Zoology of Multiple-Q Spin Textures in a Centrosymmetric Tetragonal Magnet with Itinerant Electrons

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Magnetic skyrmion is a topologically stable particle-like swirling spin texture potentially suitable for high-density information bit, which was first observed in noncentrosymmetric magnets with Dzyaloshinskii–Moriya interaction. Recently, nanometric skyrmion has also been discovered in centrosymmetric rare-earth compounds, and the identification of their skyrmion formation mechanism and further search of nontrivial spin textures are highly demanded. Here, magnetic structures in a prototypical skyrmion-hosting centrosymmetric tetragonal magnet GdRu₂Si₂ is exhaustively studied by performing the resonant X-ray scattering experiments. A rich variety of double-Q magnetic structures, including the antiferroic order of meron(half-skyrmion)/anti-meron-like textures with fractional local topological charges are identified. The observed intricate magnetic phase diagram is successfully reproduced by the theoretical framework considering the four-spin interaction mediated by itinerant electrons and magnetic anisotropy. The present results will contribute to the better understanding of the novel skyrmion formation mechanism in this centrosymmetric rare-earth compound, and suggest that itinerant electrons can ubiquitously host a variety of unique multiple-Q spin orders in a simple crystal lattice system.

1. Introduction

Multiple-Q spin order, that is, two- or three-dimensionally modulated periodic structure characterized by a multiple number of coexisting modulation vectors Q, has recently attracted attention as the source of rich emergent phenomena.[1–3] When the local magnetic moment m(r) takes a non-coplanar configuration, multiple-Q orders are often endowed with a nontrivial topology. For example, the superposition of multiple helical modulations can be regarded as the periodic array of skyrmions (Figure 1d), which appears as a stable particle-like object with a swirling m(r) texture characterized by a nonzero integer topological charge N_{sk} = \int n_{sk} dx dy.[4,5] Here, the topological charge density n_{sk}(r) is defined as

\[ n_{sk} = \frac{1}{4\pi} \mathbf{n} \cdot \left( \frac{\partial \mathbf{n}}{\partial x} \times \frac{\partial \mathbf{n}}{\partial y} \right) \] (1)

with \mathbf{n}(r) = m(r)/|m(r)|, and N_{sk} reflects how many times \mathbf{n} wraps a unit sphere. Magnetic skyrmions are now intensively studied as a potential candidate of high-density information bit[1] or building block of natural magnonic crystal[6] that is potentially suitable for magnetic data processing, and further explorations of nontrivial multiple-Q orders with exotic origin are highly demanded.

Previously, the skyrmion lattice (SkL) state with multiple-Q character was mostly observed in noncentrosymmetric...
magnets with the relativistic Dzyaloshinskii–Moriya (DM) interaction. In this case, the magnetic phase diagram is rather simple, where the single-Q helical phase is realized in zero magnetic field and the multiple-Q skyrmion lattice phase is stabilized by the application of magnetic field \( H \). On the other hand, the skyrmion formation has recently been discovered in centrosymmetric systems, such as Gd\(_2\)PdSi\(_3\), Gd\(_3\)Ru\(_4\)Al\(_{12}\), and GdRu\(_2\)Si\(_2\). Notably, these centrosymmetric Gd-compounds charged by the application of magnetic field \( H \) ≈ 2 nm, which is one order of magnitude smaller than conventional DM-based noncentrosymmetric compounds. For such systems, several alternative skyrmion formation mechanisms have been proposed, while their experimental verification has rarely been achieved.

In this work, we have exhaustively studied magnetic structures for a prototypical skyrmion-hosting centrosymmetric tetragonal magnet GdRu\(_2\)Si\(_2\), by performing the resonant X-ray scattering (RXS) experiments. We identified a rich variety of double-Q magnetic structures, including the antiferro order of meron/antimeron-like textures with fractional local topological charge. The observed intricate magnetic phase diagram is successfully reproduced by the theoretical framework considering the four-spin interactions mediated by itinerant electrons and magnetic anisotropy. The present results will contribute to the better understanding of the novel skyrmion formation mechanism in this centrosymmetric rare-earth compound, and demonstrate that a variety of unique multiple-Q spin orders can be derived from a simple crystal lattice system with itinerant electrons.

