Quantum topological phase transitions in skyrmion crystals

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Topological order is important in many aspects of condensed matter physics, and has been extended to bosonic systems. In this Letter we report on the nontrivial topology of the magnon bands in two distinct quantum skyrmion crystals appearing in zero external magnetic field. This is revealed by nonzero Chern numbers for some of the bands. As a bosonic analog of the quantum anomalous Hall effect, we show that topological magnons can appear in skyrmion crystals without explicitly breaking time-reversal symmetry with an external magnetic field. By tuning the value of the easy-axis anisotropy at zero temperature, we find eight quantum topological phase transitions signaled by discontinuous jumps in certain Chern numbers. We connect these quantum topological phase transitions to gaps closing and reopening between magnon bands.

Introduction. Topological order in fermionic condensed matter systems lies at the heart of the understanding of the quantum Hall effect (QHE) [1, 2], the quantum anomalous Hall effect (QAHE) [3, 4], the quantum spin Hall effect (QSHE) [5], and topological insulators (TIs) [6]. The QHE involves explicit time-reversal symmetry breaking by an external magnetic field, while the QSHE and TIs are found in time-reversal-symmetric systems [5–8]. The QAHE is a special case, taking place in systems where time-reversal symmetry is spontaneously, and not explicitly, broken. It is thus a manifestation of the QHE without the need for an external magnetic field [4]. It was later shown that also bosonic excitations may feature topological properties [9–12]. The collective fluctuations of quantum spins, i.e., magnons, have been shown to be topologically nontrivial in magnonic crystals [9], dipolar magnetic thin films [10], and in ferromagnets on the honeycomb lattice [13–17]. In the model used in Refs. [13–15], next-nearest neighbor Dzyaloshinskii-Moriya interaction (DMI) realizes a bosonic analog of the Haldane model [3] in a system of insulating spins.

Even though analogies were proposed [13, 17–20], topological magnon systems do not show direct equivalences of the QHE, QAHE, and QSHE [5, 6, 15]. Since bosons obey Bose-Einstein rather than Fermi-Dirac statistics, the Hall conductivity is not quantized [15]. The authors of Ref. [11] introduced the bosonic analog of a TI, a topological magnon insulator. The nontrivial topology of the magnon bands gives rise to chiral edge states within a bulk magnon gap. However, since bosonic systems lack the concept of a Fermi surface, the bulk is not guaranteed to be insulating with respect to spin currents [14]. Still, the chiral edge states resulting from topologically nontrivial magnons allow the creation of magnon currents that are insensitive to backscattering and disorder [9, 10]. These hold promising applications such as spin-current splitters, waveguides, and interferometers [9, 12]. In addition, magnetoelastic coupling leads to chiral phonon transport induced by the topological magnons [15].

The nontrivial real-space magnetic texture of skyrmions means they are topologically protected [21, 22]. Therefore, skyrmions received a great deal of interest, and are being explored for applications in magnetic memory technology, unconventional computing, and numerous other applications [22–35]. The reciprocal space topology held by the magnon bands in skyrmion crystals (SkXs) has also been explored [36–40], and a topological phase transition driven by a magnetic field was found in Ref. [38]. Furthermore, evidence of the nontrivial topology of magnon bands in SkXs was observed in an experiment [40].

In Ref. [41], we explored the quantum fluctuations of the order parameter for quantum SkXs. Quantum skyrmions are skyrmions with such a small size that the continuum limit breaks down, and the quantum nature of the individual spins is not negligible [41–43]. In this Letter we reveal eight quantum topological phase transitions (QTPTs) [7, 44–46] driven by a tunable easy-axis anisotropy in the same quantum SkXs that are explored in Ref. [41]. These QTPTs are signaled by discontinuous jumps in the Chern numbers [9] of the magnon bands. Here, we consider QTPTs to be topological phase transitions occurring at zero temperature by tuning a parameter in the Hamiltonian. The SkXs we consider are inspired by the observation of a SkX containing nanometer-sized skyrmions in a magnetic monolayer [47]. Since the SkXs are stabilized in zero external magnetic field [41, 47], the QTPTs occur in a time-reversal-symmetric model. Instead, the magnetic order of the SkX ground state (GS) spontaneously breaks time-reversal symmetry, allowing nonzero Chern numbers [12]. In that sense, our skyrmion system is analogous to the QAHE in fermionic systems. This is in contrast to previous studies of topological magnons in SkXs, where time-reversal symmetry is explicitly broken by external magnetic fields [36–40].

As pointed out in Ref. [37], the bulk-edge correspondence in not guaranteed unless the finite geometry contains an integer number of unit cells. However, by letting the GS adapt to a strip geometry, the authors of Refs. [36–38] found the expected number and chirality [9] of edge states based on the bulk Chern numbers in SkXs. Therefore, we will not explicitly prove the existence of chiral edge states here. Assuming the validity of the bulk-edge correspondence in a finite geometry, the QTPTs could be used to switch chiral edge states on and off.

Model. As in Ref. [41], we use the time-reversal-symmetric Hamiltonian

\[ H = H_{ex} + H_{DM} + H_A + H_4, \]  

(1)
The spin operator to site $D_{ij}$, acting between four sites that are oriented counterclockwise and make diamonds of minimal area [47, 50]. The reduced Planck’s constant $\hbar$ and the lattice constant $a$ are set to 1.

We refer to Refs. [41, 51] for details of the two distinct SkX GSs, which are separated by a quantum phase transition (QPT) at $K = K_t$ with $K_t/J \in (0.518, 0.519)$. The classical GSs of SkX1 and SkX2 are shown in Fig. 1(a,e) for $K/J = 0.518$ and $K/J = 0.519$, respectively. By including quantum corrections in a calculation of the expectation value of the Hamiltonian, it is found that $\langle H \rangle$ is lower than the classical GS energy. Hence, quantum skyrmions are energetically preferred over their corresponding classical GSs. [41, 51]. The introduction of the Holstein-Primakoff transformation via rotated coordinates [52] involves approximations whose validity are discussed in Refs. [41, 51]. Possible corrections to our predictions due to the ignored magnon-magnon interactions are discussed in Refs. [12, 37]. Dipolar interactions were shown to affect the high-energy magnon bands of SkXs in Ref. [39]. Since we consider a magnetic monolayer, dipolar interactions are not expected to have significant effects.
The study of dipolar interactions is also beyond the scope of this Letter. For these reasons, they are excluded from our model.

The diagonalization of the system to obtain the magnon bands [53] is shown in detail in Refs. [41, 51]. We take the transformation matrices $T_k$ and the magnon bands $E_{k,n}$ as inputs in this Letter. The 15 magnon bands are numbered from top to bottom in terms of energy. The first Brillouin zone (IBZ) is the same for all 15 sublattices in both SkX1 and SkX2 and we define the points $\Gamma = (0,0)$, $X = (2\pi/135,0)$, $M = (\pi/3,\pi/3\sqrt{3})$, $S = (2\pi/135,2\pi/3\sqrt{3})$, and $Y = (0,-2\pi/3\sqrt{3})$ in the IBZ [41, 51]. These points and the IBZ are sketched in Fig. 1.

**Chern numbers.** Let $\Gamma_n$ be a matrix whose $n$th diagonal element is 1 and all other matrix elements are zero, $\Gamma_n,_{i,j} = \delta_{ni}\delta_{jn}$. From this, we define a projection matrix $P_{k,n} = T_k^{-1}\Gamma_n T_k$. The bosonic nature of the magnons is encoded in the paraunitary transformation matrix [41, 51, 53]. Then the Berry curvature of the $n$th band is given by [2, 9]

$$B_n(k) = i\epsilon_{\mu\nu} \text{Tr}[\delta_{k,u}P_{k,n}(\delta_{k,v}P_{k,n})],$$

where $\epsilon_{\mu\nu}$ is the Levi-Civita tensor and $\mu, \nu \in \{x, y\}$. The Chern number of the $n$th band is its Berry curvature integrated over the IBZ

$$C_n = \frac{1}{2\pi} \int_{\text{IBZ}} dk B_n(k).$$

It can be shown that the Chern numbers are integers, given that the bands are isolated [9]. Here, we calculate the Chern numbers using numerical approximations to the integral. We consider equally spaced discretizations and adaptive quadratures [51, 54]. When the numerical results are found to approach integers upon increasing the density of $k$ points, we present the Chern numbers as integers.

**Quantum topological phase transitions.** In Fig. 1(b,c) we show the magnon spectrum in SkX1 for $K = 0$ and for $K/J = 0.518$, i.e., close to the QPT to SkX2. The Chern numbers of the 15 bands are given at both values of $K$ and it is clear that some of them have changed due to the change in easy-axis anisotropy. In Fig. 1(d) we plot these as a function of $K$, revealing four QPTs. $E_{k,4}$ and $E_{k,5}$ first cross at the $\Gamma$ point for the specific value $K = K_1$, where $K_1/J$ is in the interval $(0.20,0.21)$. They also cross at the $\Gamma$ point for $K/J = K_2/J \in (0.22,0.23)$. The gap between the bands closes and reopens, and their Chern numbers change, signaling QPTs. $E_{k,1}$ and $E_{k,2}$ cross at the $Y$ point for $K/J = K_3/J \in (0.26,0.27)$. The two Chern numbers annihilate, and both bands are topologically trivial for $K > K_4$. Finally, the gap between $E_{k,8}$ and $E_{k,9}$ closes between the $\Gamma$ point and the $X$ point for $K/J = K_4/J \in (0.28,0.29)$. Only $E_{k,9}$ remains topologically nontrivial when the gap reopens for $K > K_4$. In SkX1, the magnon band with lowest energy is topologically trivial, while the band with second lowest energy is topologically nontrivial. For ferromagnetic SkXs in an external magnetic field, the band with third lowest energy is topologically nontrivial while the two bands with lower energy are topologically trivial [36, 38–40].

Fig. 1(e,f) shows the magnon spectrum in SkX2 for $K/J = 0.519$ and $K/J = 0.85$. Despite the plethora of closely avoided crossings, all the bands are isolated at these values of $K$, and all 15 Chern numbers are well defined. Notice that in all cases the sum of the Chern numbers of all bands is zero, as expected [9]. It is clear that the Chern numbers have changed from the spectrum of SkX1 at $K/J = 0.518$ to the spectrum of SkX2 at $K/J = 0.519$. We do not view this as a QPT, since it is not due to gaps closing and reopening in the magnon spectrum. Rather, the magnon spectra are different from the outset, since they arise from two distinct SkXs.

In Fig. 1(g) we plot the Chern numbers that change when tuning $K$ in SkX2. We find four QPTs. The gap between $E_{k,9}$ and $E_{k,10}$ closes between $\Gamma$ and $Y$ for $K/J = K_5/J \in (0.61,0.62)$. Once the gap reopens, $E_{k,9}$ has become topologically nontrivial, while $E_{k,10}$ has become topologically trivial, with $C_{10} = -1$ has jumped to $C_{10} = -2$. $E_{k,10}$ and $E_{k,11}$ cross between $\Gamma$ and $Y$ for $K/J = K_6/J \in (0.63,0.64)$. $C_{10}$ jumps back to $-1$, allowing $E_{k,11}$ to become topologically nontrivial for $K > K_6$. The gap between $E_{k,3}$ and $E_{k,4}$ closes between $\Gamma$ and $Y$ for $K/J = K_7/J \in (0.71,0.72)$. Once the gap reopens at $K > K_7$, they are both topologically trivial. Finally, $E_{k,8}$ and $E_{k,9}$ cross at $k \approx (0.32,0.10)$ for $K/J = K_8/J \in (0.79,0.80)$ and are left topologically trivial for $K > K_8$.

**Gap closing and Berry curvature.** In Fig. 2(a) we go into detail of the two band crossings of $E_{k,4}$ and $E_{k,5}$ in SkX1. For $K < K_1$ we have $C_4 = -2, C_5 = 2$. Then, at $K = K_1$ the gap between $E_{k,4}$ and $E_{k,5}$ at the $Y$ point closes, $\Delta_{4,5} = 0$, and the two Chern numbers are undefined [9]. For $K > K_1$ the gap reopens and $C_4 = -1, C_5 = 1$, i.e., the bands remain topologically nontrivial. For $K = K_2$ the gap closes at the $\Gamma$ point, $\Delta_{1,5} = 0$. When the gap reopens for $K > K_2$ both bands have become topologically trivial. The dependence of these two gaps on the easy-axis anisotropy is shown in Fig. 2(b). It appears the two gaps close with an approximately linear dependence on $K$.

