y -axis, a continuous magnetic texture has zero *uncovered solid angle*, i.e. we assign a value of 0%. Note the stretched scale of the y -axis.

as manifested in a drop of λ_{disc} that ultimately destroys the commensurate (C) order^{39–42}. Discommensurations in two dimensions, mostly line defects, also appear for surface-adsorbed atoms and in the multi-directional charge-density wave state of transition metal dichalcogenides such as $2H-TaSe_2$, where C-IC transitions have been extensively studied^{36,37}. Among materials with two-dimensional spin textures, we compare to the C-SkL observed in an interfacial system^{24–26}. For inversion-breaking hcp-Fe/Ir(111) (for centrosymmetric $Gd_3Ru_4Al_{12}$), the spin structure is found to be commensurate within less than 10% (within 0.7%) of the magnetic period, and there is no (there is) evidence of C-IC transitions by cooling or application of a magnetic field. Based on measurements of magnetic anisotropy and modeling, we argue here that $Gd_3Ru_4Al_{12}$ has skyrmion cores on interstitial lattice sites, as does hcp-Fe/Ir(111); further that the discommensuration represents the appearance of line defects (characteristic spacing $\lambda_{disc} = 430$ nm) in the former – while phase-slip domain walls (spacing ~ 10 nm) and domains of the net magnetization (spacing ~ 30 nm) have been observed in the latter. We note that domain walls of the skyrmion helicity may appear in the bulk C-SkL of centrosymmetric materials^{6,43}; these are forbidden in inversion breaking platforms such as hcp-Fe/Ir(111).

Finally, Fig. 4d summarizes recent progress on material search and magnetic structure studies of noncoplanar magnets with lattice-averaged net spin chirality, especially SkL host compounds^{3,13,14,44,45}. As the dimension λ of the magnetic UC increases, magnetic moments cover all directions of the S_2 sphere, and the maximum *gap in angular*

coverage (as defined in percent of 4π) approaches zero. For example, noncoplanar antiferromagnets on the left side of Fig. 4d leave large fractions of S_2 out of reach of magnetic moments. Being located at the center of the plot, the C-SkL in the centrosymmetric, breathing kagome magnet $Gd_3Ru_4Al_{12}$ represents an essential link between long-period, incommensurate magnetic textures, stabilized e.g. by Dzyaloshinskii-Moriya interactions in $MnSi$ ^{3,46}, and so-called topological antiferromagnets with canted magnetic moments, such as pyrochlore systems⁴⁴. Although the present C-SkL is locked to the crystal lattice by weak in-plane magnetic anisotropy, confirmed here by ferromagnetic resonance experiments, the magnetic unit cell is large enough so that moments densely cover S_2 , leaving only marginal gaps on the order of 5% of 4π .

Methods

Cartographic projection

For visualizing the magnetic texture in Fig. 1a, we mapped a magnetic skyrmion onto a sphere in three dimensions, and subsequently mapped from the sphere onto a planar surface using the cartographic Nicolosi globular projection. Let (θ, φ) denote a point where a line between (x, y) and the north pole of the sphere penetrates the sphere's surface. These spherical coordinates are identified with (x, y) in Fig. 1a, upper side, while a point on the sphere is projected back onto the plane using the formalism in ref. 47.

Starting from polar angles θ, φ on the sphere, the x and y coordinates of the planar projection are defined as

$$x = \frac{\pi}{2}R \left(M + \operatorname{sgn}(\varphi - \varphi_0) \sqrt{M^2 + \frac{\cos^2(\theta)}{1 + b^2/d^2}} \right) \quad (3)$$

$$y = \frac{\pi}{2}R \left(N - \operatorname{sgn}(\theta) \sqrt{N^2 - \frac{d^2 \sin^2(\theta)/b^2 + d \sin(\theta) - 1}{1 + d^2/b^2}} \right) \quad (4)$$

with functions

$$M = \frac{b \sin(\theta)/d - b/2}{1 + b^2/d^2}, \quad (5)$$

$$N = \frac{d^2 \sin(\theta)/b^2 + d/2}{1 + d^2/b^2}, \quad (6)$$

$$b = \frac{\pi}{2(\varphi - \varphi_0)} - \frac{2}{\pi}(\varphi - \varphi_0), \quad (7)$$

$$c = \frac{2}{\pi}\theta, \text{ and} \quad (8)$$

$$d = \frac{1 - c^2}{\sin(\theta) - c}. \quad (9)$$

The sphere's radius R and the reference point for the longitude φ_0 can be set to 1 and 0, respectively.

