Nanometric square skyrmion lattice in a centrosymmetric tetragonal magnet

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Magnetic skyrmions are topologically stable spin swirls with a particle-like character and are potentially suitable for the design of high-density information bits. Although most known skyrmion systems arise in non-centrosymmetric systems with a Dzyaloshinskii–Moriya interaction, centrosymmetric magnets with a triangular lattice can also give rise to skyrmion formation, with a geometrically frustrated lattice being considered essential in this case. Until now, it remains an open question if skyrmions can also exist in the absence of both geometrically frustrated lattice and inversion symmetry breaking. Here we discover a square skyrmion lattice state with 1.9 nm diameter skyrmions in the centrosymmetric tetragonal magnet GdRu$_2$Si$_2$, without a geometrically frustrated lattice by means of resonant X-ray scattering and Lorentz transmission electron microscopy experiments. A plausible origin of the observed skyrmion formation is four-spin interactions mediated by itinerant electrons in the presence of easy-axis anisotropy. Our results suggest that rare-earth intermetallics with highly symmetric crystal lattices may ubiquitously host nanometric skyrmions of exotic origins.

Ordering of magnetic moments in non-collinear or non-coplanar textures has been known as a source of plentiful and intriguing phenomena. Among these, the magnetic skyrmion generally appears as a vortex-like swirling spin texture with a non-zero integer skyrmion number $n_s$ defined by:

$$n_s = \frac{1}{4\pi} \int \left( \frac{\partial n}{\partial x} \times \frac{\partial n}{\partial y} \right) dxdy$$

which represents how many times the spin directions wrap a unit sphere. Here the integral is taken over the two-dimensional magnetic unit cell, and $n(r) = m(r)/|m(r)|$ is the unit vector that points along the local magnetic moment $m(r)$. This indicates that a magnetic skyrmion has the character of a countable particle. Its size typically ranges from a few to hundreds of nanometres. In metallic materials, the skyrmion motion can be efficiently driven by the electric current through a spin transfer torque, and the interplay between the conduction electron and the skyrmion spin texture via a quantum Berry phase also leads to unique transport phenomena, such as the topological Hall effect. Such a stable particle nature, small size and electric controllability highlights the magnetic skyrmion as a potential new information carrier for high-density magnetic storage devices.