The Berry curvatures of $E_{k,4}$ and $E_{k,5}$ are shown in Fig. 2(c). At $K = 0, B_4(k)$ has extended negative valleys, giving rise to a negative Chern number. For $K/J = 0.22, \Delta_{4,5} = 0, B_4(k)$ is small and so there is a sharp negative valley in $B_4(k)$ around $k = \Gamma$. Again, this gives rise to a negative Chern number. At $K/J = 0.3$, the Berry curvature contains both positive peaks and negative valleys, which cancel each other out in the integral and lead to zero Chern number. The arguments are similar for $B_5(k)$ and $C_5$. From these figures, it is clear that the gap closings involve an exchange of Berry curvature between the bands. Also, the Berry curvature of a given band has its largest absolute values where the band has the smallest gap to neighboring bands. Similar figures and arguments can be extended to the remaining six QPTs discussed in this Letter. Notice that each time two bands cross and undergo a QPT, the sum of their Chern numbers is preserved, as expected [2].

**Predicted edge states.** The predicted number of edge states within the gap between the bands $E_{k,n}$ and $E_{k,n+1}$ is

$$\nu_n = \sum_{n'=n+1}^{15} C_{n'}. \quad (8)$$
The chiral edge states propagate clockwise (counterclockwise) for positive (negative) $\nu_y$ [9]. For instance, we predict a clockwise edge state within the bulk gap between $E_{k,13}$ and $E_{k,14}$ in SkX1. Let SkX3 be the result of applying the time-reversal operator, i.e., flipping all spins, to SkX1. In Ref. [41], we mentioned that since the Hamiltonian in Eq. (1) is time-reversal symmetric, SkX1 and SkX3 are degenerate in energy. It was also mentioned that the two states can appear concurrently, separated by domain walls. SkX3 has the same magnon spectrum as SkX1, while the Chern numbers change sign. At the interface between two topologically nontrivial systems $A$ and $B$, with $\nu_A = \nu_B$ and $\nu_\ell = \nu_J$, in the same energy interval, one expects $|\nu_A - \nu_B|$ edge states [36]. Therefore, along a domain wall between SkX1 and SkX3, we predict two chiral edge states within the gap between $E_{k,13}$ and $E_{k,14}$.

**Conclusion.** We found eight quantum topological phase transitions in two distinct skyrmion crystals that are stabilized in a time-reversal-symmetric model. Time-reversal symmetry is spontaneously broken by the magnetic ordering of the skyrmions, and therefore nonzero Chern numbers of the magnon bands are possible. This is a bosonic analog of the quantum anomalous Hall effect. The quantum topological phase transitions, driven by a tunable easy-axis anisotropy at zero temperature, are signaled by jumps in the Chern numbers. We illustrated how the closing and subsequent reopening of the gaps between magnon bands leads to these jumps in the Chern numbers, and how the Berry curvature depends on these gaps.

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[1] D. J. Thouless, M. Kohmoto, M. P. Nightingale, and M. den Nijs, Quantized Hall Conductance in a Two-Dimensional Periodic Potential, Phys. Rev. Lett. 49, 405 (1982).
[2] J. E. Avron, R. Seiler, and B. Simon, Homotopy and Quantization in Condensed Matter Physics, Phys. Rev. Lett. 51, 51 (1983).
[3] F. D. M. Haldane, Model for a Quantum Hall Effect without Landau Levels: Condensed-Matter Realization of the “Parity Anomaly”, Phys. Rev. Lett. 61, 2015 (1988).
[4] C.-X. Liu, S.-C. Zhang, and X.-L. Qi, The Quantum Anomalous Hall Effect: Theory and Experiment, Annu. Rev. Condens. Matter Phys. 7, 301 (2016).
[5] C. L. Kane and E. J. Mele, Z3 Topological Order and the Quantum Spin Hall Effect, Phys. Rev. Lett. 95, 146802 (2005).
[6] M. Z. Hasan and C. L. Kane, Colloquium: Topological insulators, Rev. Mod. Phys. 82, 3045 (2010).
[7] B. A. Bernevig, T. L. Hughes, and S.-C. Zhang, Quantum Spin Hall Effect and Topological Phase Transition in HgTe Quantum Wells, *Science* **314**, 1757 (2006).

[8] J. E. Moore and L. Balents, Topological invariants of time-reversal-invariant band structures, *Phys. Rev. B* **75**, 121306 (2007).

[9] R. Shindou, R. Matsumoto, S. Murakami, and J.-i. Ohe, Topological chiral magronic edge mode in a magronic crystal, *Phys. Rev. B* **87**, 174427 (2013).

[10] R. Shindou, J.-i. Ohe, R. Matsumoto, S. Murakami, and E. Saitoh, Chiral spin-wave edge modes in dipolar magnetic thin films, *Phys. Rev. B* **87**, 174402 (2013).

[11] L. Zhang, J. Ren, J.-S. Wang, and B. Li, Topological magnon insulator in insulating ferromagnet, *Phys. Rev. B* **87**, 144401 (2013).

[12] P. A. McClarty, Topological Magnons: A Review, *Annu. Rev. Condens. Matter Phys.* **13**, 171 (2022).

[13] S. K. Kim, H. Ochoa, R. Zarzuela, and Y. Tserkovnyak, Realization of the Haldane–Kane–Mele Model in a System of Localized Spins, *Phys. Rev. Lett.* **117**, 227201 (2016).

[14] A. Rückriegel, A. Brataas, and R. A. Duine, Bulk and edge spin transport in topological magnon insulators, *Phys. Rev. B* **97**, 081106 (2018).

[15] E. Thingstad, A. Kamra, A. Brataas, and A. Sudbø, Chiral Phonon Transport Induced by Topological Magnons, *Phys. Rev. Lett.* **122**, 107201 (2019).

[16] S. A. Owerre, A first theoretical realization of honeycomb topological magnon insulator, *J. Phys. Condens. Matter* **28**, 386001 (2016).

[17] H. Huang, T. Kariyado, and X. Hu, Topological magnon modes on honeycomb lattice with coupling textures, *Sci. Rep.* **12**, 6257 (2022).

[18] K. Nakata, J. Klinovaja, and D. Loss, Magnonic quantum Hall effect and Wiedemann-Franz law, *Phys. Rev. B* **95**, 125429 (2017).

[19] K. H. Lee, S. B. Chung, K. Park, and J.-G. Park, Magnetic quantum spin Hall state in the zigzag and stripe phases of the antiferromagnetic honeycomb lattice, *Phys. Rev. B* **98**, 180401 (2018).

[20] S. A. Owerre, Floquet topological magnons, *J. Phys. Commun.* **1**, 021002 (2017).

[21] S.-G. Je, H.-S. Han, S. K. Kim, S. A. Montoya, W. Chao, I.-S. Jeong, and M. I. Katsnelson, A DMI guide to magnets micro-world, *J. Exp. Theor. Phys.* **132**, 506 (2021).

[22] A. Fert, V. Cros, and J. Sampaio, Skyrmions on the track, *Nat. Nanotechnol.* **8**, 152 (2013).

[23] V. V. Mazurenko, Y. O. Kvashnin, A. I. Lichtenstein, and M. I. Katsnelson, A DMI guide to magnets micro-world, *J. Exp. Theor. Phys.* **132**, 506 (2021).

[24] E. A. Stepanov, C. Dutreix, and M. I. Katsnelson, Dynamical and Reversible Control of Topological Spin Textures, *Phys. Rev. Lett.* **118**, 157201 (2017).

[25] E. A. Stepanov, S. A. Nikolaev, C. Dutreix, M. I. Katsnelson, and V. V. Mazurenko, Heisenberg-exchange-free nanoskyrmion mosaic, *J. Phys.: Condens. Matter* **31**, L07T01 (2019).

[26] N. Romming, C. Hanneken, M. Menzel, J. E. Bickel, B. Wolter, K. von Bergmann, A. Kubetzka, and R. Wiesendanger, Writing and Deleting Single Magnetic Skyrmions, *Science* **341**, 636 (2013).

[27] P.-J. Hsu, A. Kubetzka, A. Finco, N. Romming, K. von Bergmann, and R. Wiesendanger, Electric-field-driven switching of individual magnetic skyrmions, *Nat. Nanotechnol.* **12**, 123 (2017).

[28] G. Yu, P. Upadhyaya, Q. Shao, H. Wu, G. Yin, X. Li, C. He, W. Jiang, X. Han, P. K. Amiri, and K. L. Wang, Room-temperature skyrmion shift device for memory application, *Nanotechnology* **17**, 261 (2017).

[29] X. Zhang, M. Ezawa, and Y. Zhou, Magnetic skyrmion logic gates: conversion, duplication and merging of skyrmions, *Sci. Rep.* **5**, 9400 (2015).

[30] C. Psaroudaki and C. Panagopoulos, Skyrmion Qubits: A New Class of Quantum Logic Elements Based on Nanoscale Magnetization, *Phys. Rev. Lett.* **127**, 067201 (2021).

[31] A. Roldán-Molina, A. S. Nunez, and J. Fernández-Rossier, Topological spin waves in the atomic-scale magnetic skyrmion crystal, *New J. Phys.* **18**, 045015 (2016).

[32] S. A. Díaz, J. Klinovaja, and D. Loss, Topological Magnons and Edge States in Antiferromagnetic Skyrmion Crystals, *Phys. Rev. Lett.* **122**, 187203 (2019).

[33] S. A. Díaz, T. Hirosawa, J. Klinovaja, and D. Loss, Chiral magnonic edge states in ferromagnetic skyrmion crystals controlled by magnetic fields, *Phys. Rev. Res.* **2**, 013231 (2020).

[34] J. Waizner, Spin wave excitations in magnetic helices and skyrmion lattices, Ph.D. thesis, Universität zu Köln (2016).

[35] T. Weber, D. M. Fobes, J. Waizner, P. Steffens, G. S. Tucker, M. Böhm, L. Beddrich, C. Franz, H. Gabold, R. Bewley, D. Voneshen, M. Skoulatos, R. Georgii, G. Ehlers, A. Bauer, C. Pfleiderer, P. Böni, M. Janoschek, and M. Garst, Topological magnon band structure of emergent Landau levels in a skyrmion lattice, *Science* **375**, 1025 (2022).

[36] K. Mæland and A. Sudbø, Quantum fluctuations in the order parameter of quantum skyrmion crystals, *Phys. Rev. B* **105**, 224416 (2022).

[37] O. M. Sotnikov, V. V. Mazurenko, J. Colbois, F. Mila, M. I. Katsnelson, and V. V. Mazurenko, Heisenberg-exchange-free nanoskyrmion mosaic, *J. Phys.: Condens. Matter* **31**, L07T01 (2019).

[38] E. Saitoh, Chiral spin-wave edge modes in dipolar magnetic thin films, *Phys. Rev. B* **87**, 157201 (2017).

[39] P. A. McClarty, Topological Magnons: A Review, *Annu. Rev. Condens. Matter Phys.* **13**, 171 (2022).

[40] T. Weber, D. M. Fobes, J. Waizner, P. Steffens, G. S. Tucker, M. Böhm, L. Beddrich, C. Franz, H. Gabold, R. Bewley, D. Voneshen, M. Skoulatos, R. Georgii, G. Ehlers, A. Bauer, C. Pfleiderer, P. Böni, M. Janoschek, and M. Garst, Topological magnon band structure of emergent Landau levels in a skyrmion lattice, *Science* **375**, 1025 (2022).

[41] K. Mæland and A. Sudbø, Quantum fluctuations in the order parameter of quantum skyrmion crystals, *Phys. Rev. B* **105**, 224416 (2022).

[42] O. M. Sotnikov, V. V. Mazurenko, J. Colbois, F. Mila, M. I. Katsnelson, and E. A. Stepanov, Probing the topology of the quantum analog of a classical skyrmion, *Phys. Rev. B* **103**, L060404 (2021).

[43] V. Lohani, C. Hickey, J. Masell, and A. Rosch, Quantum Skyrmions in Frustrated Ferromagnets, *Phys. Rev. X* **9**, 041063 (2019).

[44] C. Castelnovo and C. Chamon, Quantum topological phase transition at the microscopic level, *Phys. Rev. B* **77**, 054433 (2008).

[45] A. Hamma, W. Zhang, S. Haas, and D. A. Lidar, Entanglement, fidelity, and topological entropy in a quantum phase transition to topological order, *Phys. Rev. B* **77**, 155111 (2008).
E. Samimi, M. H. Zarei, and A. Montakhab, Global entanglement in a topological quantum phase transition, Phys. Rev. A 105, 032438 (2022).

S. Heinze, K. von Bergmann, M. Menzel, J. Brede, A. Kubetzka, R. Wiesendanger, G. Bihlmayer, and S. Blügel, Spontaneous atomic-scale magnetic skyrmion lattice in two dimensions, Nat. Phys. 7, 713 (2011).

