# EFFECT OF IMPURITIES ON STABILITY OF THE SKYRMION PHASE IN A FRUSTRATED HEISENBERG ANTIFERROMAGNET

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## KEYWORDS

Heisenberg antiferromagnet; Geometrical frustration; Skyrmion lattice; Nonmagnetic impurities; Hybrid Monte Carlo

## ABSTRACT

We employ a hybrid Monte Carlo simulation implemented on GPU to study the effect of nonmagnetic impurities in a frustrated Heisenberg antiferromagnetic (AFM) model on a triangular lattice with Dzyaloshinskii-Moriya interaction in the presence of the external magnetic field. We focus on the skyrmion lattice phase (SkX), which in the pure model is known to be stabilized in a quite wide temperature-field window. We aim to confront the effect of impurities on the SkX phase in the present frustrated AFM model with that in the nonfrustrated ferromagnetic counterpart as well as to consider more realistic conditions in the proposed experimental realizations of the present model. We show, that up to a fairly large concentration of the impurities, $p \approx 35\%$, the SkX phase can survive albeit in somewhat distorted form. Distortion of the SkX phase due to formation of bimerons, reported in the ferromagnetic model, was not observed in the present case.

## INTRODUCTION

Magnetic skyrmions, topologically nontrivial twisted magnetic spin configurations, have recently attracted a lot of attention due to the wide variety of properties, which make them promising candidates for the new generation of memory storage devices (Zhang et al. 2015b), logic gates (Zhang et al. 2015a), microwave detectors (Finocchio et al. 2015) and others. Similar to the well-known for decades one-dimensional topological objects - domain walls - and two-dimensional magnetic vortexes, skyrmions carry a certain property called topological charge or topological number, which sets them apart from the topologically trivial spin textures like ferromagnetic (FM) or antiferromagnetic (AFM) order and distinguishes one topological object from another. The idea of the existence of topologically-nontrivial structures in magnetic materials was actively theoretically developed in the end of the previous century (Belavin and Polyakov 1975; Bogdanov and Yablonskii 1989; Roessler et al. 2006), but it was not until 2009 when the first direct experimental proof of the presence of the hexagonal skyrmion crystal phase in a bulk ferromagnet was obtained in 2009 by Mühlbauer et al. (2009). After that the search for new skyrmion-hosting materials began in order to identify most promising materials for technological implementation. They were found in Hall ferromagnets, ferromagnetic monolayers, multilayers and ferrimagnets.

Stability of skyrmions is guarantied by topological charge as it is an invariant. However, in real materials it is not absolute and thermal fluctuations can bring the system over the finite energy barrier separating one spin configuration from another. The reason for the stabilization of the skyrmion lattice (SkX) state is usually the presence of Dzyaloshinskii-Moriya interaction (DMI) (Dzyaloshinsky 1958; Moriya 1960), which breaks the inversion symmetry. Nevertheless, there are some other mechanisms, that can lead to the formation of skyrmions, among which of particular interest are frustrated interactions. It was recently demonstrated by Okubo et al. (2012), that such interactions are capable of stabilizing both skyrmion and antiskyrmion crystals on any lattices of the trigonal symmetry with next-nearest interactions. Although skyrmions were first encountered in FM materials, recently the focus shifted to the alternatives (Barker and Tretiakov 2016; Bessarab et al. 2019), that were proven to be capable of hosting the SkX phase. It was shown by Rosales et al. (2015) and Osorio et al. (2017), that SkX can be stabilized in the classical Heisenberg AFM on triangular lattice with moderate DMI in a quite wide temperature-field window due to the combined effect of the frustration and the DMI. Further studies demonstrated the possibility of the SkX phase stabilization even at very small values of DMI (Mohylna and Žukovič 2020; Mohylna et al. 2021).