Our target material GdRu\(_2\)Si\(_2\) crystallizes into a centrosymmetric tetragonal structure with space group 14/mmm, which consists of alternative stacking of the Gd square lattice and Ru\(_2\)Si\(_2\) layers along the [001] axis (Figure 1a). Magnetism is governed by Gd\(^{3+}\) ion with rather isotropic magnetic moment (\( S = 7/2, L = 0 \)). Below the magnetic ordering temperature \( T_N = 46 \) K, the appearance of incommensurate magnetic order with the modulation vector \( Q = (0.22, 0, 0) \) has been reported.

In Figure 1a,b, the \( H\)–\( T \) (temperature) magnetic phase diagrams of GdRu\(_2\)Si\(_2\) for \( H\|\{001\} \) and \( H\|\{100\} \) are summarized. The phase boundaries are determined by the magnetization measurements, where \( H\)-induced multiple-step metamagnetic transitions are observed for both \( H \) directions (see Note S1 and Figure S2a, Supporting Information). The phase II for \( H\|\{001\} \) has been identified as the square skyrmion lattice state, that is, the double-Q state described by the super-position of two orthogonally modulated screw structures and a uniform magnetization (Figure 1d). Note that the recent scanning tunneling microscopy (STM) experiment revealed the two-dimensionally modulated (i.e., double-Q) electron density distribution in Phase II, III, and possibly I, while this technique is rather sensitive to charge degree of freedom and the corresponding spin texture in the latter phases has not been confirmed directly. In addition, previous experiments mostly focused on \( H\|\{001\} \), and the magnetic structures under \( H\perp\{001\} \) remain unexplored.

## 2. Results and Discussions

### 2.1. Identification of the Single-Q and Double-Q Magnetic States

To elucidate the detailed magnetic structure in each phase, magnetic resonant X-ray scattering (RXS) experiments in resonance with the Gd L\(_2\) edge at 5K were performed. First, we examined the development of the magnetic modulation vector \( Q \) for \( H\|\{100\} \), by exploring the magnetic satellite reflections around a fundamental Bragg peak indexed as \( 0, 4, 0 \) ± \( Q \) (Figure 2). Figure 2a,b indicate the line-scan profile along \( (0, 4, 0) \) and \( (0, 4, -r, 0) \), which allows the identification of the magnetic modulation vectors \( Q_1 = (q, 0, 0) \) and \( Q_2 = (0, 0, q) \) that are parallel and perpendicular to the external magnetic field \( H \), respectively. Here, the sample was initially cooled at \( \mu_0 H = 0 \) T, and the data points represented by open symbols were obtained in a field-increasing process from 0 to 5 T. Magnetic-field dependence of the wavenumber \( q \) and integrated intensity for these magnetic satellite reflections are also plotted in Figure 2c,d. After the initial zero-field cooling (i.e., the phase I), the magnetic satellite reflections are found for both \( \mu_0 H \) = 100 and \( \{100\} \) directions. By applying \( H\|\{100\} \), the phase IV is stabilized for \( 2 < \mu_0 H < 4 \) T, where the magnetic satellite reflection is observed only parallel to \( H \). This result indicates that the phase IV is the single-Q state with \( Q_1 \| H \). Above 4 T (i.e., phase III′), the magnetic satellite reflection reappears along the both directions that are parallel and perpendicular to \( H \), which suggests that the phase III′ is a double-Q state. When the \( H\)-value is reduced from the phase III′, only \( Q_1 \| H \) survives in the phase IV, but then both \( Q_1 \| H \) and \( Q_2 \perp H \) reappear in phase I. Here, the phase I is characterized by the anisotropic magnetic satellite reflections with \( Q_1 = (0.219, 0, 0) \) and \( Q_2 = (0, 0.224, 0) \). This indicates that the phase I is an anisotropic double-Q state which breaks the fourfold-symmetry. It leads to the appearance of two kinds of domains with opposite anisotropy in the phase I, and the application of \( H\|\{100\} \) selects one of the magnetic domains. As detailed in Note S5, Supporting Information, the phase V appearing for larger \( H\)-value has been identified as a single-Q state with \( Q_2 \perp H \). In case of \( H\|\{001\} \), both \( Q_1 \) and \( Q_2 \) magnetic satellite peaks are always observed in the phase I, II, and III as discussed in Note S6, Supporting Information.