L. Webster and J.-A. Yan, Strain-tunable magnetic anisotropy in monolayer CrCl$_3$, CrBr$_3$, and CrI$_3$, Phys. Rev. B 98, 144411 (2018).

R. Albaridy, A. Manchon, and U. Schwingenschlögl, Tunable magnetic anisotropy in Cr–trihalide Janus monolayers, J. Condens. Matter Phys. 32, 355702 (2020).

See Supplemental Material on page 7 for (i) details regarding the ground states and how they were obtained, (ii) the introduction of the Holstein-Primakoff transformation through rotated coordinates and the Fourier transform of the magnon description, (iii) the diagonalization to obtain the magnon bands, and (iv) details of the Chern number calculation. The Supplemental Material contains Refs. [55–62].

J. T. Haraldsen and R. S. Fishman, Spin rotation technique for non-collinear magnetic systems: application to the generalized Villain model, J. Condens. Matter Phys. 21, 216001 (2009).

J. H. P. Colpa, Diagonalization of the quadratic boson Hamiltonian, Phys. A: Stat. Mech. Appl. 93, 327 (1978).

S. E. Mousavi, J. E. Pask, and N. Sukumar, Efficient adaptive integration of functions with sharp gradients and cusps in n-dimensional parallelepipeds, Int. J. Numer. Methods Eng. 91, 343 (2012).

F. Sausset and G. Tarjus, Periodic boundary conditions on the pseudosphere, J. Phys. A: Math. Theor. 40, 12873 (2007).

S. Kirkpatrick, C. D. Gelatt, Jr., and M. P. Vecchi, Optimization by Simulated Annealing, Science 220, 671 (1983).

F. J. dos Santos, M. dos Santos Dias, F. S. M. Guimarães, J. Bouaziz, and S. Lounis, Spin-resolved inelastic electron scattering by spin waves in noncollinear magnets, Phys. Rev. B 97, 024431 (2018).

A. Roldán-Molina, M. J. Santander, A. S. Nunez, and J. Fernández-Rossier, Quantum fluctuations stabilize skyrmion textures, Phys. Rev. B 92, 245436 (2015).

C. Tsallis, Diagonalization methods for the general bilinear Hamiltonian of an assembly of bosons, J. Math. Phys. 19, 277 (1978).

M.-w. Xiao, Theory of transformation for the diagonalization of quadratic Hamiltonians, arXiv:0908.0787 (2009).

K. Mæland, Excitation Spectrum and Superfluidity of Weakly Interacting, Spin-Orbit Coupled Bose-Einstein Condensate, Master’s thesis, Norwegian University of Science and Technology, (2020).

K. Mæland, A. T. G. Janssønn, J. H. Rygh, and A. Sudbø, Plane- and stripe-wave phases of a spin-orbit-coupled Bose-Einstein condensate in an optical lattice with a Zeeman field, Phys. Rev. A 102, 053318 (2020).
Supplemental material for “Quantum topological phase transitions in skyrmion crystals”

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S1. INTRODUCTION

Sections S2 and S3 of this supplemental material repeat relevant details in Ref. [41] that are left out of the main text of “Quantum topological phase transitions in skyrmion crystals”. Section S2 concerns details of obtaining the classical ground state numerically, while Sec. S3 details the Holstein-Primakoff (HP) approach toward finding the magnon energy spectrum. In both sections, more details are included than what was previously presented in Ref. [41]. Section S4 gives details of the numerical calculation of Chern numbers.

S2. CLASSICAL GROUND STATE

S2.1. Periodicity

The classical Hamiltonian is obtained from the Hamiltonian in Eq. (1) of the main text by setting $S_i = Sm_i$. Here, $m_i$ is a unit vector along the direction of the spin at lattice site $i$, while $S$ is the uniform spin magnitude. This gives

$$H(\{m_i\}) = H_{\text{ex}} + H_{\text{DM}} + H_A + H_4,$$  \hspace{1cm} \text{(S1)}

where

$$H_{\text{ex}} = -JS^2 \sum_{\langle ij \rangle} m_i \cdot m_j,$$  \hspace{1cm} \text{(S2)}

$$H_{\text{DM}} = S^2 \sum_{\langle ij \rangle} D_{ij} \cdot (m_i \times m_j),$$  \hspace{1cm} \text{(S3)}

$$H_A = -KS^2 \sum_i m_i^{z2},$$  \hspace{1cm} \text{(S4)}
FIG. S1. SkXt for varying periodicities. Colors indicate $m_{iz}$, while the projection of $m_i$ on the $xy$ plane is shown with arrows. (a) SkXt with $\lambda_x = 3.5, \lambda_y = 1$ is a helical state. SkXt with (b) $\lambda_x = 3, \lambda_y = 6$, (c) $\lambda_x = 5, \lambda_y = 6$, (d) $\lambda_x = 4.9, \lambda_y = 6$, and (e) $\lambda_x = 9, \lambda_y = 10$ are skyrmion states. Their energies are given in Table SI.

$H_4 = U S_4^4 \sum_{ijkl} \left[ (m_i \cdot m_j)(m_k \cdot m_l) + (m_i \cdot m_l)(m_j \cdot m_k) - (m_i \cdot m_k)(m_j \cdot m_l) \right].$ (S5)

Minimizing $H(\{m_i\})$ with respect to $\{m_i\}$ will reveal the classical ground state (GS) of the system. Potential candidates are collinear states such as ferromagnetic states, coplanar states such as helical states, or noncoplanar states such as skyrmions. With $U \neq 0$ it turns out ferromagnetic and helical states will be disfavored, making skyrmion crystal (SkX) GSs more likely. The Hamiltonian is far too complicated to obtain an analytic solution for the GS, and so we must resort to numerical methods. Additionally, we wish to study a bulk system with a periodic magnetic state, in order to introduce the HP approach [52]. To obtain results within a reasonable amount of time, a limited number of spins can be included in the simulations. If the periodicity of the ground state is unknown, any chosen lattice size with periodic boundary conditions (PBCs) will introduce finite-size effects. Hence, it is a major advantage to know the periodicity of the GS, before searching for the optimal state with that periodicity.

The periodicity of the GS will depend on the parameters in the model. Our approach is to tune the parameters to ensure that a SkX with the same periodicity as the best commensurate approximation to the one observed experimentally in Ref. [47], is the GS of the system. From the supplementary information of Ref. [47] we take the skyrmion constructor $m_i = (\sin \phi_i \cos \theta_i / | \sin \phi_i |, \cos \phi_i \sin \theta_i, \cos \phi_i \cos \theta_i)$, where $\phi_i = Q_M \cdot r_i, \theta_i = Q_K \cdot r_i, Q_M = 2\pi \hat{x} / \lambda_x, Q_K = 2\pi \hat{y} / (\lambda_y \sqrt{3}/2)$. $

\lambda_x$ is the periodicity in the $x$ direction in terms of lattice sites, while $\lambda_y$ is the periodicity in the $y$ direction in terms of lattice chains. We name these trial states SkXt. For any rational numbers $\lambda_x$ and $\lambda_y$ it is possible to construct a finite-sized lattice with correct PBCs in order to calculate the energy per site.

We show a selection of SkXt states in Fig. S1. In Table SI we show the total energy of SkXt as well as the four individual contributions for various choices of periodicity. The exchange coupling, $H_{ex}$, prefers a ferromagnetic GS. Among the helical and skyrmion-like states $H_{ex}$ prefers large periodicities since then neighboring spins are more aligned. The Dzyaloshinskii-Moriya interaction (DMI), $H_{DM}$, favors helical states, especially the helical state with $\lambda_x \approx 3.4, \lambda_y = 1$. The helical state with $\lambda_x = 3.5, \lambda_y = 1$ is shown in Fig. S1(a). The easy-axis anisotropy, $H_A$, has no effect on the periodicity unless it becomes the dominant energy in the system. It prefers collinear states along the $z$ axis. Coplanar states are also preferred over noncoplanar states. The four-spin interaction, $H_4$, disfavors collinear and coplanar states. It prefers noncoplanar SkXt states with small
We now assume that with the parameters tuned to the values above, the GS is a SkX with a periodicity of $\lambda_x = 5, \lambda_y = 6$. We can then perform simulations with lattice sizes $5 \times 6, 10 \times 12$, etc., or, we can define a 15 site magnetic unit cell containing the only 15 unique spins, and implement PBCs similar to a twisted torus [55]; see Fig. S2. The latter approach gave the states with lowest energy, while the first choices gave states that visually were periodic as assumed, but where the periodicity was not exact and the resultant energy slightly higher. Hence they serve as nice tests, but were not used to obtain any results.

We implemented a Monte Carlo simulated annealing approach [37, 38, 56] to search for potential GSs. The algorithm is

1. Create a random starting distribution $\{\theta_i, \phi_i\}$ specifying $m_i = (\sin \theta_i \cos \phi_i, \sin \theta_i \sin \phi_i, \cos \theta_i)$. The inclination $\theta_i$ and the azimuth $\phi_i$ are the familiar angles in spherical coordinates.
2. Pick a random site \( j \) and update \( \theta_j, \phi_j \).

3. Calculate the change in energy, \( \Delta H \), and \( W = e^{-\beta \Delta H} \), where \( \beta \) is the inverse temperature.

4. Pick a random number, \( r \), between 0 and 1. If \( W > r \) accept the new state.

5. Decrease temperature, and repeat from step 2 for a chosen number of repetitions, usually quite large.

Several thermalization schemes were attempted in order to gradually cool the system from a high initial temperature to a low temperature compared to the relevant energy scales in the system. The idea is that starting with high temperature should reduce the risk of getting stuck in local minima. Additionally running many repetitions with a very low temperature after the initial thermalization aims to obtain the lowest energy state, i.e., the GS of the system. Alternatively, one can start with a state that is assumed to be similar to the GS of the system. One such choice would be SkXt at \( \lambda_x = 5, \lambda_y = 6 \). In the latter case, the starting temperature should also be low. If not, the initial spin updates will just bring the system out of the SkXt state into what is essentially a high energy random state.

We also employed a self-consistent iteration approach \([36, 57, 58]\), which yielded states with lower energy than any we obtained using Monte Carlo simulated annealing. Therefore, all results are calculated from numerical GSs obtained using self-consistent iteration. The algorithm is \([41, 57]\).

1. Create a random starting distribution \( \{\theta_i, \phi_i\} \) specifying \( \{m_i\} \). Alternatively, start from a judiciously chosen distribution based on the suspected GS.

2. Assume \( n \in \mathbb{N} \) iterations have been completed. Then, for each spin, calculate the magnetic torques \( T^\theta_i = \partial_\theta H \) and \( T^\phi_i = \partial_\phi H \). Use these to set new angles \( \theta_{i+1} = \theta_i - \alpha T^\theta_i \) and \( \phi_{i+1} = \phi_i - \alpha T^\phi_i \), where \( \alpha \) is the mixing parameter \([57]\). We used \( \alpha J = 0.001 \) and 0.0001.

3. Repeat step 2 until self-consistency is reached.

We checked that values of \( K/J \) in the interval \([0, 1]\) had no effect on the periodicity of the SkX GS. It is likely this interval can be extended, but we believe large magnitudes of \( K \) will eventually change the periodicity of the GS. An interesting effect was found at \( K = K_t \), where we determined that \( K_t/J \) is somewhere in the interval \((0.518, 0.519)\). There, the SkX GS changes nature, but not periodicity. For \( 0 \leq K/J \leq 0.518 \) the lowest energy state is named SkX1, which is shown in Fig. 1(a) of the main text. Tuning \( K \) in that region only adjusts the \( z \) component of all spins gradually. However, for \( 0.519 \leq K/J \leq 1 \) the center of the skyrmion relocates along the \( x \)-axis by approximately one quarter lattice constant. States where the center relocates to the left or right are degenerate in energy. We chose to focus on the state SkX2 shown in Fig. 1(e) of the main text, where the center has relocated to the left. The lower symmetry of SkX2 compared to SkX1 distinguishes it as a different phase. The phase transition between SkX1 and SkX2 occurs at zero temperature by tuning a parameter in the Hamiltonian, and is therefore a quantum phase transition (QPT) \([41]\).