The presence of nonmagnetic impurities (spin vacancies) is a common feature in magnetic solids. In the case of frustrated spin systems with a ground-state degeneracy the problem of collective impurity behaviour can become rather nontrivial due to a possible "order by quenched disorder" effect with a profound impact on the phase diagram. In particular, for the classical Heisenberg AFM on a triangular lattice in an external magnetic field but without DMI it has been shown that competition between thermal fluctuations and nonmagnetic impurities leads to a complicated temperature-field phase diagram with the emergence of a conical state at low temperatures (Maryasin and Zhitomirsky 2013). Considerable effect of nonmagnetic impurities has also been observed in the nonfrustrated FM Heisenberg model on a square lattice in the field with DMI (Silva et al. 2014), which displays the SkX phase. In particular, it was found that even very tiny concentrations of the vacancies induce the formation of bimerons in both helical (HL) and SkX states. In the considered system they show up as elongated configurations similar to a skyrmion with its disk-shaped central core

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shared by two half disks separated by a rectangular stripe domain. The presence of bimerons is found to cause deformation of both the HL and SkX states. While in the former case it occurs due to their appearance between the vacancies, thus breaking stripe-domain structures, in the latter case bimerons make the skyrmion positions in the skyrmion lattice change in a nontrivial way and decrease their overall number. The nonmagnetic impurities thus distort both the skyrmion configuration and the skyrmion lattice.

Monte Carlo (MC) simulations are among the most powerful tools in studying the behaviour of the magnetic materials at finite temperatures and, in particular, identifying the most promising candidates for the skyrmion-hosting environment. It is very important, though, to emulate the real materials used in experiments, as closely as possible. Although some studies on the Heisenberg AFM with DMI already attempted to introduce some more realistic features, such as the presence of a single-ion anisotropy, (Fang et al. 2021; Mohylna and Žukovič 2022) and quantum effects (Liu et al. 2020), the effect of nonmagnetic impurities, very common in real materials, remains to be investigated. In this work we study the influence of nonmagnetic impurities on the stability of the SkX phase in the frustrated classical Heisenberg AFM on a triangular lattice in the presence of the DMI by means of the hybrid Monte Carlo simulations. Since the problem is computationally very demanding and allows for parallelization, the simulations are implemented on a highly parallelized architecture of GPU using CUDA programming language.

## MODEL AND METHOD

We investigate the classical Heisenberg AFM on a triangular lattice with the following Hamiltonian

$$\mathscr{H} = -J\sum_{\langle i,j\rangle}\vec{S}_i\cdot\vec{S}_j + \sum_{\langle i,j\rangle}\vec{D}_{ij}\cdot\left[\vec{S}_i\times\vec{S}_j\right] - h\sum_i S_i^z, \tag{1}$$

where $\vec{S}_i$ is a classical unit-length Heisenberg spin at the $i$th site, $J<0$ is the AFM exchange coupling constant, $h$ is the external magnetic field applied perpendicular to the lattice plane (along the $z$ direction) and $\langle i,j\rangle$ denotes the sum over nearest-neighbour spins. $\vec{D}_{ij}$ is the DMI vector whose orientation is defined by the crystal symmetries. In this study it is chosen to point along the radius-vector $\vec{r}_{ij} = \vec{r}_i - \vec{r}_j$ between two neighbouring sites, i.e., $\vec{D}_{ij} = D\frac{\vec{r}_{i,j}}{|\vec{r}_{i,j}|}$ (Fig. 1), which results into the formation of the Bloch-type skyrmions. The magnitude of the parameter $D$ defines the strength of the DMI. The presence of nonmagnetic impurities is simulated by randomly replacing a certain percentage $p$ of spins on the lattice with vacancies. In the following we set $J=-1$ to fix the energy scale and absorb the Boltzmann constant in temperature by setting its value to $k_B = 1$.