Previously, the emergence of magnetic skyrmions has mostly been reported for a series of materials with non-centrosymmetric structures, in which the inherent spin-twisting interaction, termed the Dzyaloshinskii–Moriya interaction, plays a crucial role in the skyrmion formation. In such systems, the helical spin texture, $m(r) = m_0 \exp[iQ \cdot r] + c.c.$ (with $m_0$ being a complex vector and $c.c.$ representing complex conjugate), characterized by single magnetic modulation vector $Q$ (that is, a single-Q state) is realized for the magnetic field $B=0$. The application of an external magnetic field stabilizes the triangular lattice of skyrmions, which can be approximately described by $m(r) = (0, 0, m_y) + \sum_{n=1,2,3} (m_0 \exp[iQ_n \cdot r] + c.c.)$ plus higher harmonics with three distinctive fundamental modulation vectors $Q_n$ (triple-Q state). Therein, the internal spin texture of the skyrmion is determined by the crystallographic symmetry of the target system, and the observation of Bloch-type, Néel-type and anti-vortex (or antiskyrmion)-type skyrmions has been reported for chiral, polar and D$_h$ symmetry of bulk magnets, respectively. However, recent theoretical studies suggest that skyrmions can be stabilized even in centrosymmetric systems by considering different microscopic mechanisms. For example, the geometrical frustration of short-range exchange interactions on a triangular lattice is predicted to stabilize a hexagonal lattice of skyrmions. Another potential mechanism is the interplay of Ruderman–Kittel–Kasuya–Yosida (RKKY) and four-spin interactions associated with $s-d$ or $s-f$ coupling mediated by itinerant electrons, which is expected to favour a multiple-Q skyrmion lattice state for a highly symmetric (such as hexagonal or tetragonal) crystal lattice system. In the latter scenario, the wave vector of magnetic modulation is governed by the long-range RKKY interaction, which can cause a kind of magnetic frustration.
An important challenge is the search for an appropriate material system to experimentally realize these situations. Very recently, it was discovered that the centrosymmetric triangular-lattice magnet Gd₃P₄Si₁₂ hosts a hexagonal lattice of skyrmions, which is characterized by a small skyrmion diameter, \( \lambda_s \) of 2.4 nm with an associated giant topological Hall effect. A similar skyrmion formation was also reported for the centrosymmetric Gd₃Ru₄Al₁₂ with a breathing kagomé network. For these compounds, the existence of a geometrically frustrated lattice is considered to be the key for the skyrmion formation. Nevertheless, in principle, the competition of long-range magnetic interactions mediated by itinerant electrons can be allowed for any type of crystal lattice. Therefore, it remains an important question whether similar skyrmion formation in centrosymmetric magnets without geometrically frustrated lattices is possible or not. In this study, we focus on the centrosymmetric tetragonal magnet Gd₃Ru₄Si₁₂ without a geometrically frustrated lattice, and investigated its magnetic structure in detail. We found the emergence of the double-\( \mathbf{Q} \) square skyrmion lattice state in an out-of-plane magnetic field. The observed skyrmion diameter is as small as 1.9 nm, which is the smallest among those reported for single-component bulk materials. Our present results demonstrate that a skyrmion lattice can be formed even without a geometrically frustrated lattice or inversion symmetry breaking, and suggest that, in general, rare-earth intermetallics with a highly symmetric crystal lattice can be a promising material platform to realize nanometric skyrmions of exotic origins.

**Magnetic phase diagram for \( B//[001] \)**

Gd₃Ru₄Si₁₂ belongs to the family of RM₃X₄ compounds crystallized in a ThCr₂Si₂-type structure with centrosymmetric tetragonal space group \( I4/mmm \) (R, rare-earth element; M, a 3d, 4d or 5d element; X, Si or Ge). The crystal structure consists of alternate stacking of square lattice Gd layers and Ru₄Si₁₂ layers as presented in Fig. 1a, with the magnetism governed by Gd\(^{3+}\) (spin = 7/2, orbital moment = 0) ions with a Heisenberg magnetic moment. According to previous reports, this compound hosts incommensurate magnetic order below a Néel temperature of ~46 K (refs. 31,32), with the magnetic modulation vector \( \mathbf{Q} = (0.22, 0, 0) \) confined within the tetragonal basal plane. Application of a magnetic field \( B \) along the [001] axis induces several magnetic phase transitions as summarized in Fig. 1b, but the detailed magnetic structure in each phase has not been identified.

Figure 1f–h,j–l indicates the magnetic field dependence of magnetization \( M \), longitudinal resistivity \( \rho_{xx} \), and Hall resistivity \( \rho_{xy} \) measured at 5 K for \( B//[001] \). The magnetization profile shows clear
step-like anomalies at 2.0 and 2.4 T, before reaching the saturated ferromagnetic state with $M \approx 7 \mu_B$ per Gd$^{3+}$ at 10 T. These magnetic transitions are accompanied by large changes in $\rho_{xx}$ and $\rho_{yx}$, which suggests a close correlation between magnetic orders and transport properties. Figure 1b,i summarizes the $B$–$T$ magnetic phase diagram of GdRu$_2$Si$_2$, in which three distinctive magnetic phases (phases I, II and III) can be identified below 20 K.