Next, to understand the detailed relationship between the magnetic phases stabilized for \( H\|\{100\} \) and \( H\|\{001\} \), \( M\)-\( H \) profiles have been measured for various directions of \( H \) rotated within the \( \{010\} \) plane (Figure S2a, Supporting Information). On the basis of these magnetization data, magnetic phase diagram as
Figure 1. Magnetic phase diagrams and magnetic structures for GdRu₂Si₂. a,b) H(magnetic field)–T(temperature) magnetic phase diagrams for H∥[001] (a) and H∥[100] (b) determined based on the magnetization measurements. Skl and ML denote the double-Q spin orders representing the square lattice of skyrmion texture and the antiferroic lattice of meron/anti-meron-like textures, respectively. The crystal structure of GdRu₂Si₂ is also shown in the inset. c–h) The schematic illustration of spin texture for the phase I, II, III, III′, IV, and V, identified through the resonant X-ray scattering experiments with polarization analysis. The presented spin textures are based on Equations (3)–(7), with their local amplitude of Gd magnetic moment kept constant. The phases I, II, III, and III′ are the double-Q states described by the superposition of two orthogonally modulated spin textures. The background color represents the amplitude of out-of-plane spin component m².

a function of $H_{[100]}$ and $H_{[001]}$ (i.e., the [100] and [001] components of $H$) at 5 K is summarized in Figure 3j (see Note S2, Supporting Information for the detail). Here, the angle between the $H$-direction and the [001] axis is defined as $\theta$, and the corresponding $\theta$-dependence of magnetization at 3 and 7 T are plotted in Figure 3k,l, respectively. At 3 T, the $M$–$\theta$ profile exhibits a clear step-like anomaly at $\theta = 60^\circ$, which represents the transition between the phases III and IV (Figure 3k). In contrast, the $M$–$\theta$ profile at $\mu_0H = 7$ T shows a smooth behavior without any anomaly (Figure 3l), demonstrating the continuous transformation between the phase III in $H∥[001]$ and the phase III′ (double-Q state) in $H∥[100]$. This observation suggests that the phase III is also a double-Q magnetic state, which is consistent with the recent STM observation of square lattice manner of charge density modulation in the phase III. To summarize, we conclude that the phases I, II, III, and III′ are double-Q states, and that the phases IV and V are single-Q states.

2.2. Magnetic Structure Analysis

To investigate the detailed spin texture in each phase, we have performed the polarization analysis of the scattered X-ray in the RXS experiments. The schematic illustration of the experimental setup is shown in Figure 3c. The magnetic field $H$ is applied perpendicular to the scattering plane. The incident X-ray is po-
lized parallel to the scattering plane (π-polarization). The scattered X-ray beam may consist of two polarization components parallel (π') and perpendicular (σ') to the scattering plane, and their intensities (I_π' and I_σ') are measured separately. When the magnetic structure m(r) includes the modulated spin component mQ exp(iQ·r) + c.c. (with mQ being a complex vector), the magnetic scattering intensity I is given by [26]:

\[ I \propto |\langle e_\pi \cdot e_\sigma \rangle \cdot m_Q|^2 \]  

(2)

Here, e_π and e_σ are unit vectors representing the polarization of incident and scattered X-ray beams, respectively. On the basis of the above relationship, we can separately evaluate the amplitude of each modulated spin component.

