# Metastability of the Skyrmion Lattice in $GdRu_2Si_2$ Revealed by Ultrasound

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We report on the low-temperature magnetoelastic properties of the skyrmion-lattice compound $GdRu_2Si_2$ under magnetic fields applied along the [001] axis. Our sound-velocity measurements reveal a series of pronounced anomalies related to the skyrmion-lattice phase. The emergence of hysteresis and steplike features indicates intrinsic metastability of the skyrmion lattice in $GdRu_2Si_2$, deviating from other skyrmion-lattice compounds. Analysis of multiple elastic modes allows us to disentangle the compressional and shear responses, with the latter showing a stronger sensitivity to the metastability of the skyrmion lattice.

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Magnetic skyrmions are nanometric, vortexlike spin structures that represent topological defects of the magnetization. Their existence in solids was theoretically proposed in the 1980s as a result of the interplay between broken inversion symmetry, external magnetic field, and thermal fluctuations [1,2]. Experimental confirmation came much later, with the observation of a skyrmion lattice in the chiral magnet MnSi by means of small-angle neutron scattering [3]. Since then, skyrmions and related topological spin textures have been observed in a wide variety of materials, opening a vibrant research field with promising prospects for spintronic applications [4].

While broken inversion symmetry and the associated Dzyaloshinskii-Moriya interaction (DMI) were long considered to be essential for stabilizing skyrmion lattices, recent discoveries have challenged this view. Skyrmion phases have been observed in centrosymmetric magnets such as $MnSc_2S_4$ [5] and $GdRu_2Si_2$ [6], where they arise instead from frustrated or long-range exchange interactions. Remarkably, in both systems, the skyrmion lattice remains stable down to low temperatures, suggesting that thermal fluctuations, also traditionally viewed as a key stabilizing mechanism, may not play a dominant role in these materials.

Understanding the stabilization mechanisms and physical properties of such unconventional skyrmion phases requires complementary experimental methods. An alternative and powerful approach to small-angle neutron scattering is to use ultrasound, which probes the coupling between magnetic textures and the crystal lattice. This technique, for skyrmion systems first applied to MnSi, revealed pronounced anomalies in the elastic constants associated with the skyrmion phase [7,8]. Furthermore, such measurements show a strong dependence on the propagation and polarization direction of the sound waves with respect to the skyrmion lattice.

In this Letter, we employ ultrasound techniques to investigate the skyrmion phase of $GdRu_2Si_2$ at low temperatures. We show that, unlike MnSi, the elastic properties of the skyrmion lattice in $GdRu_2Si_2$ exhibit a strong variation with temperature. Moreover, we observe pronounced hystereses and evidence of intrinsic skyrmion-lattice metastabilities below 4 K. Previously reported metastable skyrmion states have so far been realized either through nonequilibrium preparation protocols [9,10], or as thermodynamically disconnected from the equilibrium skyrmion state [11]. Here, we show that the equilibrium skyrmion phase of $GdRu_2Si_2$ instead evolves into a manifold of intrinsic metastable states, without crossing a phase boundary or requiring any special preparation protocol.

We grew a single crystal of $GdRu_2Si_2$ (tetragonal $ThCr_2Si_2$ type structure, space group $I4/mmm$) using the floating zone method, as described in more detail in Refs. [12,13]. We selected a crystal of $2.5 \times 2.0 \times 1.5$ mm, with the largest dimension along the [110] axis.

We applied magnetic fields up to 3 T along the [001] axis in a superconducting 20 T magnet from Oxford Instruments equipped with a commercial $^3$He cryostat. The field-sweep rate was set at 0.1 T/min, which allows to precisely resolve steplike features (of typical width around 7 mT) and plateaus in all acoustic modes. Between successive field scans, the sample was heated at least to 2.5 K at zero