Sample preparation and characterization

We prepared polycrystals of $\text{Gd}_3\text{Ru}_4\text{Al}_{12}$ by arc melting of the constituent elements in argon atmosphere, carefully turning pellets at least three times. Subsequently, single crystals were grown from these polycrystals by the floating zone technique under argon flow. Before the melting step, the halogen-lamp based furnace was evacuated to a base pressure of $8 \cdot 10^{-4}$ Pa, pumping for about three hours. The growth speed in the float zoning step was 2–4 mm/hr. We crushed single crystalline pieces into a fine powder for x-ray diffraction and refined the data by the Rietveld method using the RIETAN software package⁴⁸. In this analysis, we did not find impurity phases with volume fraction larger than 4%. Further, single-crystalline pieces were oriented using a Laue camera, cut with a diamond saw, and hand-polished to a high sheen ($\sim 1 \mu\text{m}$ grit) for single crystal x-ray diffraction measurements. Using a microscope with Nicolet prism, single-crystalline pieces with small amounts of RuAl_3 impurity phase, which forms tear-drop shaped inclusions of < 2% volume fraction that are hard to detect by laboratory x-rays, were excluded based on patterns in the surface-reflected light. Finally, magnetization measurements (M - H curve at $T = 2$ K, M - T curve at $\mu_0 H = 0.1$ T) were used to confirm a systematic evolution of long-range magnetic order in these single crystals.

Ferromagnetic resonance measurements

In order to avoid unwanted shape anisotropy effects, we polished the $\text{Gd}_3\text{Ru}_4\text{Al}_{12}$ samples cut within the a - c plane into a cylindrical disc form, with ellipticity below 4%. The ratio of diameter to thickness was 12 for the final disc, with a thickness of 120 μm .

High-field ferromagnetic resonance (FMR) measurements were performed at École Polytechnique Fédérale de Lausanne (EPFL, Switzerland) using a home built, high-sensitivity, quasi-optical spectrometer operating in the range of 50–420 GHz. This instrument covers a broad magnetic field regime up to 16 T, using a superconducting magnet. Its variable temperature insert operates across the temperature range 1.5–300 K, using the dynamic flow of helium gas, or liquid helium, through a heat exchanger right below the sample space. For more details, see ref. 49.

The overall temperature accuracy of the system is 0.1 K. The polished disc-like sample is mounted on a goniometer with the a - c plane coinciding with plane of rotation for the static magnetic field, the angular position of which is controllable and detectable via a potentiometer. Rotation proceeded in 5–10 degree steps, at sample temperature $T = 2$ K. The signal-to-noise ratio of the spectra is improved by recording the field-derivative of the absorbed power dP/dH using a lock-in technique with magnetic field modulation. The angular-dependent resonance data can be evaluated using the Smit-Beliers-Suhl formula as described in ref. 50. Using the saturation magnetization $M_S = 7 \mu_B / \text{Gd}^{3+}$, our fit yields the easy-plane anisotropy constant $K_1 = -1.944 \cdot 10^6 \text{ erg/cm}^3$ ($-0.13 \text{ meV} / \text{Gd}^{3+}$) and the temperature independent, isotropic g -factor 2.005.

Resonant x-ray scattering experiments (Gd- $L_{2,3}$)

Resonant elastic x-ray scattering (REXS) experiments are carried out in reflection geometry at RIKEN beamline BL19LXU of SPring-8 and beamline BL-3A of Photon Factory, KEK, with the sample mounted inside a cryomagnet. The preparation of Sample A, used to obtain the data in Fig. 3b, is discussed in ref. 11. Sample B, which was used to obtain all other data in this manuscript, has a surface perpendicular to the [110] axis, and was polished to reduce loss of intensity by diffuse scattering of x-rays. The energy of incident x-rays is matched to the L_2 or L_3 absorption edge of Gadolinium, where magnetic scattering involves virtual excitations from the $2p$ to the $5d$ atomic shells (Supplementary Fig. 2). The $5d$ shell is coupled to the dominant magnetic species, the half-filled $4f$ orbitals, by intra-atomic exchange correlations.

For data in Figs. 1 and 4, which are collected at the Gd- L_3 edge ($E_{x\text{-ray}} = 7.243 \text{ keV}$) at BL19LXU of SPring-8, we do not carry out polarization analysis of the diffracted beam. For data in Fig. 3 and Supplementary Fig. 3, which were collected at the Gd- L_2 edge ($E_{x\text{-ray}} = 7.932 \text{ keV}$) at BL-3A of Photon Factory, $\pi - \sigma'$ and $\pi - \pi'$ components of the diffracted beam are separated using an analyser plate made from pyrolytic graphite (PG-006).