In the following, we discuss the case for the double-Q structure in phase III' at H = 5 T applied along the [100] direction, as an example. First, the magnetic satellite peaks at (0, 4, 0) ± Q are investigated with the setup shown in Figure 3a. In this configuration, the propagation vector k of incident X-ray is almost parallel to the [011] direction. Equation (2) suggests that I_{π'} and I_{σ'} mainly reflect the [100] and [011] components of m_Q, respectively. Figure 3d-g indicates the line scan profiles of the I_{π'} and I_{σ'} intensities for Q_1 = (q, 0, 0) and Q_2 = (0, q, 0), respectively. In both cases, I_{σ'} is clearly observed, while I_{π'} is almost negligible. This suggests that m_a and m_b contain [010] or [001] component, but do not have the [100] component. We have further performed a similar polarization analysis for the magnetic reflections at (0, 4, 0) ± Q with the setup shown in Figure 3b, where k is almost parallel to the [010] direction. In this case, I_{π'} and I_{σ'} mainly reflect the [100] and [010] components of m_Q, respectively. Figure 3e-g indicates the line scan profiles of I_{π'} and I_{σ'} for Q_2. The observed presence of I_{π'} and absence of I_{σ'} suggest that m_Q mainly consists of the [010] component. The corresponding line scan profile for Q_3 is also shown in Figure 3h, where the absence of I_{π'} and I_{σ'} intensities demonstrates that m_Q contains neither [010] nor [100] component. Since the aforementioned results for (0, 4, 0) ± Q_1 (Figure 3g) suggest that the projection of m_a along the [011] direction is zero, m_a should possess the [001] component. On the basis of the above results, we conclude that the spin texture in the phase III' for H||[100] can be approximately described as

\[ m(r) \propto [(0, m_2, 0) \exp(iQ_2 \cdot r) + (0, 0, m_1) \exp(iQ_1 \cdot r) + \text{c.c.}] + M_b(1, 0, 0) \]

(3)

with M_b representing the H-induced uniform magnetization component. The resultant anisotropic double-Q spin texture...
Figure 3. Polarization analysis of RXS profiles in the phase III', and magnetic phase diagram as a function of $H_{[100]}$ and $H_{[001]}$. a,b) Experimental configuration of RXS measurement to investigate magnetic satellite peaks around the fundamental Bragg spots (0, 4, 0) and (0, 4, 4), respectively. Scattering plane is always perpendicular to the [100] axis. $k_i$ and $k_f$ are the propagation vectors of incident and scattered X-ray, respectively. In (a) and (b), $k_i$ is almost parallel to the [011] and [010] direction, respectively. c) Schematic illustration of experimental setup for polarization analysis. $\pi$ ($\pi'$) and $\sigma$ ($\sigma'$) are the polarization direction of incident (scattered) X-ray. d,e) Line profiles for ($\delta$, 4, 0) and ($\delta$, 4, 4) scans for $Q_1$ (||H) satellite reflection, measured at 5 K with $\mu_0H = 5$ T applied along the [100] direction (phase III'). g,h) The corresponding line profiles for (0, 4 $-$ $\tau$, 0) and (0, 4 + $\tau$, 4) scans for $Q_2$ (\perp H) satellite reflection. f,i) Real-space illustration of modulated spin components that belong to each magnetic modulation vector $Q_1$ and $Q_2$. j) Magnetic phase diagram as a function of $H_{[100]}$ and $H_{[001]}$ (i.e., the [100] and [001] component of H, respectively), determined from the $M$–$H$ profiles measured for various direction of H at 5 K (Figure S2, Supporting Information). The angle between the H-direction and the [001] axis is defined as $\theta_k$. k,l) $\theta$-dependence of magnetization measured at $\mu_0H = 3$ T (k) and 7 T (l).
is the double-Q state given by
\[
m(r) \propto \left[ (0, m_x, 0) \exp(iQ_1 \cdot r) + (m_y, 0, 0) \exp(iQ_2 \cdot r) + c.c. \right] + M_0(0, 0, 1)
\]
which represents the square vortex lattice state described by the superposition of two sinusoidally modulated spin components (Figure 1e). The phases IV and V induced by \( H \parallel [100] \) are the single-Q states described by
\[
m(r) \propto \left[ (0, m_x, 0) \exp(iQ_1 \cdot r) + c.c. \right] + M_0(1, 0, 0)
\]
and
\[
m(r) \propto \left[ (0, 0, m_y) \exp(iQ_2 \cdot r) + c.c. \right] + M_0(1, 0, 0)
\]
representing the screw spin order with \( Q_1 \parallel H \) (Figure 1g) and the sinusoidally modulated fan-like spin order with \( Q_2 \perp H \) (Figure 1h), respectively. Notably, the experimentally identified \( m_{q_0} \) in the \( H \)-induced phases III, III', IV, V and VI are always normal to the \( H \)-direction (Figure 1e–h). In general, the magnetic moments in the antiferromagnetic and helimagnetic states favor to be aligned perpendicular to the \( H \)-direction, and the observed \( m_{q_0} \parallel H \) relationship is reasonable in this context. At 7 T, the rotation of magnetic field direction from the [001] axis (phase III represented by Equation (5)) to the [100] axis (phase III' represented by Equation (3)) leads to the continuous rotation of \( m_{q_0} \) within the (010) plane keeping the \( m_{q_0} \perp H \) relationship, which well explains the observed smooth \( M-\Theta \) profile in Figure 3l.