SkX1 and SkX2 are found to have lower energy than the trial state SkXt that was used to determine the periodicity. New GSs are found at each considered value of \( K/J \). To double check the proposed GSs we also started from random states on larger lattices of varying sizes using Monte Carlo simulated annealing. Due to the complicated energy landscape, Monte Carlo simulations might get stuck in local minima. To check if this prevented finding helical states, we also searched specifically for the best possible coplanar state, without finding any states with lower energy than SkX1 and SkX2.

\[ S2.2.1. \text{Net magnetization in ground states} \]

We define the GS net magnetizations as

\[ m_{\alpha} = \frac{1}{N} \sum_i m_{i\alpha}. \]  \hspace{1cm} (S6)

In SkX1 \( m_x = m_y = 0 \) within numerical accuracy, while \( m_z \) is small and negative. The net magnetization is \( m_z \approx -0.002 \) at low \( K/J \), with decreasing magnitude for increasing \( K/J \). In SkX2 \( m_y = m_z = 0 \) within numerical accuracy, while \( m_x \) is small and negative. The net magnetization is \( m_x \approx -0.014 \) with a minor dependence on \( K \). Hence, both states have a small net magnetization even though there is no external magnetic field. Apparently, a small net magnetization allows the formation of a state with lower energy than any state with zero net magnetization.
S3. HOLSTEIN-PRIMAKOFF APPROACH

S3.1. Inserting rotated coordinates

Following Ref. [52] we introduce a local orthonormal frame \( \{ \hat{e}_1, \hat{e}_2, \hat{e}_3 \} \) with \( \hat{e}_3 = m_i \). I.e., a coordinate system where the third axis points along the magnetization in the classical GS. We let \( \hat{e}_{\alpha} = (R_{\alpha\beta})^{-1} \hat{r}_{\beta} \), where \( \hat{r}_{\beta} \) are the Cartesian axes. Inverting we get \( \hat{r}_{\alpha} = R_{\alpha\beta} \hat{e}_{\beta} \),

\[
\begin{pmatrix}
\hat{r}_x \\
\hat{r}_y \\
\hat{r}_z
\end{pmatrix} = \begin{pmatrix}
\cos \theta_i \cos \phi_i & -\sin \theta_i \sin \phi_i & \cos \phi_i \\
\cos \theta_i \sin \phi_i & \cos \theta_i \cos \phi_i & -\sin \phi_i \\
-\sin \theta_i & 0 & \cos \theta_i
\end{pmatrix}
\begin{pmatrix}
\hat{e}_1 \\
\hat{e}_2 \\
\hat{e}_3
\end{pmatrix}. \tag{S7}
\]

Note that \((R^t)^{-1} = (R^t)^T\) since it is an SO(3) rotation matrix. Values of \( \{ \theta_i, \phi_i \} \) are given by the GS.

This can then be inserted in the Hamiltonian,

\[ H_{\text{ex}} = -\sum_{(i,j)} \sum_{\alpha=\{x,y,z\}} J_{\alpha} S_{i\alpha} S_{j\alpha} = -\sum_{(i,j)} \sum_{(\alpha,\beta,\gamma=\{1,2,3\}}} J_{\alpha\beta\gamma} (S_i \cdot \hat{r}_\alpha)(S_j \cdot \hat{r}_\alpha) \]

\[ = -\sum_{(i,j)} \sum_{\alpha\beta\gamma=\{1,2,3\}}} J_{\alpha\beta\gamma} (S_i \cdot \hat{e}_{\beta})(S_j \cdot \hat{e}_{\beta}) R^i_{\alpha\beta} R^j_{\alpha\beta} = -\sum_{(i,j)} J_{\alpha\beta\gamma} S_{i\beta} S_{j\gamma} R^i_{\alpha\beta} R^j_{\alpha\beta}, \]

\[ H_{\text{DM}} = \sum_{(i,j)} D_{ij} \cdot [S_i \times S_j] = \sum_{(i,j)} \sum_{\alpha\beta\gamma=\{1,2,3\}}} \epsilon_{\alpha\beta\gamma} D_{ij\gamma} (S_i \cdot \hat{e}_{\beta})(S_j \cdot \hat{e}_{\beta}) R^i_{\beta\gamma} R^j_{\beta\gamma} \]

\[ = \sum_{(i,j)} \epsilon_{\alpha\beta\gamma} D_{ij\gamma} S_{i\beta} S_{j\gamma} R^i_{\beta\gamma} R^j_{\beta\gamma}, \]

\[ H_A = -K \sum_i S^2_{iz} = -K \sum_i \left[ \sum_{\alpha} (S_i \cdot \hat{e}_{\alpha}) R^i_{3\alpha} \right]^2 = -K \sum_i (S_{i\alpha} R^i_{3\alpha})^2, \]

\[ H_4 = U \sum_{ijkl} \left[ (S_i \cdot S_j)(S_k \cdot S_l) + (S_i \cdot S_l)(S_j \cdot S_k) - (S_i \cdot S_k)(S_j \cdot S_l) \right] \]

\[ = U \sum_{ijkl} \left[ (S_i S_j R^i_{\alpha\beta} R^j_{\alpha\beta})(S_k S_l R^k_{\gamma\delta} R^l_{\gamma\delta}) + (S_i S_l R^i_{\alpha\beta} R^j_{\alpha\beta})(S_k S_j R^k_{\gamma\delta} R^l_{\gamma\delta}) \right] \]

\[ - (S_i S_j S_k S_l R^i_{\alpha\beta} R^j_{\alpha\beta} R^k_{\gamma\delta} R^l_{\gamma\delta}). \tag{S8} \]

We have defined \( S_{i\alpha} \equiv S_i \cdot \hat{e}_{\alpha} \) for \( \alpha = \{1,2,3\} \) and adopted the Einstein summation convention over Greek letters running over \( \{1,2,3\} \). For the sake of generality, we keep the possibility of anisotropic exchange open here.

The HP transformation is introduced as \( S_i \cdot m_i = S - a_i^\dagger a_i, \) \( S_{i\pm} = S_i \cdot \hat{e}_1 \pm i S_i \cdot \hat{e}_2, \) \( S_{i=} = \sqrt{2S - a_i^\dagger a_i}, \) \( S_{i=} = a_i \sqrt{2S - a_i^\dagger a_i}. \) Here, \( a_i^\dagger (a_i) \) creates (destroys) a magnon at lattice site \( i \). We truncate at second order in magnon operators from now on, an approximation which should be valid at low temperature compared to the magnon gap [41]. \( S_{i\pm} = S_{i1} \pm iS_{i2} \) leads to

\[ S_{i1} = \frac{1}{2} (S_{i+} + S_{i-}) = \sqrt{\frac{S}{2}} (a_i + a_i^\dagger), \quad S_{i2} = \frac{1}{2t} (S_{i+} - S_{i-}) = i \sqrt{\frac{S}{2}} (a_i^\dagger - a_i). \tag{S9} \]

Finally, \( S_{i3} = S - a_i^\dagger a_i \) and we are ready to introduce the HP transformation. We consider one term of the Hamiltonian at a time, and write out the sums over indices in \( S_i \). For \( H_{\text{ex}} \) we write out the sums over \( \beta \) and \( \gamma \). Separating into operator-independent terms, linear terms and quadratic terms yields

\[ H_{\text{ex},0} = -S^2 \sum_{(ij)} J_{\alpha\beta\gamma} R^i_{\alpha\beta} R^j_{\alpha\beta}, \]

\[ H_{\text{ex},1} = -S \sqrt{2S} \sum_{(ij)} [J_{\alpha\beta\gamma} (R^i_{\alpha\beta} R^j_{\alpha\beta} - i R^i_{\alpha\gamma} R^j_{\alpha\gamma}) a_i + \text{H.c.}], \]

\[ H_{\text{ex},2} = -\frac{S}{2} \sum_{(ij)} [ -4 J_{\alpha\beta\gamma} R^i_{\alpha\beta} R^j_{\alpha\beta} a_i^\dagger a_i. \]
Here, we performed some rewrites so that, e.g., \(H_{\text{ex},1}\) only depends on operators at lattice site \(i\). H.c. denotes the Hermitian conjugate of the preceding term. Specializing to isotropic exchange, defining \(\tilde{e}^{\pm}_i = e^{\pm}_i \pm i \tilde{e}^{\pm}_i\) and using that the columns of \(R^i\) are the unit vectors \(\hat{e}^i\), we can simplify,

\[
H_{\text{ex},2} = -\frac{JS}{2} \sum_{\langle ij \rangle} \left[ -4 \tilde{e}^{\pm}_i \cdot \tilde{e}^{\pm}_j a_i^\dagger a_j + \tilde{e}^\pm_i a_i a_j + \text{H.c.} + \tilde{e}^\pm_i \cdot \tilde{e}^\pm_j a_i^\dagger a_j^\dagger + \text{H.c.} \right].
\]  

(S11)

For \(H_{\text{DM}}\) we write out the sums over \(\delta\) and \(\epsilon\) and get

\[
H_{\text{DM,0}} = S^2 \sum_{\langle ij \rangle} \epsilon_{\alpha \beta \gamma} D_{iji\alpha} R^i_{\gamma \beta} R^j_{\gamma \beta},
\]

\[
H_{\text{DM,1}} = S \sqrt{2S} \sum_{\langle ij \rangle} \epsilon_{\alpha \beta \gamma} D_{iji\alpha} [(R^i_{\beta \gamma} R^j_{\gamma \beta} - i R^i_{\beta \gamma} R^j_{\gamma \beta}) a_i + \text{H.c.}]
\]

\[
+ (R^i_{\beta \gamma} R^j_{\gamma \beta} - i R^i_{\beta \gamma} R^j_{\gamma \beta}) a_i a_j + \text{H.c.},
\]

\[
H_{\text{DM,2}} = \frac{S}{2} \sum_{\langle ij \rangle} \epsilon_{\alpha \beta \gamma} D_{iji\alpha} [-2 R^i_{\gamma \beta} R^j_{\gamma \beta} (a_i^\dagger a_i + a_j^\dagger a_j)
\]

\[
+ (R^i_{\gamma \beta} R^j_{\gamma \beta} - R^i_{\beta \gamma} R^j_{\gamma \beta} - i R^i_{\beta \gamma} R^j_{\gamma \beta} - i R^i_{\beta \gamma} R^j_{\gamma \beta}) a_i a_j + \text{H.c.}
\]

\[
+ (R^i_{\gamma \beta} R^j_{\gamma \beta} + R^i_{\beta \gamma} R^j_{\gamma \beta} + i R^i_{\beta \gamma} R^j_{\gamma \beta} - i R^i_{\beta \gamma} R^j_{\gamma \beta}) a_i^\dagger a_j^\dagger + \text{H.c.}].
\]  

(S12)

We seek to rewrite this to a slightly simpler form, but need to be more careful than for the exchange interaction. In \(H_{\text{DM,1}}\) we let \(i \leftrightarrow j\) in the term with \(a_j\). This is ok for a sum \(\sum_{\langle ij \rangle}\). Then we rename \(\beta \leftrightarrow \gamma\) and use \(\epsilon_{\alpha \beta \gamma} = -\epsilon_{\alpha \gamma \beta}\) and \(D_{iji\alpha} = -D_{iji\alpha}\) to find that the \(a_j\) term can be rewritten to be equivalent to the \(a_i\) term. Similar rewrites also show that the \(a_i^\dagger a_j\) terms can be rewritten as the \(a_i^\dagger a_i\) terms. Hence,

\[
H_{\text{DM,1}} = S \sqrt{2S} \sum_{\langle ij \rangle} \epsilon_{\alpha \beta \gamma} D_{iji\alpha} (R^i_{\beta \gamma} R^j_{\gamma \beta} - i R^i_{\beta \gamma} R^j_{\gamma \beta}) a_i + \text{H.c.},
\]

\[
H_{\text{DM,2}} = \frac{S}{2} \sum_{\langle ij \rangle} D_{ij} \cdot [-4 (\tilde{e}^\pm_i \times \tilde{e}^\pm_j) a_i^\dagger a_i + (\tilde{e}^\pm_i \times \tilde{e}^\pm_j) a_i a_j + \text{H.c.} + (\tilde{e}^\pm_i \times \tilde{e}^\pm_j) a_i^\dagger a_j^\dagger + \text{H.c.}].
\]  

(S13)

In \(H_A\), we perform the sum over \(\alpha\), then square the sum and replace all \(S_{i\alpha} S_{i\alpha'}\) by their HP transformation. This gives

\[
H_{A,0} = -KS^2 \sum_i (R^i_{\beta \gamma})^2 = -KS^2 \sum_i \cos^2 \theta_i,
\]

\[
H_{A,1} = -KS \sqrt{2S} \sum_i [(R^i_{\beta \gamma} - i R^i_{\beta \gamma}) a_i + \text{H.c.}] = KS \sqrt{2S} \sum_i (\sin \theta_i \cos \theta_i a_i + \text{H.c.}),
\]

\[
H_{A,2} = -KS \sum_i \left[ -4 (R^i_{\beta \gamma})^2 + (R^i_{\beta \gamma})^2 + (R^i_{\beta \gamma})^2 a_i^\dagger a_i + [(R^i_{\beta \gamma})^2 + (R^i_{\beta \gamma})^2] a_i a_i^\dagger \right.
\]

\[
+ [(R^i_{\beta \gamma})^2 - (R^i_{\beta \gamma})^2 - 2i R^i_{\beta \gamma} R^i_{\beta \gamma}] a_i a_i + \text{H.c.})
\]

\[= -KS \sum_i \left[ (\sin^2 \theta_i - 4 \cos^2 \theta_i) a_i^\dagger a_i + \sin^2 \theta_i a_i a_i^\dagger + \sin^2 \theta_i a_i a_i + \text{H.c.} \right].
\]  

(S14)

We inserted the definition of the rotation matrix \(R^i\) from Eq. (S7) in order to clean up the expressions. The fact that \(R^i_{\beta \gamma} = 0\) allows significant simplification.