**Magnetic structure analysis**

To resolve the detailed magnetic structure in each phase, we performed magnetic resonant X-ray scattering (RXS) experiments in resonance with the Gd $L_3$ edge at 5 K. By exploring the magnetic satellite peaks around the fundamental Bragg reflection indexed as $(4, 0, 0) \pm Q$, the magnetic modulation vector $Q$ in each phase was investigated directly. In Fig. 2a,d,g, the scattering profiles along $(4 - \delta, 0, 0)$ measured at 0, 2.1 and 3 T (which correspond to phases I, II and III, respectively) are plotted, in which the existence of the magnetic modulation vector $Q = (q, 0, 0)$ with $q \approx 0.22$ is clearly identified in all three phases. The tetragonal symmetry of the crystal structure means the appearance of an equivalent magnetic modulation vector $Q = (0, q, 0)$ is expected, which has also been identified in the scattering profiles along $(4, -\delta, 0)$, shown in Fig. 2b,e,h. In Fig. 2j–l, the magnetic-field dependence of magnetization, wave-number $q$ of magnetic modulation and integrated intensity of the magnetic peak are plotted. On the magnetic phase transitions at 2.0 and 2.4 T, both the $q$ value and the peak intensity show clear step-like anomalies.

Here the simultaneous appearance of $Q = (q, 0, 0)$ and $Q = (0, q, 0)$ can be interpreted as either a double-$Q$ magnetic state or multiple domains of a single-$Q$ magnetic state. One of the most effective ways to distinguish these two possibilities is the identification of additional $Q + Q$ modulations, which are allowed to appear only for the former double-$Q$ magnetic state. In Fig. 2c,f,i, the corresponding scattering profiles along $(4 - \delta, -\delta, 0)$ are plotted, in which the $Q + Q$ modulation is identified only in the intermediate phase II, but not in phases I and III. Such a feature can be more directly confirmed in the magnetic field dependence of the integrated intensity for the $Q + Q$ magnetic peak (Fig. 2m), in which the appearance of the $Q + Q$ modulation is clearly correlated with the transition into phase II. On the basis of these results, we conclude that phase II is a double-$Q$ magnetic state.

**Spin texture of magnetic phases**

Next, to investigate the spin orientation in each magnetic phase, we analysed the polarization of a scattered X-ray. The experimental geometry is illustrated in Fig. 3a,b, in which the magnetic satellite peaks at the $(4, -2, 0) \pm Q$ position are investigated. Here, the incident beam is always polarized parallel to the scattering plane ($\pi$ polarized). The scattered beam may include two polarization components: parallel ($\pi$) and perpendicular ($\sigma$) polarizations. The intensities of the two components of the scattered beam ($I_{\pi}$ and $I_{\sigma}$, respectively) are measured separately. When the magnetic structure contains the modulated spin component ($\mathbf{m}_Q \exp[\mathbf{Q} \cdot \mathbf{r}] + \text{c.c.}$), the corresponding magnetic scattering intensity is given by

$$I \propto |(\mathbf{e}_x \times \mathbf{e}_y) \cdot \mathbf{m}_Q|^2 \quad \text{(2)}$$

where $\mathbf{e}_x$ and $\mathbf{e}_y$ represent the polarization vectors of incident and scattered beams, respectively. As $\mathbf{e}_z$ is almost parallel to the [010] axis in the present experimental geometry, $I_{\pi}$ and $I_{\sigma}$ should mainly reflect the [001] and [100] components of $\mathbf{m}_Q$. On the basis of the above discussion, we first investigated the magnetic structure at 0 T, which corresponds to Phase I. In Fig. 3c, the existence of the $I_{\pi}$-absence of $I_{\sigma}$ intensity indicates the existence of [001]-modulated spin component for $Q = (q, 0, 0)$. Likewise, Fig. 3e demonstrates the coexistence of both [001]- and [100]-modulating spin components for $Q = (0, q, 0)$. Note that there appears an additional peak at $\delta \approx 0.226$ in the $(4, -2, 0) \pm Q$ scan having the same selection rule as phase III, whose origin is discussed in Supplementary Note II. These data suggest that the spin texture in phase I can be approximately described by the screw structure, in which neighbouring magnetic moments rotate within a plane normal to the magnetic modulation vector $Q$, as shown in Fig. 3g,h. Similar measurements were also performed for phase II.
 approximately described by: 

\[ \mathbf{m}(r) = S(r)/|S(r)| \]  