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field and then back to the chosen temperature. We performed ultrasound measurements utilizing the transmission pulse-echo technique with a phase-sensitive detection as described in Refs. [14,15]. We investigated four different elastic modes: a longitudinal wave with propagation $\mathbf{k}$ and polarization $\mathbf{u}$ along [001] ($v_0 = 4200 \pm 40$ m/s), another longitudinal mode with $\mathbf{k}\|\mathbf{u}\|[110]$ ($v_0 = 4600 \pm 50$ m/s), and two transverse modes, one with propagation $\mathbf{k}\|[001]$ and polarization $\mathbf{u}\|[110]$ ($v_0 = 2800 \pm 30$ m/s) and another one with $\mathbf{k}\|[110]$ and $\mathbf{u}\|[1\bar{1}0]$ ($v_0 = 2600 \pm 30$ m/s): in all cases, $v_0$, the absolute sound velocity, was determined at zero field and $T = 4$ K. Note that contrary to $v_0$, the resolution for $\Delta v/v = [v(H,T) - v(H_0,T_0)]/v(H_0,T_0)$ is of the order of $10^{-6}$ [14]. We bonded $LiNbO_3$ transducers (36°-Y cut and 41°-X cut for longitudinal and transverse modes, respectively, obtained from Yamaju Ceramics) to the polished surfaces of the single crystal. The transducers were attached using Thiokol LP-32 as a bond. We used ultrasound frequencies between 60 and 90 MHz.

The skyrmion phase of $GdRu_2Si_2$ has been reported in a narrow field window between 2 and 2.4 T below 20 K [6]. In this Letter, we concentrate on the sound-velocity changes below 10 K, both as function of temperature and magnetic field. In Figs. 1(a) and 1(b), we show our results below 1 K for the two longitudinal acoustic modes. We observed very sharp, steplike anomalies as a function of magnetic field, inside the region corresponding to the skyrmion-phase boundaries [6] (gray hatched areas in Fig. 1). The skyrmion phase leads to a hardening (increase of the sound velocity) of all studied acoustic modes compared to the low-field phase. Furthermore, we observe a strong hysteresis, which evidences first-order phase transitions into, as well as out of the skyrmion phase. Remarkably, the sound-velocity changes are quite different between the sweep up and down curves. For decreasing field, we identify a broad elasticity plateau in the skyrmion phase, which extends from 1.88 to 2.10 T at and below 0.5 K. In contrast, for increasing field, we detect two smaller plateaus between 2.12 and 2.20, and between 2.27 and 2.30 T, respectively. Remarkably, there is no overlap in field between these field-up- and field-down-sweep plateaus. Altogether, these results evidence a metastability of the skyrmion lattice.

For the transverse acoustic modes [Figs. 1(c) and 1(d)], We observe pronounced plateaus and step-like anomalies, comparable to our results for the longitudinal modes. The skyrmion phase leads to a hardening of both transverse modes with respect to the low-field phase. However, for the $\mathbf{k}\|[001]$, $\mathbf{u}\|[110]$ geometry [Fig. 1(c)] we do not detect an elastic plateau for deceasing field, but rather two sharp step-like anomalies and a region of constant slope of the sound velocity between 1.88 and 2.10 T. Similar features are visible for the mode with $\mathbf{k}\|[110]$, $\mathbf{u}\|[1\bar{1}0]$ [Fig. 1(d)]. This shows a different response of the skyrmion lattice to compression and shear modes. The overall elastic response of the skyrmion phase is larger for the transverse than the longitudinal acoustic modes.