2.3. Formation Mechanism of each Spin Texture

Next, we discuss the microscopic mechanism to realize such an intricate magnetic phase diagram with a rich variety of double-Q orders. In the following, we demonstrate that the observed phase diagram and spin textures can be well reproduced by the effective Hamiltonian given by
\[
H = 2N \sum_{q \neq 0} \left[ -J \left( \sum_{q \neq 0, q' \neq 0} \Gamma_{q, q'} m''_{q} m''_{q'} \right) + K \left( \sum_{q \neq 0, q' \neq 0} \Gamma_{q, q'} m''_{q} m''_{q'} \right)^2 \right] - \sum_{i} H \cdot m_i
\]
which is derived from a Kondo lattice model consisting of localized magnetic moments and itinerant electrons. This theoretical framework has originally been proposed in refs. [18, 27], where a set of wavenumbers \( Q_1 = (q \pm 0, 0) \) and \( Q_2 = (0, q \pm 0) \) are assumed to take peaks in the bare susceptibility under the tetragonal lattice symmetry. \( J(\parallel 1) \) represents the amplitude of Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction stabilizing the magnetic modulation with the wavevector \( Q_1 \), and \( K \) represents the amplitude of four-spin interaction favoring multiple-Q orders. \( \Gamma_{Q_0} = (\gamma_x, \gamma_y, \gamma_z) \) and \( \Gamma_{Q_2} = (\gamma_x, \gamma_y, \gamma_z) \) describe the anisotropy of associated interactions originating from the spin-orbit coupling. The last term represents the Zeeman coupling in an external magnetic field \( H \), and \( m_i \) represents the local magnetic moment at the position \( i \) (See Experimental Section for the detailed parameters and procedures for the theoretical simulation.)

Figure 4a shows the magnetization profile for \( H \parallel [001] \) theoretically calculated by Equation (8) with \( K = 0.3 \). It shows a series of step-like anomalies, in good agreement with the experimental results (Figure S2a, Supporting Information). The corresponding \( H \)-dependence of individual \( m''_{Q_0} \) component and the simulated spin texture at selected \( H \)-values are shown in Figure 4b–i, respectively. We have found that the experimentally identified spin textures for phases I (anisotropic double-Q state (Figure 1c)), II (double-Q spin skyrmion lattice state (Figure 1d)), and III (double-Q square vortex lattice state (Figure 1e)), as well as the \( H \)-induced transitions among these phases (i.e., \( I \rightarrow II \rightarrow III \rightarrow FM \) (Figure 1a)), are successfully reproduced in the present simulation. The same behaviors are also confirmed for a wide range of \( K \) parameter of 0.1 < \( K \) < 0.5.