(3)

\[ S(r) = \{(0, -i, 1)\exp[iQ_1 \cdot r] + (i, 0, 1)\exp[iQ_2 \cdot r] + (0, 0, S_0)\} + c.c. \]  

(4)

Here, \( \mathbf{S} \) is a vector giving the direction of local magnetic moment, and \( \mathbf{S} \) approximately scales with the \( B \)-induced out-of-plane uniform magnetization component. The normalization process leads to the emergence of higher harmonic terms in \( \mathbf{m}(r) \) with the wave vectors such as \( 2Q_1 \), \( 2Q_2 \), and \( Q_1 + Q_2 \), whose existence has also been confirmed experimentally (Fig. 2f, Supplementary Fig. 6 and the related discussion in Supplementary Note V). Figure 1d describes the spin texture given by equation (3), which is characterized by the integer skyrmion number \( n_s = -1 \) per magnetic unit cell according to equation (1) and can be considered as the square lattice of magnetic skyrmions (Supplementary Fig. 4 and Supplementary Note IV). Similar polarization analysis was also performed for phase III, whose magnetic structure is displayed in Fig. 1c. (see Supplementary Note II for the details). In general, the skyrmion lattice phase is separated by first-order phase-transition boundaries with other topologically trivial magnetic phases \(^{12,15,29,30}\), which is consistent with the observed \( M-B \) profile with two discontinuous magnetization steps with a clear hysteresis (Fig. 2g).

Note that the conduction electrons that interact with the skyrmion spin texture gain an extra Berry phase that acts on the conduction electrons as an emergent magnetic field, which causes an additional contribution to the Hall resistivity proportional to the skyrmion density, that is, the topological Hall effect. As shown in Fig. 1h, an enhancement of \( \rho_y / \rho_x \) is identified in phase II (that is, in the SkL phase), which may contain the possible contribution from the topological Hall effect. Reflecting the strong correlation between magnetism and transport properties, GdRu\(_2\)Si\(_2\) also shows a non-monotonous magnetoresistance and large anomalous Hall effect (Fig. 1g,h). In such a situation, the interpretation of transport properties is not straightforward. Our tentative analysis in terms of the topological Hall effect is provided in Supplementary Note VI, but the quantitative evaluation remains a future challenge.

Real-space imaging of square skyrmion lattice

Figure 4a indicates the under-focused Lorentz transmission electron microscopy (L-TEM) image obtained at 1.95 T and 8 K (that is, in phase II), with a Fourier transform pattern in Fig. 4b,c. Here, the peaks marked by white circles are related to the crystal structure (whose corresponding Miller indices are also indicated) and the fourfold peaks marked by yellow circles (which correspond to the magnetic modulation vectors \( Q_1 = (0.22, 0, 0) \) and \( Q_2 = (0, 0.22, 0) \)) are related to the magnetic structure. These results are in good agreement with the RXS results. To filter the undesired background noise, we picked up the circled areas (that is, the reflections related to crystalline II and magnetic structures) in Fig. 4b and reversed the Fourier transformation. The result is shown in Fig. 4e, which clearly visualizes the square-lattice-like magnetic superstructure with a

![Image](https://www.nature.com/naturenanotechnology)