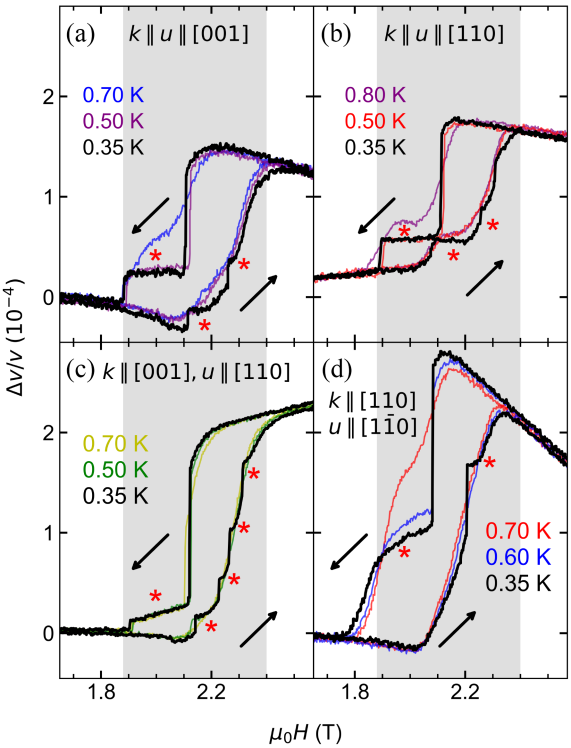



FIG. 1. Sound-velocity changes versus magnetic field at selected temperatures below 0.9 K, for the studied acoustic modes in $GdRu_2Si_2$: (a) $\mathbf{k}\|\mathbf{u}\|[001]$, (b) $\mathbf{k}\|\mathbf{u}\|[110]$, (c) $\mathbf{k}\|[001]$ and $\mathbf{u}\|[110]$, and (d) $\mathbf{k}\|[110]$ and $\mathbf{u}\|[1\bar{1}0]$. Magnetic field was applied along [001]. The skyrmion phase is indicated by the gray area, which we define as the region where up and down sweeps start to differ in (a). These phase boundaries are consistent with the results of Ref. [12] obtained at higher temperatures. The red stars show the different elasticity plateaus discussed in the text.

Furthermore, we observe a strong difference between the two transverse modes for the field-up sweeps at 0.35 K: We detect four elasticity plateaus for $\mathbf{k}\|[001]$, $\mathbf{u}\|[110]$, and only one for $\mathbf{k}\|[110]$, $\mathbf{u}\|[1\bar{1}0]$. The appearance of multiple elasticity plateaus might correspond to the devil's staircase scenario, as discussed in the following section.

Above 0.9 K, the hystereses in the longitudinal modes is strongly reduced [Figs. 2(a) and 2(b)]. Furthermore, for the mode with $\mathbf{k}\|\mathbf{u}\|[110]$, a small softening inside the skyrmion phase starts to develop above 2 K. This evidences that the elastic properties of the skyrmion phase in $GdRu_2Si_2$ are strongly sensitive to temperature. For the transverse modes, we observe the same trend with a closing of the hysteresis loop above 0.9 K [Figs. 2(c) and 2(d)].

In order to emphasize the hysteretic behavior of the sound velocity in the skyrmion phase at low temperatures, we show $H - T$ color plots obtained from the derivatives of our sound-velocity data of the longitudinal acoustic mode with $\mathbf{k}\|\mathbf{u}\|[001]$ in Figs. 3(a) and 3(b), and from the difference between the sound-velocity data for up and down field sweeps in Fig. 3(c). Since the skyrmion phase

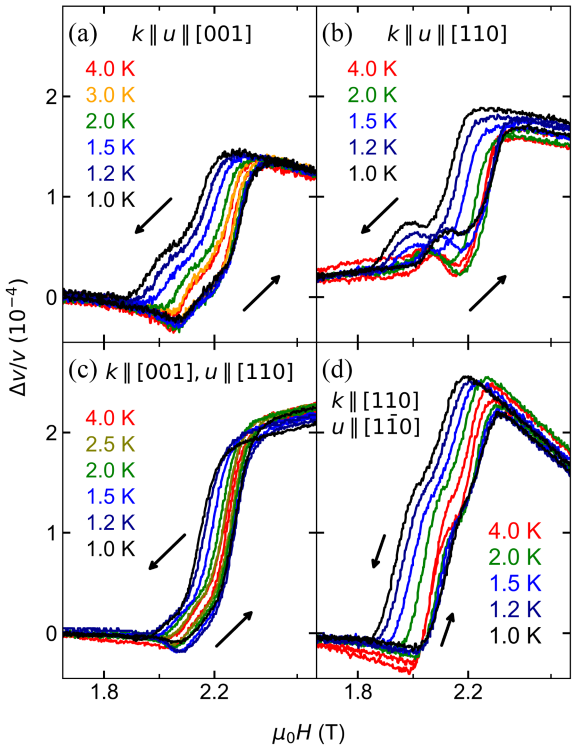



FIG. 2. Sound-velocity changes versus magnetic field at selected temperatures above 0.9 K, for the studied acoustic modes in $GdRu_2Si_2$: (a) $\mathbf{k}\|\mathbf{u}\|[001]$, (b) $\mathbf{k}\|\mathbf{u}\|[110]$, (c) $\mathbf{k}\|[001]$ and $\mathbf{u}\|[110]$, and (d) $\mathbf{k}\|[110]$ and $\mathbf{u}\|[1\bar{1}0]$. Magnetic field was applied along [001].

results in jumps in the sound velocity with respect to magnetic field, the derivatives of the curves show sharp peaks, which allows us to identify easily the skyrmion-phase boundaries [Figs. 3(a) and 3(b)]. In particular, the anomalies coincide for up [Fig. 3(a)] and down sweeps [Fig. 3(b)] above 4 K. Below 4 K, the sharp sound-velocity steps appear at largely different fields for up and down sweeps. This relates to the large hystereis developing below 4 K, as shown in the difference between the up- and down-sweep data [Fig. 3(c)].