In \(H_4\) we perform the sums over \(\beta, \gamma, \epsilon\) and \(\zeta\). Then, we need only keep the terms involving at least two \(S_{i\beta} S_{j\gamma} S_{k\epsilon} S_{l\zeta}\). All other terms will be more than quadratic in magnon operators. We focus on the first term \(ijjk\) when performing the HP transformation. The second, \(iijk\), and third, \(-ijkj\), terms can then be obtained by permuting \(ijkl\) appropriately.

\[
H_{4,0} = US^4 \sum_{ijkl} (R^i_{\alpha \beta} R^i_{\alpha \beta} R^i_{\alpha \beta} R^i_{\alpha \beta} + R^i_{\alpha \beta} R^i_{\alpha \beta} R^i_{\alpha \beta} R^i_{\alpha \beta} - R^i_{\alpha \beta} R^i_{\alpha \beta} R^i_{\alpha \beta} R^i_{\alpha \beta}).
\]  

(S15)
The linear terms originating from the $ijkl$ term are

$$[ijkl]_1 = U S^3 \sqrt{\frac{5}{2}} \sum_{ijkl} (R_{\alpha_1}^{i} R_{\alpha_2}^{j} R_{\delta_1}^{k} R_{\delta_2}^{l} - i R_{\alpha_1}^{i} R_{\alpha_2}^{j} R_{\delta_1}^{k} R_{\delta_2}^{l} a_i + \text{H.c.})$$

$$+ (R_{\alpha_3}^{i} R_{\alpha_4}^{j} R_{\delta_3}^{k} R_{\delta_4}^{l} - i R_{\alpha_3}^{i} R_{\alpha_4}^{j} R_{\delta_3}^{k} R_{\delta_4}^{l} a_i + \text{H.c.})$$

$$+ (R_{\alpha_3}^{i} R_{\alpha_4}^{j} R_{\delta_3}^{k} R_{\delta_4}^{l} - i R_{\alpha_3}^{i} R_{\alpha_4}^{j} R_{\delta_3}^{k} R_{\delta_4}^{l} a_i + \text{H.c.})$$

$$+ (R_{\alpha_3}^{i} R_{\alpha_4}^{j} R_{\delta_3}^{k} R_{\delta_4}^{l} - i R_{\alpha_3}^{i} R_{\alpha_4}^{j} R_{\delta_3}^{k} R_{\delta_4}^{l} a_i + \text{H.c.})].$$

(S16)

The best way to treat the linear terms is to rewrite all of them to $a_i$ form and then check that the total coefficient in front of $a_i$ is zero. All these terms originate from $(S_i \cdot S_j)(S_k \cdot S_l)$. We can clearly interchange $i \leftrightarrow j$ and/or $k \leftrightarrow l$ without changing this expression. We can also interchange $(i,j) \leftrightarrow (k,l)$ since $(S_i \cdot S_j)(S_k \cdot S_l) = (S_k \cdot S_l)(S_i \cdot S_j)$. Hence, in the second line of $[ijkl]_1$ we can let $i \leftrightarrow j$ to see that it is the same as the first line. In the third line we can let $(i,j) \leftrightarrow (k,l)$ to see that it is the same as the first line. In the fourth line we let $(i,j) \leftrightarrow (k,l)$ and then $i \leftrightarrow j$ to see that it is the same as the first line. In total therefore, we simply have 4 times the first line:

$$[ijkl]_1 = 2 U S^3 \sqrt{25} \sum_{ijkl} (R_{\alpha_1}^{i} R_{\alpha_2}^{j} R_{\delta_1}^{k} R_{\delta_2}^{l} - i R_{\alpha_1}^{i} R_{\alpha_2}^{j} R_{\delta_1}^{k} R_{\delta_2}^{l} a_i + \text{H.c.})$$

(S17)

Permuting to get the $iljk$ and $-ikjl$ terms gives

$$H_{4,1} = 2 U S^3 \sqrt{25} \sum_{ijkl} (R_{\alpha_1}^{i} R_{\alpha_2}^{j} R_{\delta_1}^{k} R_{\delta_2}^{l} + R_{\alpha_1}^{i} R_{\alpha_2}^{j} R_{\delta_1}^{k} R_{\delta_2}^{l} - R_{\alpha_1}^{i} R_{\alpha_2}^{j} R_{\delta_1}^{k} R_{\delta_2}^{l} - i R_{\alpha_1}^{i} R_{\alpha_2}^{j} R_{\delta_1}^{k} R_{\delta_2}^{l} a_i + \text{H.c.})$$

(S18)

We take a similar approach to the quadratic terms. From $ijkl$ we get

$$[ijkl]_2 = U S^3 \sum_{ijkl} (R_{\alpha_1}^{i} R_{\alpha_2}^{j} R_{\delta_1}^{k} R_{\delta_2}^{l} (a_i^\dagger a_i + a_j^\dagger a_j + a_k^\dagger a_k + a_l^\dagger a_l)$$

$$+ (R_{\alpha_3}^{i} R_{\alpha_4}^{j} R_{\delta_3}^{k} R_{\delta_4}^{l} - i R_{\alpha_3}^{i} R_{\alpha_4}^{j} R_{\delta_3}^{k} R_{\delta_4}^{l} a_i + \text{H.c.})$$

$$+ (R_{\alpha_3}^{i} R_{\alpha_4}^{j} R_{\delta_3}^{k} R_{\delta_4}^{l} - i R_{\alpha_3}^{i} R_{\alpha_4}^{j} R_{\delta_3}^{k} R_{\delta_4}^{l} a_i + \text{H.c.})$$

$$+ (R_{\alpha_3}^{i} R_{\alpha_4}^{j} R_{\delta_3}^{k} R_{\delta_4}^{l} - i R_{\alpha_3}^{i} R_{\alpha_4}^{j} R_{\delta_3}^{k} R_{\delta_4}^{l} a_i + \text{H.c.})$$

(S19)

We attempt to simplify this by making more of the sum centered at site $i$, instead of, e.g., counting contributions from only sites $k$ and $l$ here. For the $a_j a_j$ term we let $i \leftrightarrow j$. For $a_k a_k$ we let $(i,j) \leftrightarrow (k,l)$ and then rename $\alpha \leftrightarrow \delta$. For $a_l a_l$ we let $(i,j) \leftrightarrow (k,l)$, then $i \leftrightarrow j$, and finally rename $\alpha \leftrightarrow \delta$. Then, this is just 4 times the $a_i a_i$ term. We can bring the $a_k a_k$ and $a_l a_l$ terms on the same form as the $a_j a_j$ and $a_j a_j$ terms by interchanging $i \leftrightarrow j$ and $k \leftrightarrow l$. We can bring the $a_j a_k$ and $a_j a_l$ terms on the same form as the $a_j a_j$ and $a_j a_j$ terms by interchanging $i \leftrightarrow j$ and $k \leftrightarrow l$. 
In total, we therefore have

\[
[ijk]_2 = \sum_{i,j,k,l} \left[-4R_{a3}^i R_{a3}^j R_{a3}^k R_{a3}^l a_i a_j + (R_{a1}^i R_{a1}^j R_{a3}^k R_{a3}^l - R_{a2}^i R_{a2}^j R_{a3}^k R_{a3}^l - iR_{a1}^i R_{a2}^j R_{a3}^k R_{a3}^l - iR_{a2}^i R_{a1}^j R_{a3}^k R_{a3}^l) a_i a_j \right] + H.c.
\]

Permuting to get the \(ijk\) and \(ikj\) terms gives

\[
H_{4,2} = U S^2 \sum_{i,j,k,l} \left[\sum_{r} \left(-4\left(\hat{e}_3^i \cdot \hat{e}_3^j \hat{e}_3^k \cdot \hat{e}_3^l + \hat{e}_3^i \cdot \hat{e}_3^j \hat{e}_3^k \cdot \hat{e}_3^l - \hat{e}_3^i \cdot \hat{e}_3^j \hat{e}_3^k \cdot \hat{e}_3^l\right) a_i a_j \right] + H.c.
\]

\[
[ijk]_2 = \sum_{i,j,k,l} \left[-4R_{a3}^i R_{a3}^j R_{a3}^k R_{a3}^l a_i a_j + (R_{a1}^i R_{a1}^j R_{a3}^k R_{a3}^l - R_{a2}^i R_{a2}^j R_{a3}^k R_{a3}^l - iR_{a1}^i R_{a2}^j R_{a3}^k R_{a3}^l - iR_{a2}^i R_{a1}^j R_{a3}^k R_{a3}^l) a_i a_j \right] + H.c.
\]

\[
H_{4,2} = U S^2 \sum_{i,j,k,l} \left[\sum_{r} \left(-4\left(\hat{e}_3^i \cdot \hat{e}_3^j \hat{e}_3^k \cdot \hat{e}_3^l + \hat{e}_3^i \cdot \hat{e}_3^j \hat{e}_3^k \cdot \hat{e}_3^l - \hat{e}_3^i \cdot \hat{e}_3^j \hat{e}_3^k \cdot \hat{e}_3^l\right) a_i a_j \right] + H.c.
\]

\[
S3.2. \text{ Fourier transform}
\]

The Fourier transform (FT) is introduced as

\[
a_i = \frac{1}{\sqrt{N^{(r)}}} \sum_{k} e^{i k \cdot r_i} a_k^{(r)}, \tag{S22}
\]

\[
a_k^{(r)} = \frac{1}{\sqrt{N^{(r)}}} \sum_{i} e^{-i k \cdot r_i} a_i. \tag{S23}
\]

We assume lattice site \(i\), located at \(r_i\), resides on sublattice \(r\). \(N^{(r)}\) is the number of lattice sites on sublattice \(r\), the sum over \(k\) is restricted to the first Brillouin zone (1BZ) of sublattice \(r\), 1BZ\((r)\), and \(a_k^{(r)}\) is a magnon destruction operator associated with sublattice \(r\).

The sum in the exchange Hamiltonian is converted in the following way

\[
\sum_{(ij)} = \sum_{(rs)} \sum_{s} \sum_{\delta_{(r,s)}}. \tag{S24}
\]

Here, we sum over all sublattices \(r\) and for each \(r\) we sum over those sublattices \(s\) that contain sites which are nearest neighbors to a site on sublattice \(r\). We sum over all sites \(i\) on sublattice \(r\) and all possible vectors \(\delta_{(r,s)}\) connecting two sites \(i\) and \(j\) that are nearest neighbors, located on sublattice \(r\) and \(s\), respectively.