**Fig. 3 | Polarization analysis of RXS profiles in phase I and phase II.** a, Experimental set-up for RXS measurements. Satellite peaks around the fundamental Bragg spot \((4, -2, 0)\) are investigated. Scattering plane lies perpendicular to the [001] axis. \( \mathbf{k} \) and \( \mathbf{h} \) are the propagation vectors of the incident and scattered X-ray. \( \pi (\mathbf{r}') \) and \( \sigma (\mathbf{r}') \) represent the polarization direction of the incident (scattered) X-ray. b, The measurement configuration projected along the [001] direction. In this set-up, \( \mathbf{k} \) is almost parallel to the [100] axis. c-e, Line profiles for \((4, -\delta, 2, 0)\) and \((4, -2, 0)\) scans, which correspond to \( Q_1 \) and \( Q_2 \), measured at 0 T (phase I). d,f, Corresponding line profiles at 2.1 T (phase II). As discussed in the main text, the intensities of the \((\pi - \pi')\) and \((\pi - \sigma')\) channels mainly reflect the [001] and [100] components of \( m_\parallel \), respectively. g,h, Line-scan direction in reciprocal space, as well as a real-space illustration of modulated spin components that belong to each magnetic modulation vector \( Q_1 \) (g) and \( Q_2 \) (h). In e, an additional peak (highlighted by *) is observed at \( \delta \approx 0.226 \), whose origin is discussed in Supplementary Note II. Error bars indicate one s.d.
period of 1.9 nm formed on the atomic lattice. Figure 4f,g show filtered images obtained from the under-focused and over-focused L-TEM images, respectively, based only on the magnetic reflections circled in Fig. 4c. Both under-focused and over-focused images show a square-lattice-like pattern but with an opposite black/white contrast. By performing the transport-of-intensity equation analysis based on these two images, the spatial distribution of the in-plane local magnetization is deduced as shown in Fig. 4h. We can confirm the square lattice of the vortex-like spin arrangements, which is revealed to be consistent with the spin texture in Fig. 1d and represents the square skyrmion lattice described by equation (3). Above Curie temperature, all the magnetic contrast and the corresponding magnetic reflections disappear, as shown in Fig. 4d.

**Microscopic origins of square skyrmion lattice**

Theoretically, several distinct mechanisms for the emergence of multiple-Q spin textures have been proposed, such as (1) the Dzyaloshinskii–Moriya interaction in non-centrosymmetric magnets, (2) the competition of short-ranged exchange interactions in geometrically frustrated magnets and (3) the interplay of RKKY and four-spin interactions related to the coupling between a conduction electron and localized moment in itinerant magnets with a highly symmetric lattice. Here, mechanisms (2) and (3) are associated with magnetic frustration in a broad sense, and the magnetic modulation is generated by the sign-alternating competing spin exchange interactions in both cases. In the latter case for itinerant magnets, the presence or absence of magnetic frustration is not directly linked to the underlying crystal lattice geometry.

As the crystal structure of GdRu2Si2 is centrosymmetric, the contribution from the Dzyaloshinskii–Moriya interaction is not relevant in the present case. Instead, the family of RuRu2 systems commonly shows incommensurate magnetic modulation along in-plane directions, which has been discussed in terms of \((m_0 \cdot m_0)^\text{-type RKKY interactions that reflect Fermi surface properties}. A similar mechanism was also discussed to describe the coplanar triple-Q magnetic order in the hexagonal magnet YCo2Sn5 (Ref. 27). Here, the four-spin interaction is generally described as \(\sum K_{ijkl}[(m_i \cdot m_j)(m_k \cdot m_l)]\) among the four magnetic sites \(i, j, k, l\) in real space or \(\sum K_{ijkl}[(m_0 \cdot m_0)(m_0 \cdot m_0)]\delta(Q_1 + Q_2 + Q_3 + Q_4)\) among the wavenumbers \(Q\), \(\nu = 1, 2, 3, 4\) to give multiple maxima in the bare susceptibility in reciprocal space, which lifts the degeneracy between multiple-Q and single-Q magnetic orders. The present discovery of a square skyrmion lattice in GdRu2Si2 suggests that a similar mechanism may also promote the SKL formation in single-component bulk materials. In this case, the tetragonal symmetry of the underlying crystal lattice allows the existence of a
multiple number of equivalent magnetic modulation vectors $Q$, and $Q$, determined by RKKY interaction, and the additional contribution from the four-spin interaction stabilizes the double-$Q$ square skyrmion lattice orders. Recently, the tetragonal itinerant magnet CeAuSb$_2$ was reported to host a double-$Q$ collinear spin density wave state under $B_{||}[001]$, but this material is characterized by a strong Ising magnetic anisotropy, so skyrmion formation is not allowed. In contrast, magnetism in GdRu$_2$Si$_2$ is dominated by Gd$^{3+}$ Heisenberg spins, which should be another important factor for the present skyrmion formation. Note that GdRu$_2$Si$_2$ is characterized by a moderate amplitude of easy-axis anisotropy, which can also provide the effective four-spin interaction and stabilize the multiple-$Q$ order. Such an interplay among the magnetic interactions mediated by itinerant electrons and the easy-axis magnetic anisotropy is perhaps responsible for the present SKL formation.