Similar theoretical calculations have also been performed for \( H \parallel [100] \), and the obtained \( H \)-dependence of \( M \) and \( (m''_{Q_0})^2 \), as well as the associated spin textures at selected \( H \)-values, are summarized in Figure 4c–f,j–l. The simulation well reproduces the experimentally identified spin textures for the phases IV (single-Q screw state (Figure 1g)), III' (anisotropic double-Q state (Figure 1f)) and V (single-Q fan state (Figure 1h)), while the manner of \( H \)-induced phase transition depends on the magnitude of \( K \). The present calculation predicts the successive transitions in order of \( I \rightarrow IV \rightarrow V \rightarrow FM \) (Figure 4c) and \( I \rightarrow III' \rightarrow V \rightarrow FM \) (Figure 4e) for \( K = 0.2 \) and \( K = 0.3 \), respectively, which suggests that the free energies for phases III' and IV are almost degenerated. In the experiment, the transition in order of \( I \rightarrow IV \rightarrow III' \rightarrow V \rightarrow FM \) is observed (Figure 1b) in accord with theoretical prediction, implying that the fine tuning of \( K \) value and/or magnetic anisotropy would allow the full reproduction of the experimental phase transition process for \( H \parallel [100] \). The above results suggest that Equation (8) well captures the physics behind the skyrmion formation and the related multiple-Q magnetism in this system.

To understand the topological aspect of each spin texture, their corresponding spatial distribution of topological charge density \( n_{q_0}(r) \) are plotted in Figure 4m–r. The double-Q phases I, II, and III are characterized by the common texture of in-plane spin component (Figure 4g–i), which possesses two vortices and two anti-vortices in the magnetic unit cell. By further considering the out-of-plane spin component, they can be considered as a periodic lattice of meron and anti-meron-like texture, where magnetic moments in meron and anti-meron wrap a half of unit sphere and are characterized by \( N_{sk} = -1/2 \) and \( N_{sk} = +1/2 \), respectively.[29–31] According to Equation (1), the sign of \( N_{sk} \) is affected by the orientation of out-of-plane spin component at their core positions and the swirling manner of in-plane spin component in the surrounding area. For example, the magnetic unit cell of the phase II consists of a core-down vortex (i.e., a meron with \( N_{sk} = -1/2 \)), a core-up vortex (i.e., an anti-meron with \( N_{sk} = +1/2 \) and two core-up anti-vortices (i.e., two merons), which can be viewed as the square lattice of skyrmion (\( N_{sk} = -1 \) (Figure 4n). On the other hand, the phases I and III are characterized by the different orientation of core magnetization, and can be considered as the antiferroic order of meron/anti-meron-like
Figure 4. Numerical simulation of magnetic phase diagram and magnetic structures. a,b) Magnetic field dependence of magnetization $M$ and modulated spin component $(m_{Q_1}^2 + m_{Q_2}^2)^2$, theoretically calculated by simulated annealing for $H || [001]$ with Equation (8) and $K = 0.3$. Here, the uniform ferromagnetic (FM) state gives $M \approx 1$, and we defined $(m_{Q_1}^2 + m_{Q_2}^2)^2 \equiv (m_{xQ_1}^2 + m_{yQ_1}^2)^2$. The corresponding results for $H || [100]$ calculated with $K = 0.2$ (c) and (d) and $K = 0.3$ (e) and (f). c–f) The real space distribution of local magnetization $m(r)$ (g–l) and topological charge density $n_{sk}(r)$ (m–r) as well as (s–x) reciprocal-space distribution of $|m_Q|$ for the phase I, II, III, IV, III', and V obtained by simulated annealing. The background color represents the amplitude of $m_z$, $n_{sk}$ and $|m_Q|$. The results for the single-Q state are obtained by averaging over 500 Monte Carlo samplings to reduce the effect of thermal fluctuations.