Finally, we highlight the hysteretic behavior of $GdRu_2Si_2$ at low temperatures by showing sound-velocity data at constant magnetic fields in Fig. 4. The measurements were performed under zero-field-cooled (ZFC) and field-cooled (FC) conditions. The zero-field-cooled curves were obtained upon warming up the sample at a rate of 0.2 K/ min, and the field-cooled ones were measured upon cooling the sample at the same temperature-change rate. At zero field, we observe a hardening of all elastic modes down to 1 K, followed by a slight softening at lower temperatures. The total variation of the sound velocity between 0.35 and 5 K is approximately $2 \times 10^{-5}$, almost 1 order of magnitude smaller than the field-induced changes within the skyrmion phase. For all elastic modes, we detect a difference between the ZFC and FCC procedures developing below 2 K and at 2.1 and 2.3 T. These features are associated with the hysteretic behavior of the skyrmion phase at low temperature.

Our ultrasound results reveal a pronounced hysteresis at the skyrmion-lattice phase of $GdRu_2Si_2$ below 4 K. Furthermore, the different sound-velocity plateaus for up and down sweeps evidence intrinsic metastabilities inside the skyrmion phase. This feature contrasts with the archetypical skyrmion system MnSi, where the skyrmion lattice is stabilized by the interplay between Dzyaloshinskii-Moriya interactions (DMI) and thermal fluctuations, and does not show any significant hysteresis [16]. In MnSi, both longitudinal and transverse acoustic modes display sharp discontinuities at the skyrmion-phase boundaries [8], in accord with a thermally stabilized, equilibrated skyrmion crystal.

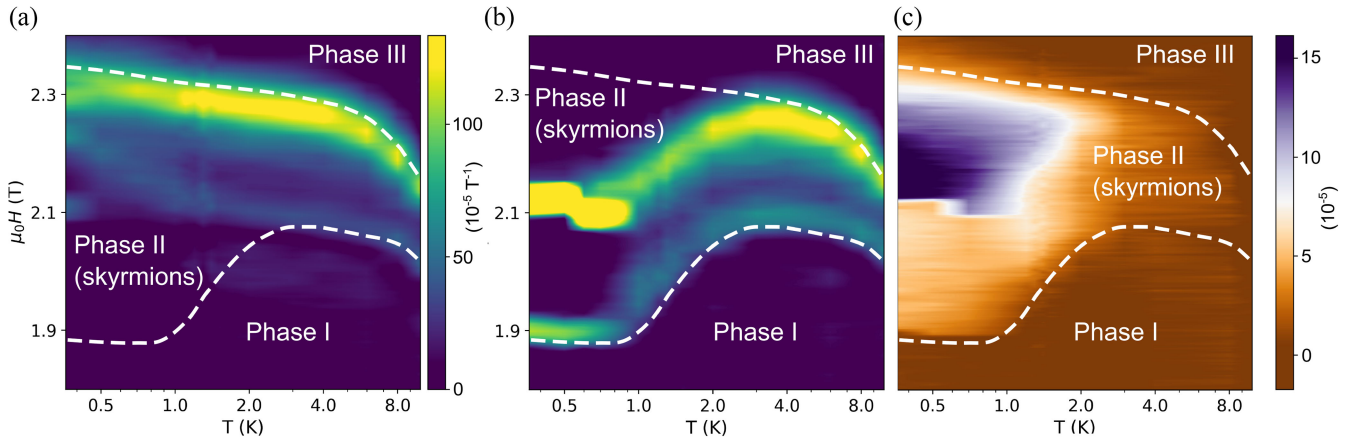



FIG. 3. $H-T$ semilogarithmic color plot of the sound-velocity derivative with respect to magnetic field for (a) up- and (b) down-field sweeps. (c) Difference between the sound-velocity changes for up- and down-field sweeps. All data are obtained from the longitudinal acoustic mode with $\mathbf{k}\|\mathbf{u}\|H\|[001]$ in $GdRu_2Si_2$ [Figs. 1(a) and 2(a)]. The white dashed lines mark the skyrmion-phase boundaries. The labeling of phases is taken from Ref. [12].