Conclusions
In the case of Dzyaloshinskii–Moriya-induced skyrmions, a close-packed triple-$Q$ hexagonal SKL is usually favoured irrespective of the underlying crystal symmetry, and the double-$Q$ square SKL is rather exceptional. The present observation of a square SKL in tetragonal GdRu$_2$Si$_2$ suggests a strong correlation between lattice symmetries of SKL and the crystal structure, which can be a unique feature of centrosymmetric rare-earth intermetallics. Interestingly, GdRu$_2$Si$_2$ has the same crystal structure as the 122-type Fe-based superconductors, and this family of materials allows a wide variety of chemical parameter tuning. Because of the lack of geometrically frustrated lattice and inversion symmetry breaking, the present RM$_X$S$_2$ system with its centrosymmetric tetragonal lattice may offer an ideal material platform to explore the novel skyrmion formation mechanism potentially mediated by itinerant electrons. The small skyrmion size and spin-helicity degree of freedom are other advantages of GdRu$_2$Si$_2$, and further systematic investigation of the detailed magnetic structure as well as its relationship with the electronic structure and emergent electromagnetic responses for this material family will be essential. Our results establish that skyrmions can be stabilized without the need for a geometrically frustrated lattice or inversion symmetry breaking, which suggests a new route for the design of nanometric topological spin textures in single-component systems. Further experimental identification of an isolated skyrmion particle in such systems is an important challenge for potential applications.