An example of a FT of a term in \(H_{ex}\)

\[
\sum_{(ij)} \hat{e}_3^i \cdot \hat{e}_3^j a_i a_j = \sum_{(rs)} \hat{e}_3^r \cdot \hat{e}_3^s \sum_{s} \sum_{\delta_{(r,s)}} \frac{1}{\sqrt{N^{(r)}}} \sum_{k} e^{i k \cdot r_i} a_k^{(r)} \frac{1}{\sqrt{N^{(s)}}} \sum_{k'} e^{i k' \cdot (r_i + \delta_{(r,s)})} a_k^{(s)}. \tag{S25}
\]
Since the orthonormal frame \( \{ \hat{e}_1^r, \hat{e}_2^r, \hat{e}_3^r \} \) is the same for all lattice sites \( i \) on sublattice \( r \), we can move \( \hat{e}_r^\alpha \) outside the sum over \( i \) by writing \( \hat{e}_r^\alpha \). Now focus on the FT:

\[
\frac{1}{\sqrt{N(s)N(r)}} \sum_{k} \sum_{k'} \sum_{i} e^{i(k+k'_r) \cdot r_i} N(r) \sum_{\delta_{(r,s)}} \sum_{\delta_{(r,s)}} e^{i k'_r \cdot \delta_{(r,s)}} a_k^{(r)} \gamma \delta \cdot a_{k'}^{(r)} = \sqrt{N(r)N(s)} \sum_{k} \sum_{k'} \delta_{k,-k'} \sum_{\delta_{(r,s)}} e^{i k'_r \cdot \delta_{(r,s)}} a_k^{(r)} a_{k'}^{(r)}.
\]

We used that \( \sum_{i} e^{i(k+k'_r) \cdot r_i} = N(r) \delta_{k,-k'} \). It is clear that in the end we only get contributions from those \( k \) such that \( k \in 1BZ^{(r)} \) and \( -k \in 1BZ^{(s)} \). This is indicated by \( (r,-s) \) above the summation sign. We also introduce a quantity \( \gamma_{NN} \) which is the number of lattice sites \( j \) on a specific sublattice \( s \) that are nearest neighbors (NN) to lattice site \( i \) on sublattice \( r \). Following this approach, the end result is

\[
H_{\text{ex},2} = \sum_{(r,s)} \left( F_{\text{ex},rdr}^{(r,s)} \sum_{k} a_k^{(r)} a_{r_k}^{(r)} + F_{\text{ex},r}^{(r,s)} \sum_{k} \gamma_{k}^{(r,s)} a_k^{(r)} a_{-k}^{(r)} + H.c. \right) ,
\]

where we defined

\[
F_{\text{ex},rdr}^{(r,s)} = 2JS \hat{e}_3^r \cdot \hat{e}_3^{r,s} \gamma_{NN} \gamma_{NN} ,
\]

\[
F_{\text{ex},r}^{(r,s)} = -\frac{JS}{2} \sqrt{\frac{N(s)}{N(r)}} \hat{e}_-^r \cdot \hat{e}_+^{r,s} ,
\]

\[
F_{\text{ex},r}^{(r,s)} = -\frac{JS}{2} \sqrt{\frac{N(r)}{N(s)}} \hat{e}_-^r \cdot \hat{e}_+^{r,s} .
\]

Subscript \( r, s \) denote which kind of sublattice combination the following operators belong to, and \( d \) is added to indicate a creation operator instead of an annihilation operator. Superscript \( rs \) indicates that the prefactor depends on the specific sublattices.

For the DMI terms, we need to take into account that the DMI vector depends on the direction of the vector connecting \( i \) and \( j, namely \( \delta_{(r,s)} \). Hence, \( D_{ij} = D_{(r,s)} \hat{e}_r^\alpha \), when we rewrite the sum. Note that \( \delta_{(r,s)} \) is the shortest vector connecting two lattice sites on sublattice \( r \) and \( s \). Then, we find

\[
H_{\text{DM},2} = \sum_{(r,s)} \left( F_{\text{DM},rdr}^{(r,s)} \sum_{k} a_k^{(r)} a_{r_k}^{(r)} + F_{\text{DM},r}^{(r,s)} \sum_{k} \gamma_{k}^{(r,s)} a_k^{(r)} a_{-k}^{(r)} + H.c. \right) ,
\]

with

\[
F_{\text{DM},rdr}^{(r,s)} = -2S D_{(r,s)} \cdot (\hat{e}_3^r \times \hat{e}_3^{r,s}) \gamma_{NN} \gamma_{NN} ,
\]

\[
F_{\text{DM},r}^{(r,s)} = \frac{S}{2} \sqrt{\frac{N(s)}{N(r)}} D_{(r,s)} \cdot (\hat{e}_-^r \times \hat{e}_+^{r,s}) ,
\]

\[
F_{\text{DM},r}^{(r,s)} = \frac{S}{2} \sqrt{\frac{N(r)}{N(s)}} D_{(r,s)} \cdot (\hat{e}_-^r \times \hat{e}_+^{r,s}) .
\]

For \( H_{A,2} \) we get

\[
H_{A,2} = \sum_{r} \left( \sum_{k} \left[ F_{A,rdr}^{(r)} a_k^{(r)} a_{r_k}^{(r)} + F_{A,rr}^{(r)} a_k^{(r)} a_{r_k}^{(r)} \right] + \sum_{k} \left[ F_{A,rrd}^{(r)} a_k^{(r)} a_{-k}^{(r)} + H.c. \right] \right) ,
\]
\[ F_{A,rdr}^{(r)} = -\frac{KS}{2} \left( \sin^2 \theta_r - 4 \cos^2 \theta_r \right) \quad \text{and} \quad F_{A,rr}^{(r)} = -\frac{KS}{2} \sin^2 \theta_r \]  

(S36)

For \( H_4 \) we rewrite the sum as

\[ \sum_{ijkl} \sum_{rstu} \sum_{r,s,t,u} \sum_{\delta(r,s)} \sum_{\delta(r,t)} \sum_{\delta(r,u)} \]  

(S37)

We sum over all sublattices \( r \). Then, for each \( r \) we sum over sublattices \( s, t, u \) such that \( i, j, k, l \) on sublattices \( r, s, t, u \) form diamonds oriented counterclockwise of minimal area [47, 50]. Then we sum over all sites \( i \) on sublattice \( r \) and all vectors connecting \( i \) to sites \( j, k, l \) such that \( i, j, k, l \) form counterclockwise diamonds and where \( j, k, l \) are located on sublattices \( s, t, u \). Let us introduce \( z_{rs}^z \), as the number of sites on sublattice \( s \) that can form diamonds with a specific site on sublattice \( r \). Often this is one, but let us be general. Then, we find that

\[ H_{4,2} = \sum_{rstu} \sum_{r,s,t,u} \sum_{r,s,t,u} \sum_{\delta(r,s)} \sum_{\delta(r,t)} \sum_{\delta(r,u)} \]  

(S38)

with

\[ F_{4,rdr}^{(r,s,t,u)} = -4US^3z_{rs}^z \delta_{rt} \delta_{ru} \left( (\hat{e}_3 \cdot \hat{e}_s)(\hat{e}_3 \cdot \hat{e}_u) + (\hat{e}_3 \cdot \hat{e}_u)(\hat{e}_3 \cdot \hat{e}_s) - (\hat{e}_3 \cdot \hat{e}_s)(\hat{e}_3 \cdot \hat{e}_u) \right), \]  

(S39)

\[ F_{4,rs}^{(r,s,t,u)} = US^3 \delta_{rt} \delta_{ru} \left( \frac{N(r)}{N(s)} \left[ (\hat{e}_3^r \cdot \hat{e}_u^s)(\hat{e}_3^r \cdot \hat{e}_3^u) + (\hat{e}_3^r \cdot \hat{e}_3^u)(\hat{e}_3^r \cdot \hat{e}_u) - (\hat{e}_3^r \cdot \hat{e}_u)(\hat{e}_3^r \cdot \hat{e}_3^u) \right] \right), \]  

(S40)

\[ F_{4,rsd}^{(r,s,t,u)} = US^3 \delta_{rt} \delta_{ru} \left( \frac{N(r)}{N(s)} \left[ (\hat{e}_3^r \cdot \hat{e}_s^u)(\hat{e}_3^r \cdot \hat{e}_3^u) + (\hat{e}_3^r \cdot \hat{e}_3^u)(\hat{e}_3^r \cdot \hat{e}_s^u) - (\hat{e}_3^r \cdot \hat{e}_s^u)(\hat{e}_3^r \cdot \hat{e}_3^u) \right] \right), \]  

(S41)

\[ F_{4,rt}^{(r,s,t,u)} = US^3 \delta_{rs} \delta_{ru} \left( \frac{N(r)}{N(s)} \left[ (\hat{e}_3^r \cdot \hat{e}_s^r)(\hat{e}_3^r \cdot \hat{e}_3^r) + (\hat{e}_3^r \cdot \hat{e}_3^r)(\hat{e}_3^r \cdot \hat{e}_s^r) - (\hat{e}_3^r \cdot \hat{e}_s^r)(\hat{e}_3^r \cdot \hat{e}_3^r) \right] \right), \]  

(S42)

\[ F_{4,rtd}^{(r,s,t,u)} = US^3 \delta_{rs} \delta_{ru} \left( \frac{N(r)}{N(s)} \left[ (\hat{e}_3^r \cdot \hat{e}_s^r)(\hat{e}_3^r \cdot \hat{e}_3^u) + (\hat{e}_3^r \cdot \hat{e}_3^u)(\hat{e}_3^r \cdot \hat{e}_s^r) - (\hat{e}_3^r \cdot \hat{e}_s^r)(\hat{e}_3^r \cdot \hat{e}_3^u) \right] \right), \]  

(S43)

\[ F_{4,ru}^{(r,s,t,u)} = US^3 \delta_{rs} \delta_{ru} \left( \frac{N(r)}{N(s)} \left[ (\hat{e}_3^u \cdot \hat{e}_3^r)(\hat{e}_3^u \cdot \hat{e}_3^r) + (\hat{e}_3^u \cdot \hat{e}_3^r)(\hat{e}_3^u \cdot \hat{e}_3^r) - (\hat{e}_3^u \cdot \hat{e}_3^r)(\hat{e}_3^u \cdot \hat{e}_3^r) \right] \right), \]  

(S44)

\[ F_{4,rud}^{(r,s,t,u)} = US^3 \delta_{rs} \delta_{ru} \left( \frac{N(r)}{N(s)} \left[ (\hat{e}_3^u \cdot \hat{e}_3^r)(\hat{e}_3^u \cdot \hat{e}_3^r) + (\hat{e}_3^u \cdot \hat{e}_3^r)(\hat{e}_3^u \cdot \hat{e}_3^r) - (\hat{e}_3^u \cdot \hat{e}_3^r)(\hat{e}_3^u \cdot \hat{e}_3^r) \right] \right), \]  

(S45)

S3.3. Collecting the total Hamiltonian

Collecting all terms we get

\[ H_0 = -JS^2 \sum_{(ij)} R_{\alpha \beta} \bar{R}_{\alpha \beta}^i + S^2 \sum_{(ij)} \epsilon_{\alpha \beta \gamma} D_{ij \gamma} R_{\beta \gamma}^i \bar{R}_{\beta \gamma}^i - KS^2 \sum_i \cos^2 \theta_i \]

\[ + US^4 \sum_{ijkl} \left( R_{\alpha \beta}^i R_{\alpha \beta}^j R_{\beta \gamma}^i R_{\beta \gamma}^j + R_{\alpha \beta}^i R_{\alpha \beta}^j R_{\beta \gamma}^i \bar{R}_{\beta \gamma}^j - R_{\alpha \beta}^i R_{\alpha \beta}^j R_{\beta \gamma}^i \bar{R}_{\beta \gamma}^j \right), \]  

(S46)

This is simply the classical Hamiltonian from Eq. (S1).
All linear terms in the Hamiltonian should vanish if we expand around the correct ground state of the system [52]. Collecting, we find

$$H_1 = S\sqrt{2S} \sum_{i} \left[ \sum_{j \in \text{NN}} \left[ - J (R^i_{\alpha_1} R^j_{\alpha_3} - i R^i_{\alpha_2} R^j_{\alpha_3}) + \epsilon_{\alpha_\gamma} \gamma D_{ij} \epsilon \gamma (R^i_{\beta_1} R^j_{\gamma_3} - i R^i_{\beta_2} R^j_{\gamma_3}) \right] + K \sin \theta_i \cos \theta_i \right]$$

$$+ 2US^2 \sum_{jkl} \left( R^j_{\alpha_1} R^j_{\alpha_2} R^l_{\alpha_3} R^l_{\alpha_3} - R^j_{\alpha_1} R^j_{\alpha_2} R^l_{\alpha_3} R^l_{\alpha_3} \right) - R^j_{\alpha_1} R^j_{\alpha_2} R^l_{\alpha_3} R^l_{\alpha_3} + i R^j_{\alpha_1} R^j_{\alpha_2} R^l_{\alpha_3} R^l_{\alpha_3} + i R^j_{\alpha_1} R^j_{\alpha_2} R^l_{\alpha_3} R^l_{\alpha_3} \right) a_i + \text{H.c.} \quad (S47)$$

We sum over all $j$ that are nearest neighbors (NNs) to $i$, and all $jkl$ that can form counterclockwise diamonds of minimal area with $i$. The linear terms are zero if the quantity inside the large square brackets is zero at all $i$. Collecting the FTed quadratic part gives

$$H_2 = \sum_{\langle rs \rangle} \sum_k \left[ F_{\text{ex,rd}}^{(r,s)} a_k^{(r)} a_k^{(r)} \right] + \sum_{\langle r,s,t,u \rangle} \sum_k \left[ F_{\text{ex,rd}}^{(r,s)} a_k^{(r)} a_k^{(r)} \right] + \sum_{\langle r,s,t,u \rangle} \sum_k \left[ F_{\text{ex,rd}}^{(r,s)} a_k^{(r)} a_k^{(r)} \right] + \text{H.c.} \quad (S48)$$

Here, we include an analytic proof that the terms that are linear in magnon operators are zero when expanding around the GS of the system. Checking numerically that the linear terms in Eq. (S47) are zero therefore serves as a check on whether we have obtained the true GS from the numerical simulations. In Appendix A2 of Ref. [41] we argue that the linear terms are zero within numerical accuracy.