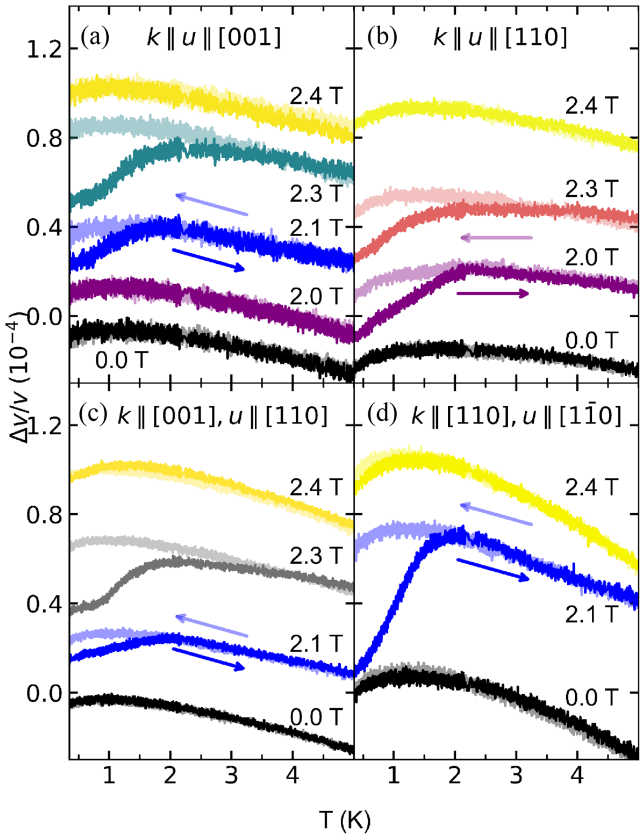



FIG. 4. Sound-velocity changes versus temperature at selected magnetic fields, for the studied elastic modes: (a) $\mathbf{k}\|\mathbf{u}\|[001]$, (b) $\mathbf{k}\|\mathbf{u}\|[110]$, (c) $\mathbf{k}\|[001]$ and $\mathbf{u}\|[110]$, and (d) $\mathbf{k}\|[110]$ and $\mathbf{u}\|[1\bar{1}0]$. The measurement were performed under zero-field-cooled (darker colored) and field cooled (lighter colored) conditions. The magnetic field was aligned along [001]. The curves are shifted along $y$-axis for clarity.

A closer analogy to our observations may be found in $Cu_2OSeO_3$, another magnet with a DMI-driven skyrmion lattice. At high temperatures, the skyrmion phase of $Cu_2OSeO_3$ behaves similarly to that of MnSi: it is free of hysteresis and metastability [17]. However, $Cu_2OSeO_3$ hosts a second, low-temperature skyrmion phase, stabilized by a strong spin-orbit induced magnetocrystalline anisotropy [11]. The high-temperature skyrmion phase of $Cu_2OSeO_3$ can also be extended towards low temperatures by quenching [9,10]. At low temperatures, all skyrmion phases of $Cu_2OSeO_3$ are highly hysteretic, and only accessed through specific cooling protocols [9–11]. They can be further stabilized by tailoring the chemical composition [18] or the sample's shape [19]. Moreover, the authors of Ref. [11] suggest a coexistence of two distinct stabilization mechanisms for the skyrmion lattice: one driven by the DMI interactions and thermal fluctuations, another one by magnetic anisotropy and frustration.

The latter mechanism appears relevant to centrosymmetric magnets such as $MnSc_2S_4$, where skyrmions arise from the competition between magnetic anisotropy and frustrated exchange interactions [5]. In this compound, ultrasound measurements also reveal a strong hysteresis upon entering the skyrmion phase at low temperature [20]. However, in $MnSc_2S_4$ the elastic constants show the same plateau for up and down sweeps, unlike $GdRu_2Si_2$, where the elastic response is strongly asymmetric. This difference suggests that while magnetic frustration likely contributes to the hysteresis in $GdRu_2Si_2$, this alone cannot account for the observed metastabilities in the skyrmion phase.