The details of the polarization analysis in the latter case are as follows. Let \mathbf{k}_i (\mathbf{k}_f) be the wavevector of the incoming (outgoing) x-ray beam with polarization vector $\boldsymbol{\varepsilon}_i$ ($\boldsymbol{\varepsilon}_f$), where the scattering plane is spanned by the two wavevectors; c.f. purple plane in Fig. 1c. Let z be the direction perpendicular to the scattering plane. The incident beam is π -polarized, so that $\boldsymbol{\varepsilon}_i \perp \mathbf{e}_z, \mathbf{k}_i$. In the resonant elastic scattering process, the scattering cross-section f_{res} contains a term $\propto (\boldsymbol{\varepsilon}_i \times \boldsymbol{\varepsilon}_f) \cdot \mathbf{m}(\mathbf{q})$, where $\mathbf{q} = \mathbf{k}_f - \mathbf{k}_i$, and $\mathbf{m}(\mathbf{q})$ are the momentum transfer and the periodically modulated magnetic moment, respectively (Fig. 1c). In the present case, where $\boldsymbol{\varepsilon}_i \propto \mathbf{k}_i \times \mathbf{e}_z$ and the scattering plane is aligned with the crystal's hexagonal ab plane, we separate scattered x-rays with $\boldsymbol{\varepsilon}_f \parallel \mathbf{e}_z \parallel \mathbf{c}^*$ (i.e., $\pi - \sigma'$) and $\boldsymbol{\varepsilon}_f \propto \mathbf{k}_f \times \mathbf{e}_z$ (i.e., $\pi - \pi'$). Hence $f_{\text{res}}^{\pi - \sigma'} \propto \mathbf{k}_i \cdot \mathbf{m}(\mathbf{q})$ and $f_{\text{res}}^{\pi - \pi'} \propto (\mathbf{k}_i \times \mathbf{k}_f) \cdot \mathbf{m}(\mathbf{q}) \propto m_z(\mathbf{q}) \sin(2\theta)$, where 2θ is the scattering angle between \mathbf{k}_i and \mathbf{k}_f . The observed scattered intensities are $I \propto |f_{\text{res}}|^2$.

We obtain integrated intensities $I_{\pi - \sigma'}$ and $I_{\pi - \pi'}$ from Gaussian fits to line-cuts in momentum space. As described, the intensity ratio $R = I_{\pi - \sigma'} / I_{\pi - \pi'}$ at a given magnetic reflection is sensitive to the cycloidal (proper screw) character of the magnetic order by virtue of being large (small) when $\mathbf{k}_i \cdot (\mathbf{q} - \mathbf{G}')$ is large (small). For proper screw order, magnetic moments arrange themselves perpendicular to the propagation direction ($\mathbf{q} - \mathbf{G}'$), with a finite projection onto both \mathbf{e}_z (i.e., m_z) and the in-plane vector $\mathbf{e}_\perp = (\mathbf{q} - \mathbf{G}') \times \mathbf{e}_z$ (i.e., \mathbf{m}_{ip}). Here we introduce \mathbf{G}' , the closest reciprocal lattice vector to \mathbf{q} , and \mathbf{e}_z a unit vector along the c -direction. It follows $R \sin^2(2\theta) \propto [\mathbf{k}_i \cdot \mathbf{e}_\perp(\mathbf{q})]^2$ for a proper screw-type order, as demonstrated for $\text{Gd}_3\text{Ru}_4\text{Al}_{12}$ in Fig. 3b, c. Moreover, R provides information on the degree of elliptical distortion, capturing e.g. the ratio $(m_y/m_z)^2$ for a proper screw propagating along \mathbf{e}_x , written as $\mathbf{e}_y \cdot m_y \sin(2\pi x/\lambda) + \mathbf{e}_z \cdot m_z \cos(2\pi x/\lambda)$, where $\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z$ are Cartesian unit vectors aligned so that $\mathbf{e}_x \parallel \mathbf{q}$. In real materials with large (classical) magnetic moments, where the length of the moment is fixed to be spatially uniform in space, this type of deformed screw can be realized by anharmonic distortion of the proper screw texture, as shown in Fig. 3b (inset).

Data availability

The data supporting the findings of this study are available from the corresponding author upon reasonable request.

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Author contributions

Ma.Hi., I.K., Y.To. and T.-h.A. conceived the project. Ma.Hi. synthesized and characterized the single crystals. M.He., B.G.S., H.-A.K.v.N. and L.F. carried

out electron spin-resonance experiments at EPFL. Ma.Hi., L.S., H.O. and Y.Ta. performed REXS at SPring-8. Ma.Hi., L.S., S.G., K.K., H.S., H.N. and Y.Y. carried out REXS measurements at BL-3A of Photon Factory (KEK). Ma.Hi. and S.E. were in charge of spin model calculations. Ma.Hi. wrote the manuscript in close collaboration with M.M.H. and S.E., and with contributions and comments from all co-authors.

Competing interests

The authors declare no competing interests.

Additional information

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Correspondence and requests for materials should be addressed to Max Hirschberger.

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