Picking a specific lattice site $i'$ we can require that $H_1 = 0$ by setting the real and imaginary parts of the coefficients to zero,

$$\sum_{j' \in \text{NN}} \left( - J_{i'} R^i_{\alpha_1} R^j_{\alpha_3} + \epsilon_{\alpha_\gamma} \gamma D_{i'j} \epsilon \gamma (R^i_{\beta_1} R^j_{\gamma_3} - i R^i_{\beta_2} R^j_{\gamma_3}) \right) + K \sin \theta_i \cos \theta_i$$

$$+ 2US^2 \sum_{j'k'l'} \left( R^i_{\alpha_1} R^j_{\alpha_2} R^l_{\alpha_3} R^l_{\alpha_3} + R^i_{\alpha_1} R^j_{\alpha_2} R^l_{\alpha_3} R^l_{\alpha_3} - R^i_{\alpha_1} R^j_{\alpha_2} R^l_{\alpha_3} R^l_{\alpha_3} + R^i_{\alpha_1} R^j_{\alpha_2} R^l_{\alpha_3} R^l_{\alpha_3} \right) = 0, \quad (S49)$$

$$\sum_{j' \in \text{NN}} \left( - J_{i'} R^i_{\alpha_2} R^j_{\alpha_3} + \epsilon_{\alpha_\gamma} \gamma D_{i'j} \epsilon \gamma (R^i_{\beta_1} R^j_{\gamma_3} - i R^i_{\beta_2} R^j_{\gamma_3}) \right)$$

$$+ 2US^2 \sum_{j'k'l'} \left( R^i_{\alpha_2} R^j_{\alpha_1} R^l_{\alpha_3} R^l_{\alpha_3} + R^i_{\alpha_2} R^j_{\alpha_1} R^l_{\alpha_3} R^l_{\alpha_3} - R^i_{\alpha_2} R^j_{\alpha_1} R^l_{\alpha_3} R^l_{\alpha_3} + R^i_{\alpha_2} R^j_{\alpha_1} R^l_{\alpha_3} R^l_{\alpha_3} \right) = 0. \quad (S50)$$

We now show that setting $\frac{\partial H_1}{\partial \theta_i} = 0$ and $\frac{\partial H_1}{\partial \phi_i} = 0$ leads to the same constraints. It is important to take into account that any $ijk$ in the sum could be $i'$. If, e.g., $j = i'$, we cyclically permute so that $k = j'$, $l = k'$ and $i = l'$. Then we see that all choices
and obtain a matrix version of the total quadratic Hamiltonian. We use the numbering shown in Fig. S2. With reference to the definition in Eq. (S7),

\[
\frac{\partial R_{\alpha}^{i'}}{\partial \theta_{i'}} = R_{\alpha 1}^{i'}, \quad \text{and} \quad \frac{\partial R_{\alpha}^{i'}}{\partial \theta_{i'}} = \sin \theta_{i'} R_{\alpha 2}^{i'}. \tag{S51}
\]

So we get

\[
\frac{\partial H_0}{\partial \theta_{i'}} = 2S^2 \left( \sum_{j' \in N_N} \left( -J_{j} R_{\alpha 1}^{i'} R_{\alpha 3}^{j'} + \epsilon_{\alpha \beta \gamma} D_{j'} j' \cdot \hat{r}_\alpha R_{\beta 1}^{i'} R_{\gamma 3}^{j'} + K \sin \theta_{i'} \cos \theta_{i'} \right) + 2US^2 \sum_{j' k' \in N} \left( R_{\alpha 1}^{i'} R_{\alpha 3}^{j'} R_{\alpha 3}^{k'} R_{\alpha 3}^{l'} + R_{\alpha 1}^{i'} R_{\alpha 3}^{j'} R_{\alpha 3}^{k'} R_{\alpha 3}^{l'} - R_{\alpha 1}^{i'} R_{\alpha 3}^{j'} R_{\alpha 3}^{k'} R_{\alpha 3}^{l'} \right) \right),
\]

\[
\frac{\partial H_0}{\partial \theta_{i'}} = 2S^2 \sin \theta_{i'} \left( \sum_{j' \in N_N} \left( -J_{j} R_{\alpha 2}^{i'} R_{\alpha 3}^{j'} + \epsilon_{\alpha \beta \gamma} D_{j'} j' \cdot \hat{r}_\alpha R_{\beta 2}^{i'} R_{\gamma 3}^{j'} + 2US^2 \sum_{j' k' \in N} \left( R_{\alpha 2}^{i'} R_{\alpha 3}^{j'} R_{\alpha 3}^{k'} R_{\alpha 3}^{l'} + R_{\alpha 2}^{i'} R_{\alpha 3}^{j'} R_{\alpha 3}^{k'} R_{\alpha 3}^{l'} - R_{\alpha 2}^{i'} R_{\alpha 3}^{j'} R_{\alpha 3}^{k'} R_{\alpha 3}^{l'} \right) \right) \right). \tag{S52}
\]

The case \( \sin \theta_{i'} = 0 \) for all \( i' \) is uninteresting in this context, so we see that requiring \( \frac{\partial H_0}{\partial \theta_{i'}} = 0 \) and \( \frac{\partial H_0}{\partial \phi_{i'}} = 0 \) leads to the same constraints as we had for \( H_1 = 0 \) in Eqs. (S49) and (S50).

Hence, if we are in an extremum of \( H_0 \), e.g. the GS, \( H_1 = 0 \). This is also fairly obvious from arguments presented in Ref. [52]; the function \( H(\{a_i, a_i\}) \) should be in a minimum with respect to the operators, and so all linear terms must vanish.

### S3.5. Specializing to the ground states

SkX1 and SkX2 contain 15 sublattices that are equal, centered rectangular lattices with primitive vectors \( a_1 = (5/2, -3\sqrt{3}/2) \) and \( a_2 = (5/2, 3\sqrt{3}/2) \). Hence, all sublattices have the same 1BZ and the same number of lattice sites, i.e., all \( N^{(r)} \) are equal. We name them \( N' = N/N_{\text{SL}} \), where \( N \) is the total number of lattice sites, and \( N_{\text{SL}} = 15 \) is the number of sublattices. In reciprocal space the primitive vectors are \( b_1 = 2\pi (1/5, -1/3\sqrt{3}) \) and \( b_2 = 2\pi (1/5, 1/3\sqrt{3}) \). Hence, the 1BZ is a nonregular hexagon with corners at \( (\pm 52\pi/135, 0) \), \( (2\pi/135, \pm 2\pi/3\sqrt{3}) \) and \( (\pm 2\pi/135, \pm 2\pi/3\sqrt{3}) \). The 1BZ is invariance symmetric. Therefore, all sums over momentum are restricted to the same values of \( k \), and we can replace all of them with \( \sum_k \) with the understanding that \( k \) is restricted to the 1BZ of the sublattices. See Fig. S3 for sketches of the Wigner-Seitz cell in real space and the 1BZ in reciprocal space.

Furthermore, the SkX1 and SkX2 GSs are such that we never encounter a situation where more than one site on the same sublattice is a nearest neighbor to a specific site on another sublattice, nor can make diamonds in \( H_1 \). Hence, all the \( z_{rs}^{NN}, z_{rs}^{S} \) factors are 1, and all sums over vectors connecting lattice sites only contain one term, \( \gamma_{k}^{(r,s)} = e^{-i k \cdot \delta_{(r,s)}} \).

To get any further we need to adopt a numbering of the sublattices, and then perform the sums over the sublattices to obtain a matrix version of the total quadratic Hamiltonian. We use the numbering shown in Fig. S2. With reference...
to that numbering we state for reference the 12 counterclockwise diamonds in $H_4$ where sublattice $r = 10$. These are $(r, s, t, u) = \{(10, 11, 7, 6), (10, 11, 6, 5), (10, 6, 2, 5), (10, 6, 5, 9), (10, 5, 4, 9), (10, 5, 9, 13), (10, 9, 3, 13), (10, 9, 13, 14), (10, 13, 15, 14), (10, 13, 14, 11), (10, 14, 1, 11), (10, 14, 11, 6)\}.$

### S3.6. Quadratic part

Having specialized to our SkX GSs, the quadratic part can be written

\[
H_2 = \sum_k \left\{ \left[ (F_{ex,rs})^{(r,s)}_{k} + (F_{DM,rsd})^{(r,s)}_{k} \right] a_k^{(r)} a_k^{\dagger (s)} + (F_{ex,rs} + F_{DM,rs}) \gamma_k a_k^{(r)} a_{-k}^{\dagger} + \text{H.c.} \right. \\
+ \left. (F_{ex,rd}^{(r,s)} + F_{DM,rsd}^{(r,s)}) \gamma_k a_k^{(r)} a_k^{\dagger} + \text{H.c.} \right]\]

\[
+ \sum_r \left[ \left[ (F_{A,rr}^{(r,s,t,u)} a_k^{(r)} a_k^{\dagger} + F_{A,rr}^{(r,s,t,u)} \gamma_k a_k^{(r)} a_{-k}^{\dagger} + \text{H.c.} \right] \\
+ F_{4,rr}^{(r,s,t,u)} \gamma_k a_k^{(r)} a_{-k}^{\dagger} + \text{H.c.} \right] \\
+ \left[ \left[ (F_{A,rr}^{(r,s,t,u)} a_k^{(r)} a_k^{\dagger} + F_{A,rr}^{(r,s,t,u)} \gamma_k a_k^{(r)} a_{-k}^{\dagger} + \text{H.c.} \right] \\
+ F_{4,rr}^{(r,s,t,u)} \gamma_k a_k^{(r)} a_{-k}^{\dagger} + \text{H.c.} \right]. \right\} \right) \text{.} \tag{S54}
\]

We used a commutator which leads to a shift of $H_0$,

\[
H_0' = H_0 + N' \sum_r F_{A,rr}^{(r)}. \tag{S55}
\]

Upon writing out the sums over sublattices, we organize the factors into the following types: $a_k^{(r)} a_k^{(s)}$, $a_k^{(r)} a_k^{(s)}$ with $r$ and $s > r$ nearest neighbors, $a_k^{(r)} a_k^{(s)}$ with $r$ and $t > r$ next nearest neighbors, $a_k^{(r)} a_k^{(s)}$ with $r$ and $s > r$ nearest neighbors, and $a_k^{(r)} a_k^{(s)}$ with $r$ and $t > r$ next nearest neighbors. These are then later rewritten using commutators and letting $k \rightarrow -k$ in the sum where necessary, e.g.,

\[
\sum_k c_k a_k^{(r)} a_{-k}^{(s)} = \frac{1}{2} \sum_k (c_{-k} a_{-k}^{(r)} a_k^{(s)} + c_k a_k^{(r)} a_{-k}^{(s)}) \tag{S56}
\]

where $c_k$ is the coefficient in front of $a_k^{(r)} a_{-k}^{(s)}$. Contributions to $a_k^{(r)} a_k^{(s)}$ with $r$ and $s > r$ nearest neighbors comes from two sources; the H.c. of $a_k^{(r)} a_k^{(s)}$ and from $a_k^{(r)} a_k^{(s)}$ with $r' = s > r$, $s' = r$. We find that these two coefficients are the same, and so we store the prefactor as twice the H.c. of $a_k^{(r)} a_k^{(s)}$.

Similarly, $a_k^{(r)} a_{-k}^{(s)}$ with $r$ and $s > r$ nearest neighbors comes from two sources: $a_k^{(r)} a_{-k}^{(s)}$ and $a_k^{(r)} a_{-k}^{(s)}$ with $r' = s > r$, $s' = r$ and $k \rightarrow -k$. The latter is permissible due to the sum over $k$ over an inversion symmetric BZ. Again, these two coefficients are found to be the same, and are stored as twice the coefficient of $a_k^{(r)} a_{-k}^{(s)}$. Finally, similar considerations apply to the terms involving $r$ and $t$ as next-nearest neighbors.