textures into the stripe and checkerboard patterns, respectively (Figure 4m,o). Likewise, the double-Q phase III' can be also interpreted as the checkerboard antiferroic order of meron/antimeron-like textures (Figure 4q). Here, the phases III and III' smoothly transform into each other by rotating magnetic field, keeping the $m_{Q} \bot H$ relationship. Since $n_{sk}(r)$ is invariant under the global rotation of magnetic moments according to Equation (1), these two phases are characterized by the common spatial distribution of $n_{sk}$ as shown in Figure 4o,q.

In general, conduction electrons interacting with such non-coplanar magnetic textures feel the local emergent magnetic field $b_{em}^z(r) = (h/e)n_{sk}(r)$ due to the additional quantum Berry phase. In case of GdRu$_2$Si$_2$ with magnetic modulation period $\lambda \approx 1.9$ nm, the amplitude of $b_{em}^z(r)$ should be in order of $\approx 100$ T. The above analysis suggests that the presently identified double-Q phases are characterized by the periodic modulation of giant emergent magnetic field $b_{em}^z(r)$, which will lead to the exotic manner of local electron dynamics (such as meandering motion of electrons). Note that $n_{sk}(r)$ is always negligible for the phase IV and V with single-Q character (Figure 4p,r), suggesting that the multiple-Q nature is essential for the appearance of local topological charge and associated emergent magnetic field.

In Figure 4s–x, we plot theoretically calculated reciprocal-space distribution of $|m_Q|$ for each phase. They always exhibit peak structures corresponding to $Q_1$ and $Q_2$. On the other hand, the higher order peak at the $Q_1 + Q_2$ position can be identified.
only in phase II (square skyrmion lattice phase), but not in the other double-Q phases. Since the magnetic scattering intensity in the diffraction experiment is generally proportional to $|m_{\sigma}|^2$, the absence of $Q_1 + Q_2$ reflection in phases I and III reported in ref. [13] would be reasonable.

3. Conclusion

In this study, we have identified a variety of double-Q spin textures (including the antiferroic order of meron/antimeron-like textures with fractional local topological charge) for a prototype skyrmion-hosting centrosymmetric tetragonal magnet GdRu$_2$Si$_2$. The observed good agreement between the experimental and theoretical phase diagrams suggests a potential mechanism for the skyrmion formation in this system, where the magnetic interactions mediated by itinerant electrons and the associated magnetic anisotropy play a significant role. Note that several alternative theoretical frameworks based on magnetic frustration has also been proposed recently, some of which may be effectively mapped into the present model and possibly reproduce the observed phase diagrams by tuning their parameters.[22,23] These novel theoretical frameworks predict the appearance of numerous multiple-Q orders consisting of various topological solitons such as merons, skyrmions and higher-order skyrmions with $N_{\sigma} = \pm 1/2$, 1, and 2, respectively, depending on the symmetry of underlying crystal lattice and magnetic anisotropy.[16–20,27] Previously, such multiple-Q spin textures with fractional or higher-order topological charge have rarely been identified experimentally.[29,30] The present results glimpse how rich variety of multiple-Q orders with unique topology/symmetry can be derived from a simple crystal lattice system with itinerant electrons, and highlight rare-earth intermetallics as a promising platform for the further search of exotic topological soliton ensembles with nontrivial functionality. The direct real-space observation of these textures and full theoretical reproduction of finite temperature phase diagram are the issue for the future study.

4. Experimental Section

Crystal Growth: Single crystals of GdRu$_2$Si$_2$ were grown by the optical floating zone method as reported in ref. [13]. The phase purity of the sample was confirmed by the powder X-ray diffraction. The orientations of single crystals were determined using the back-reflection X-ray Laue photography method.