Online content
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References
1. Nagaosa, N. & Tokura, Y. Emergent electromagnetism in solids. *Phys. Scripta* **T146**, 014020 (2012).
2. Rößler, U. K., Bogdanov, A. N. & Pfleiderer, C. Spontaneous skyrmion ground states in magnetic metals. *Nature* **442**, 797–801 (2006).
3. Rößler, U. K., Leonov, A. A. & Bogdanov, A. N. Chiral skyrmionic matter in non-centrosymmetric magnets. *J. Phys. Conf. Ser.* **303**, 012105 (2011).
4. Nagaosa, N. & Tokura, Y. Topological properties and dynamics of magnetic skyrmions. *Nat. Nanotechnol.* **8**, 899–911 (2013).
5. Neubaer, A. et al. Topological Hall effect in the A phase of MnSi. *Phys. Rev. Lett.* **102**, 136602 (2009).
6. Schulz, T. et al. Emergent electrodynamics of skyrmions in a chiral magnet. *Nat. Phys.* **8**, 301–304 (2012).
7. Fert, A., Cros, V. & Sampaio, J. Skyrmions on the track. *Nat. Nanotechnol.* **8**, 152–156 (2013).
8. Kanazawa, N., Seki, S. & Tokura, Y. Noncentrosymmetric magnets hosting magnetic skyrmions. *Adv. Mater.* **29**, 1603227 (2017).
9. Müllerbauer, S. et al. Skyrmion lattice in a chiral magnet. *Science* **323**, 915–919 (2009).
10. Yu, X. Z. et al. Real-space observation of a two-dimensional skyrmion crystal. *Nature* **465**, 901–904 (2010).
11. Seki, S., Yu, X. Z., Ishiwata, S. & Tokura, Y. Observation of skyrmions in a multiferroic material. *Science* **336**, 198–201 (2012).
12. Tokunaga, Y. et al. A new class of chiral materials hosting magnetic skyrmions beyond room temperature. *Nat. Commun.* **6**, 7638 (2015).
13. Karube, K. et al. Robust metastable skyrmions and their triangular–square lattice structural transition in a high-temperature chiral magnet. *Nat. Mater.* **11**, 1237–1242 (2016).
14. Kakihana, M. et al. Giant Hall resistivity and magnetoresistance in cubic chiral antiferromagnet EuPtS$_3$. *J. Phys. Soc. Jpn* **87**, 023701 (2018).
15. Tanigaki, T. et al. Real-space observation of short-period cubic lattice of skyrmions in MnGe. *Nano Lett.* **15**, 5438–5442 (2015).
16. Kurumaji, T. et al. Néel-type skyrmion lattice with confined orientation in the polar magnetic semiconductor Ga$_2$V$_5$S$_7$. *Nat. Mater.* **14**, 1116–1122 (2015).
17. Kurumaji, T. et al. Néel-type skyrmion lattice in the tetragonal polar magnet VO$_x$Se$_y$. *Phys. Rev. Lett.* **119**, 237201 (2017).
18. Nayak, A. K. et al. Magnetic antiskyrmions above room temperature in noncolinear Heusler materials. *Nature* **548**, 561–566 (2017).
19. Batista, C. D., Lin, S. Z., Hayami, S. & Kamiya, Y. Frustration and chiral orderings in correlated electron systems. *Rep. Prog. Phys.* **79**, 084504 (2016).
20. Okubo, T., Chung, S. & Kawamura, H. Multiple-q states and the skyrmion lattice of the triangular-lattice Heisenberg antiferromagnet under magnetic fields. *Phys. Rev. Lett.* **108**, 017206 (2012).
21. Leonov, A. O. & Mostovoy, M. Multiply periodic states and isolated skyrmions in an anisotropic frustrated magnet. *Nat. Commun.* **6**, 8275 (2015).
22. Lin, S. Z., Saxena, A. & Batista, C. D. Skyrmion fractionalization and merons in chiral magnets with easy-plane anisotropy. *Phys. Rev. B* **91**, 224407 (2015).
23. Ozawa, R. et al. Vortex crystals with chiral stripes in itinerant magnets. *J. Phys. Soc. Jpn* **85**, 103703 (2016).
24. Ozawa, R., Hayami, S. & Motome, Y. Zero-field skyrmions with a high topological number in itinerant magnets. *Phys. Rev. Lett.* **118**, 147205 (2017).
25. Hayami, S., Ozawa, R. & Motome, Y. Effective bilinear–biquadratic model for noncoplanar ordering in itinerant magnets. *Phys. Rev. B* **95**, 224424 (2017).
26. Heinze, S. et al. Spontaneous atomic-scale magnetic skyrmion lattice in two dimensions. *Nat. Phys.* **7**, 713–718 (2011).
27. Takagi, R. et al. Multiple-q noncollinear magnetism in an itinerant hexagonal magnet. *Sci. Adv.* **4**, eaau3002 (2018).
28. Martin, I. & Batista, C. D. Itinerant electron-driven chiral magnetic ordering and spontaneous quantum Hall effect in triangular lattice models. *Phys. Rev. Lett.* **101**, 156402 (2008).
29. Kurumaji, T. et al. Skyrmion lattice with a giant topological Hall effect in a frustrated triangular-lattice magnet. *Science* **365**, 914–918 (2019).
30. Hirschberger, M. et al. Skyrmion phase and competing magnetic orders on a breathing kagomé lattice. *Nat. Commun.* **10**, 5831 (2019).
31. Ślaski, M., Szytuła, A., Leciejewicz, J. & Zygmunt, A. Magnetic properties of RERu$_2$Si$_2$ (RE = Pr, Nd, Gd, Tb, Dy, Er) intermetallics. *J. Mag. Mag. Mater.* **46**, 114 (1984).
32. Garnier, A. et al. Anisotropic metamagnetism in GdRu$_2$Si$_2$. *J. Magn. Magn. Mater.* **140**, 899–900 (1995).
33. Devishivi, A. Magnetic Properties of Gd$^{3+}$ Based Systems. PhD Thesis, Univ. Vienna (2010).
34. Samanta, T., Dao, I. & Banerjee, S. Comparative studies of magnetocaloric effect and magnetotransport behavior in GdRu$_2$Si$_2$ compound. *J. Appl. Phys.* **104**, 123901 (2008).
35. Adams, T. et al. Long-range crystalline nature of the skyrmion lattice in MnSi. *Phys. Rev. Lett.* **107**, 217206 (2011).
36. Blume, M. In Resonant Anomalous X-Ray Scattering (eds Materlik, G., Sparks, C. J. and Fischer, K.) 495–512 (Elsevier, 1994).
37. Marcus, G. C. et al. Multi-q mesoscale magnetism in CeAuSb$_2$. *Phys. Rev. Lett.* **120**, 097201 (2018).
38. Yu, X. Z. et al. Transformation between meron and skyrmion topological spin textures in a chiral magnet. *Nature* **564**, 95–98 (2018).
39. Nakajima, T. et al. Skyrmion lattice structural transition in MnSi. *Sci. Adv.* **3**, 1602562 (2017).