Recent work on the closely related material $GdRu_2Ge_2$ [21] revealed multiple field-induced skyrmion phases, pointing to a broader trend among rare-earth-based metallic skyrmion compounds. In these systems, the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between pairs of localized moments arises from the interplay between localized and conduction electrons, and which usually is long range [22–24]. Theoretical studies of itinerant magnets with RKKY interactions predict the emergence of competing multi-$Q$ textures and metastable skyrmion lattices [25,26]. Our results, therefore, support a picture in which the interplay between localized and itinerant electrons in RKKY-mediated systems gives rise to a complex energy landscape hosting multiple metastable skyrmion configurations in $GdRu_2Si_2$.

Furthermore, the emergence of multiple elasticity plateaus in $GdRu_2Si_2$ suggests the possible realization of a skyrmion analog of the devil's staircase. Originally discussed for the collinear magnet CeSb [27], the devil's staircase denotes a sequence of commensurate magnetic states stabilized by the interplay between frustration, fluctuations, and magnetic anisotropy. This cascade of nearly degenerate phases gives rise to a series of weakly first-order transitions, manifested as sharp anomalies in the specific heat [28]. Signatures of such states can also be observed in ultrasound experiments [29]. Thus, the appearance of devil's-staircase behavior and of skyrmion lattices share common prerequisites such as competing interactions, magnetic frustration, and anisotropy. Skyrmion-like analogs of the devil's staircase have indeed been predicted in frustrated magnetic films [30]. An alternative scenario might be the formation of a bubble crystal, a collinear modulated phase that exhibits devil's-staircase characteristics and competes with skyrmion states when the easy-axis anisotropy is sufficiently strong [31]. Such competition between collinear, staircaselike structures and skyrmion lattices has recently been reported in Fe/Gd thin films [32]. Consequently, such a bubble-crystal phase may emerge in $GdRu_2Si_2$ at low temperatures, providing a consistent explanation for our observations. Furthermore, the evolution from a skyrmion phase at high temperature to a bubble-crystal phase at low temperature suggests an increase of the magnetic anisotropy at lower temperature, also in agreement with theory [26].

Finally, our results require a closer examination of the magnetoelastic coupling in metallic skyrmion compounds.

For $GdRu_2Si_2$, the absence of single-ion anistropy for the Gd ions suggests that the elastic properties of the skyrmion lattice might be related to the coupling with itinerant electrons. While exchange-striction-like contributions can be derived from the RKKY interactions [33], the magnetoelastic coupling arising from higher-order RKKY processes and the resulting effective magnetic anisotropy discussed in Refs. [25,26] remain largely unexplored.

In summary, we have investigated the low-temperature magnetoelastic properties of the skyrmion phase in $GdRu_2Si_2$ using ultrasound techniques. Our measurements reveal a series of pronounced anomalies in the sound velocity with a strongly hysteretic behavior developing below 4 K. Furthermore, we observed a significant asymmetry of the elastic response in increasing and descreasing magnetic fields, which suggests the presence of multiple metastable skyrmion configurations depending on the magnetic-field history. Moreover, our observation of a sequence of elastic plateaus for the transverse mode with $\mathbf{k}\|[001]$ and $\mathbf{u}\|[110]$ suggests devil's-staircase behavior, which has been discussed for skyrmion thin films [30]. By analyzing different acoustic modes, we identify distinct responses of the compression and shear acoustic waves, showing a stronger coupling between the metastable skyrmion states and the transverse acoustic modes.

Furthermore, within the skyrmion phase the size of the sound-velocity jumps and plateaus evolve significantly with temperature, in sharp contrast to the behavior observed in Dzyaloshinskii-Moriya magnets such as MnSi and $Cu_2OSeO_3$, where the skyrmion phases are largely reversible and thermally stabilized. These findings indicate that in RKKY systems, such as $GdRu_2Si_2$, skyrmion lattices can persist and reorganize even in the absence of strong thermal fluctuations.

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*Data availability*—The data that support the findings in this article are available from the authors upon reasonable request.

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