Magnetic and Electrical Transport Property Measurements: Magnetization measurements were performed using a superconducting quantum interference device magnetometer (MPMS, Quantum Design), which was accompanied with a rotation probe to characterize the angle dependence of magnetization. A single crystal of GdRu$_2$Si$_2$ was cut into a rectangular plate shape and silver paste was painted as electrodes for five terminal plate shape anisotropy.

Monochromator. A single crystal of GdRu$_2$Si$_2$ with flat (010) plane was fixed on an Al plate using varnish and put into a vertical-field superconducting magnet. In this setup, the scattering plane $(0, K, L)$ was perpendicular to the $[100]$ axis. The incident X-ray was linearly polarized parallel to the scattering plane $(\sigma)$. To analyze the polarization of the scattered X-ray, the (006) reflection of a pyrolytic graphite (PG) plate was used. The $2\theta$ angle for the analyzer at Gd $L_3$ edge was 88°. The polarization components $\sigma_\parallel$ (perpendicular to the scattering plane) and $\sigma_\perp$ (parallel to the scattering plane) of scattered X-ray beams can be chosen by rotating the PG plate about the scattered beam.

Theoretical Calculation: The results in Figure 4a–f were obtained by performing simulated annealing for the model in Equation (8). In the simulations, starting from a random spin configuration, the temperature was gradually reduced from high temperature to the final temperature $T = 0.01$ with the rate $T_n + 3 = a T_n$, where $T_n$ was the temperature in the nth step and $a$ was set to be 0.99995–0.99999. The standard Metropolis local updates may be effectively mapped into the present model and pose- tionally frustrated has also been proposed recently, some of which the final temperature. To determine the phase boundaries between different magnetic phases, the simulations were also performed from the spin patterns obtained at low temperatures.

The magnetic phases in Figure 4 are calculated for the model parameters $q = x/3$, $\gamma = 0.855$, $\gamma_0 = 0.5$, $\gamma_1 = 1$, and $K = 0.3$ (Figure 4a,b,e,f) and $K = 0.2$ (Figure 4c,d) in the system size with $N = 96^2$. In the case of $\text{H}[\langle 001 \rangle]$, three distinct phases were obtained in addition to the fully-polarized state at high fields; phase I for $0 < H \leq 0.6$, phase II for $0.6 < H \leq 0.7$, and phase III for $0.7 < H \leq 1.8$. On the other hand, three phases were stabilized in the case of $\text{H}[\langle 100 \rangle]$; phase I for $0 < H \leq 0.55$, phase IV for $0.55 < H \leq 1.6$, and phase V for $1.6 < H \leq 2$ at $K = 0.2$ and phase I for $H \leq 0.5$, phase III for $0.5 < H \leq 1.6$, and phase V for $1.6 < H \leq 2$ at $K = 0.3$. Note that the similar low-temperature phase sequence against magnetic field $H$ was obtained to reproduce that in experiments in the wide range of model parameters $K$ and magnetic anisotropy, as detailed in ref. [27].

In Figure 4g–l, the spin textures for the phases I, II, III, IV, III′, and V were calculated with ($H_{\langle 100 \rangle}$, $H_{\langle 001 \rangle}$) = (0, 0.1, 0.3), (0.65, 0.3), (0, 1.0, 0.3), (1.0, 0, 0.2), (1.0, 0, 0.3), and (1.8, 0, 0.3), respectively. For Figure 4m–r, the topological charge density $n_m(r)$ was calculated following the procedure in refs. [27, 34].

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.
Author Contributions

S.S., T.A., and Y.T. supervised the project. N.D.K. grew samples and measured the magnetic and transport properties with the assistance of R.T. RXS measurements were carried out by T.N., S.G., and N.D.K., with the assistance of K.K., Y.Y., H.S., and H.N. S.H. and Y.M. performed Monte Carlo simulation. N.D.K., S.S., and S.H. wrote the manuscript with inputs from co-authors. All the authors discussed the results and commented on the manuscript.

Data Availability Statement

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

Keywords

centrosymmetric, intermetallics, magnetism, spintronics, topological spin textures

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