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Methods

Crystal growth. Single crystals of GdRu2Si2 were grown by the optical floating zone method. Poly-crystals were prepared by the arc-melt technique from pieces of high-quality elements Gd (3N), Ru (3N) and Si (3N) using a water-cooled copper crucible under an Ar atmosphere. The obtained crystals were characterized by powder X-ray diffraction, which confirmed the purity of the sample. Crystal orientations were determined using the back-reflection X-ray Laue photography method.

Magnetic and electrical transport property measurements. Measurements of magnetic susceptibility (Supplementary Note I) and magnetization were performed using a superconducting quantum interference device magnetometer (MPMS 3, Quantum Design). To characterize the electrical transport properties, the rectangular bar-shaped samples were used with silver paste as an electrode. Measurements of $\rho_{xx}$ and $\rho_{yx}$ were performed using the AC-transport option in a physical property measurement system (PPMS, Quantum Design). To avoid the possible discrepancy due to shape anisotropy caused by the demagnetization effect, the same crystal was used for the measurements of magnetic and electrical transport properties.

RXS measurement. RXS measurements were performed on a BL-3A (Photon Factory, KEK). The photon energy of the incident X-ray was turned near the Gd L2 edge (~7.935 keV) with a Si(111) double-crystal monochromator. A Factory, KEK). The photon energy of the incident X-ray was turned near the Gd RXS measurement.

L-TEM measurements. L-TEM provides information on the in-plane magnetization of the sample. The bright or dark contrast in the defocused L-TEM images reflects the convergence or divergence of the deflected electron beam induced by a Lorentz force, which corresponds to the direction and magnitude of in-plane magnetizations. The under-focused and over-focused L-TEM images should provide the reversed contrast. Accordingly, magnetic skyrmions with clockwise or anticlockwise helicity can be observed as bright or dark dots on the under-focused or over-focused image plane.

To examine the modulated magnetic structure in the present magnet, we thinned the bulk sample with Ar+ milling after mechanical polishing and then put the thin sample on a double-tilt He cooling sample holder with a temperature controller (Gatan ULTDT) to control the sample temperature from 6 to 70 K, which is attached to a commercial transmission electron microscope (JEOL, JEM2800). L-TEM measurements were performed under a normal magnetic field of 1.95 T. The field map for SkL was obtained by analysing defocused L-TEM images using a commercial software package Qpt based on the transport-of-intensity equation.

Data availability

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

References

40. Yu, X. Z. et al. Aggregation and collapse dynamics of skyrmions in a non-equilibrium state. Nat. Phys. 14, 832–836 (2018).

41. Ishizuka, K. & Allman, B. Phase measurement of atomic resolution image using transport of intensity equation. J. Electron Microsc. 54, 191–197 (2005).

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