As presented in Ref. [41] we can write

\[
H_2 = \frac{1}{2} \sum_k a_k^\dagger M_k a_k, \tag{S57}
\]

where $a_k^\dagger = (a_k^{(1)}, a_k^{(2)}, \ldots, a_k^{(15)}, a_{-k}^{(1)}, \ldots, a_{-k}^{(15)})$ and

\[
M_k = \begin{pmatrix}
\eta_k r \\
\nu_k r \\
\eta_{-k} r
\end{pmatrix}, \tag{S58}
\]

The matrix elements can be written,

\[
\eta_k r, s = \eta k r s + S e^{ik r s} \Lambda^{r, s}, \tag{S59}
\]
\[ \eta_r = 2S \sum_s |J_{(r,s)}| \hat{e}_r^s \cdot \hat{e}_r^s - D_{(r,s)} \cdot (\hat{e}_r^s \times \hat{e}_r^s)| - KS[1 - 3(\hat{e}_r^s \cdot \hat{z})^2] - 4S^2 \sum_{s,t,u} U_{(r,s,t,u)}[(\hat{e}_s^r \cdot \hat{e}_s^r)(\hat{e}_t^s \cdot \hat{e}_u^t) + (\hat{e}_s^r \cdot \hat{e}_u^t)(\hat{e}_t^s \cdot \hat{e}_s^r) - (\hat{e}_s^r \cdot \hat{e}_t^s)(\hat{e}_t^s \cdot \hat{e}_s^r)], \]  

(S60)

\[ \nu_{r,s} = \nu_r \delta_{r,s} + Se^{ik\delta_{r,s}} \Lambda_{r,s}^{r,s}, \]  

(S61)

\[ \nu_r = -KS(\hat{e}_r^s \cdot \hat{z})^2. \]  

(S62)

Here, \( J_{(r,s)} = J \) if there exists \( i \in r, j \in s \) such that \( i \) and \( j \) are nearest neighbors. Otherwise \( J_{(r,s)} = 0 \). \( D_{(r,s)} = D \delta_{(r,s)} \times \hat{z} \) if there exist \( i \in r, j \in s \) such that \( i \) and \( j \) are nearest neighbors. Otherwise \( D_{(r,s)} = 0 \). \( U_{(r,s,t,u)} = U \) if there exist \( i \in r, j \in s, k \in t, l \in u \) such that sites \( i, j, k, l \) make a counterclockwise diamond of minimal area. Otherwise \( U_{(r,s,t,u)} = 0 \).

S3.7. Diagonalization

We employ the method described in Ref. [53] to diagonalize the system. This is an alternative to the method described in Refs. [59, 60]. The first method works only if the matrix \( M_k \) in the Hamiltonian is positive definite, unlike the latter which is more general. Following Ref. [53] the matrix \( M_k \) in Eq. (S58) is a \( 30 \times 30 \) matrix as opposed to \( 60 \times 60 \) if following Ref. [59]. Hence, the first method gives 15 energy bands, while the latter originally gives 30 bands, which can be reduced to 15 bands using methods similar to those we presented in Refs. [61, 62]. We find that both methods give the same results in the present system. In SkX2, where the excitation spectrum is not inversion symmetric it would be challenging to perform the reduction from 30 to 15 bands without the result from following Ref. [53] as a guide.

The matrix form of the Hamiltonian is Hermitian, \( M_k^\dagger = M_k \), while the submatrices obey \( \eta_k^\dagger = \eta_k, \nu_k^T = \nu_{-k} \). The system is diagonalized with a transformation matrix \( T_k \) as follows:

\[ a_k^\dagger M_k a_k = (a_k^\dagger T_k^\dagger)[(T_k^{-1})^\dagger M_k T_k^{-1}](T_k a_k) = b_k^\dagger D_k b_k, \]  

(S64)

where \( D_k \) is diagonal. The diagonalized operator vector is \( b_k = (b_{k,1}, b_{k,2}, \ldots, b_{k,15}, b_{-k,1}, \ldots, b_{-k,15})^T \). The diagonalized operators retain bosonic commutation relations since the transformation matrix is paraunitary [53], \( T_k^{-1} = J T_k^\dagger J \). Here, \( J \) is a diagonal matrix whose first 15 diagonal elements are 1, and final 15 diagonal elements are \(-1\).

The algorithm for obtaining the transformation matrix is described in Ref. [53] as follows:

1. Find \( K_k \) from the Cholesky decomposition \( M_k = K_k^\dagger K_k \). \( K_k^\dagger \) is lower triangular, while \( K_k \) is upper triangular.
2. Find eigenvectors \( w_{k,1}, \ldots, w_{k,2m} \) and eigenvalues \( E_{k,1}, \ldots, E_{k,2m} \) of the Hermitian \( 2m \times 2m \) matrix \( K_k J K_k^\dagger \). We have \( E_{k,n} = -E_{-k,n+m} \) for \( n \leq m \), where \( E_{k,n} > 0 \) and \( E_{-k,n+m} < 0 \). The \( m \) positive eigenvalues are the excitation spectrum of the system [53].
3. Construct a unitary matrix \( W_k = [w_{k,1}, \ldots, w_{k,2m}] \) from the orthonormal eigenvectors.
4. Construct \( D_k = \text{diag}(E_{k,1}, \ldots, E_{k,m}, -E_{k,m+1}, \ldots, -E_{k,2m}) \) from the eigenvalues \( E_{k,n} \).
5. Calculate \( T_k^{-1} \) row by row from \( K_k T_k^{-1} = W_k D_k^\dagger \) starting at the last row since \( K_k \) is upper triangular.
Step 5 can be performed as follows,

\[
(W_k D_k^{\frac{1}{2}})_{\text{row } 2m-i} = \sum_{j=0}^{i-1} K_{k,2m-i,2m-j} (T_k^{-1})_{\text{row } 2m-j}. 
\]

(S65)

Using \( T_k^{-1} = \mathcal{J} T_k^{\dagger} \mathcal{J} \) gives \( T_k = \mathcal{J} (T_k^{-1})^{\dagger} \mathcal{J} \). Let

\[
T_k = \begin{pmatrix} U_k & W_k \\ V_k & Z_k \end{pmatrix}. 
\]

(S66)

From \( b_k = T_k a_k \), we find

\[
b_{k,n} = \sum_{r=1}^{m} \left[ U_{k,n,r} a_k^{(r)} + W_{k,n,r} a_k^{(r)^\dagger} \right], 
\]

(S67)

\[
b_{-k,n}^\dagger = \sum_{r=1}^{m} \left[ V_{k,n,r} a_k^{(r)} + Z_{k,n,r} a_k^{(r)^\dagger} \right], 
\]

(S68)

\[
b_{-k,n}^\dagger = \sum_{r=1}^{m} \left[ W_{-k,n,r}^* a_k^{(r)} + U_{-k,n,r}^* a_k^{(r)^\dagger} \right]. 
\]

(S69)

Eq. (S68) was found using \( T_k \), while Eq. (S69) was found directly from Eq. (S67) by letting \( k \to -k \) and taking the H.c. This shows that \( W_k = V_{-k}^* \), while \( Z_k = U_{-k}^* \). Hence,

\[
T_k = \begin{pmatrix} U_k & V_{-k}^* \\ V_k & U_{-k}^* \end{pmatrix}. 
\]

(S70)

Using \( T_k^{-1} = \mathcal{J} T_k^{\dagger} \mathcal{J} \) gives

\[
T_k^{-1} = \begin{pmatrix} U_k^{\dagger} & -V_{-k}^{\dagger} \\ -V_k^{\dagger} & U_{-k}^{\dagger} \end{pmatrix}. 
\]

(S71)

Diagonalizing \( H_2 \) in Eq. (S57) yields [53]

\[
H_2 = \frac{1}{2} \sum_{k} \sum_{n=1}^{15} \left( E_{k,n} b_{k,n}^\dagger b_{k,n} - E_{k,n+m} b_{-k,n}^\dagger b_{-k,n} \right). 
\]

(S72)

Letting \( -k \to k \) in last term of the sum gives

\[
H_2 = \frac{1}{2} \sum_{k} \sum_{n=1}^{15} \left( E_{k,n} b_{k,n}^\dagger b_{k,n} - E_{-k,n+m} b_{k,n}^\dagger b_{k,n} \right), 
\]

(S73)

Using a commutator in the final term gives

\[
H_2 = \sum_{k} \sum_{n=1}^{15} E_{k,n} \left( b_{k,n}^\dagger b_{k,n} + \frac{1}{2} \right). 
\]

(S74)

The excitation spectrum is shown in Fig. 1 of the main text, and discussed further in Ref. [41].

S3.7.1. Proofs

a. Statement. The diagonalized operator vector is

\[
b_k = (b_{k,1}, b_{k,2}, \ldots, b_{k,15}, b_{-k,1}^\dagger, \ldots, b_{-k,15}^\dagger)^T. 
\]

(S75)
The expectation value of the Hamiltonian is

\[ \langle H \rangle = H_0 + \sum_{r=1}^{N'} \left( 2F_{k,r}^{(r)} - \eta_r \right) \]

(S85)

The commutator leads to an additional shift of the operator-independent part of the Hamiltonian,

\[ H'_0 = H_0 + \sum_{r} \left( 2F_{k,r}^{(r)} - \eta_r \right) = H_0 + \frac{N'}{2} \sum_{r} (v_r - \eta_r) \]

(S85)

The expectation value of the Hamiltonian is

\[ \langle H \rangle = H_0 + (H'_0 - H_0) + \langle H_2 \rangle \]

(S85)
at zero temperature. In SkX1, we find

\[ \langle H \rangle / NJ \approx 7.360, \quad (H_0^2 - H_0)/NJ \approx 6.490, \quad \langle H_2 \rangle / NJ \approx 6.925, \quad \langle H \rangle / NJ \approx 7.725, \quad (\langle H \rangle - H_0)/NJ \approx 0.195. \] (S91)

The parameters are \( D/J = 2.16, U/J = 0.35, K/J = 0.1, S = 1 \) and we used 40000 points in the sum over \( k \). Notice that \( \langle H \rangle < H_0 \) which shows that quantum fluctuations stabilize the SkX. This agrees with Refs. [42, 58]. The quantum state is energetically preferred over the classical GS. Along with their small size, this is our justification for referring to the skyrmions in SkX1 and SkX2 as quantum skyrmions [41–43].

### S4. DETAILS OF CHERN NUMBER CALCULATION

As shown in Fig. 2(c) in the main text, the Berry curvature develops strong peaks or valleys at values of \( k \) where the band has closely avoided crossings with other bands. This presents a numerical challenge in calculating the integral of the Berry curvature over the 1BZ. This is especially a challenge in SkX2, where many bands have closely avoided crossings at all values of \( K/J \). We used the recursive algorithm in Ref. [54] to obtain adaptive quadratures, where the density of \( k \) values is largest around the sharp peaks and valleys. In general, this gave accurate numerical results, with deviations from integer Chern numbers decreasing to \( O(10^{-5}) \) or better with increasing number of \( k \) points in the quadrature.

For \( K/J = 0.71 \) in SkX2, \( E_{k,3} \) and \( E_{k,4} \) are very close to crossing, with a gap of \( O(10^{-7}J) \). This yields extremely sharp peaks in the Berry curvatures and numerical difficulties led to \( C_3 \approx 0.995 \) and \( C_4 \approx -0.995 \) for both 10^6 and 10^7 \( k \) points in the adaptive quadrature. These are the greatest deviations from integers in our results. We view this as a numerical artifact since \( K/J = 0.71 \) is very close to the value \( K = K_7 \) where \( E_{k,3} \) and \( E_{k,4} \) cross. The numerical Chern numbers are better approximations of integers at values of \( K \) farther away from \( K = K_7 \) than \( K/J = 0.71 \).

The derivatives in the Berry curvature in Eq. (6) in the main text were calculated using forward difference with \( \Delta k_\mu = 10^{-8} \). In the case of an evenly spaced discretization, the integral is converted to a sum via \( \int dk = (A_{1BZ}/N') \sum_k \), where \( A_{1BZ} \) is the area of the 1BZ, and \( N' \) is the number of magnetic unit cells, i.e., the number of \( k \) points in the sum.

The adaptive quadrature relies on subdivisions of the integration interval whenever the difference between two Gaussian quadratures of degree five and eight exceed a chosen tolerance [54]. The application of Gaussian quadratures involve a change of variables in each subdivided integration interval and a generalization to a two-dimensional (2D) integral. The change of interval since Gaussian quadratures are designed for the interval \([-1, 1]\) can be performed as

\[ \int_a^b f(x)dx = \frac{b-a}{2} \int_{-1}^1 dx' f \left( \frac{b-a}{2} x' + \frac{a+b}{2} \right) \] (S92)

\[ = \frac{b-a}{2} \sum_{i=1}^{n} w_i f \left( \frac{b-a}{2} x_i' + \frac{a+b}{2} \right) \] (S93)

\[ = \sum_{i=1}^{n} w_i f(x_i), \] (S94)

with \( w_i = \frac{b-a}{2} w_{i}', x_i = \frac{b-a}{2} x_{i}' + \frac{a+b}{2} \). The weights \( w_i \) and points \( x_i \) are provided by a Gaussian quadrature of degree \( n \). The extension of Gaussian quadratures to 2D is given by

\[ \int_{-1}^1 \int_{-1}^1 f(x, y)dydx = \sum_i \sum_j w_i w_j f(x_i, y_j). \] (S95)