In order to identify the presence of the SkX phase we use the skyrmion chirality, a discretization of a continuum topological charge (Berg and Lüscher 1981), which reflects the number and the nature of topological objects present in the system. The topological charge of a single skyrmion is $\pm1$ for the core magnetization $\pm|\vec{S}|$ (Zhang et al. 2020). The skyrmion chirality $\kappa$ and the corresponding susceptibility $\chi_\kappa$ are defined as follows:

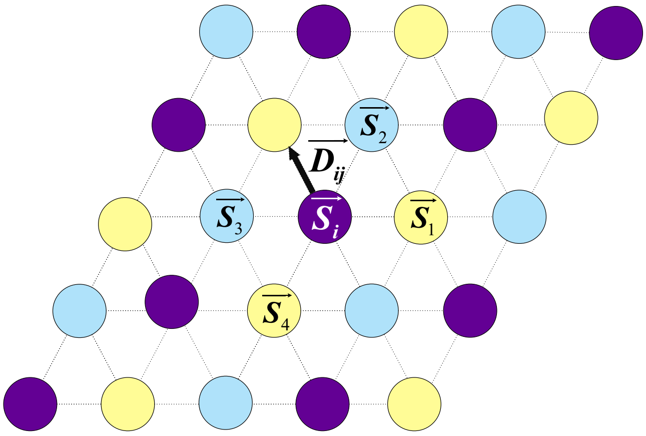

Fig. 1. Three-sublattice decomposition of the triangular lattice shown by different colors. $\vec{S}_i$ is the central spin, $\vec{S}_1,\ldots,\vec{S}_4$ are the spins involved in calculation of the local chirality, and $\vec{D}_{ij}$ represents the Dzyaloshinskii-Moriya vector.

$$\kappa = \frac{\langle K\rangle}{N} = \frac{1}{8\pi N}\left\langle\sum_i\left(\kappa_i^{12}+\kappa_i^{34}\right)\right\rangle, \tag{2}$$

$$\chi_\kappa = \frac{\langle K^2\rangle - \langle K\rangle^2}{NT}, \tag{3}$$

where $\kappa_i^{ab} = \vec{S}_i\cdot[\vec{S}_a\times\vec{S}_b]$ is the chirality of a triangular plaquette of three neighbouring spins (area of the triangle spanned by those spins) and $\langle\cdots\rangle$ denotes the thermal average. The chirality is calculated for the whole lattice and the summation runs through all the spins with $\{\vec{S}_a,\vec{S}_b\}$ corresponding to $\{\vec{S}_1,\vec{S}_2\}$ and $\{\vec{S}_3,\vec{S}_4\}$ in Fig. 1. Spins are taken in counter-clockwise fashion to keep the sign in accordance with the rules in (Berg and Lüscher 1981). For construction of the phase diagram it is useful to calculate some other basic thermodynamic quantities, such as the magnetization $m$, the magnetic susceptibility $\chi_m$, and the specific heat $c$, as follows:

$$m = \frac{\langle M\rangle}{N} = \frac{1}{N}\left\langle\sum_i S_i^z\right\rangle, \tag{4}$$

$$\chi_m = \frac{\langle M^2\rangle - \langle M\rangle^2}{NT}, \tag{5}$$

$$c = \frac{\langle\mathscr{H}^2\rangle - \langle\mathscr{H}\rangle^2}{NT^2}. \tag{6}$$

In order to identify the presence of the skyrmion phase and construct the phase diagrams we implement the hybrid Monte Carlo (HMC), which combines the standard Metropolis algorithm with the over-relaxation (OR) method (Creutz 1987). The OR method is a deterministic energy preserving perturbation method, which leads to the faster relaxation of the system due to faster decorrelation. To perform configurational averaging we run independent simulations on 50 replicas with different configurations of randomly distributed nonmagnetic impurities for each value of the impurity concentration $p$. In simulations we use $2\cdot10^6$ MC sweeps, half of which are used for the equilibration. The lattice size in all the simulations is $L=48$ and periodic boundary conditions are implemented. Due to high

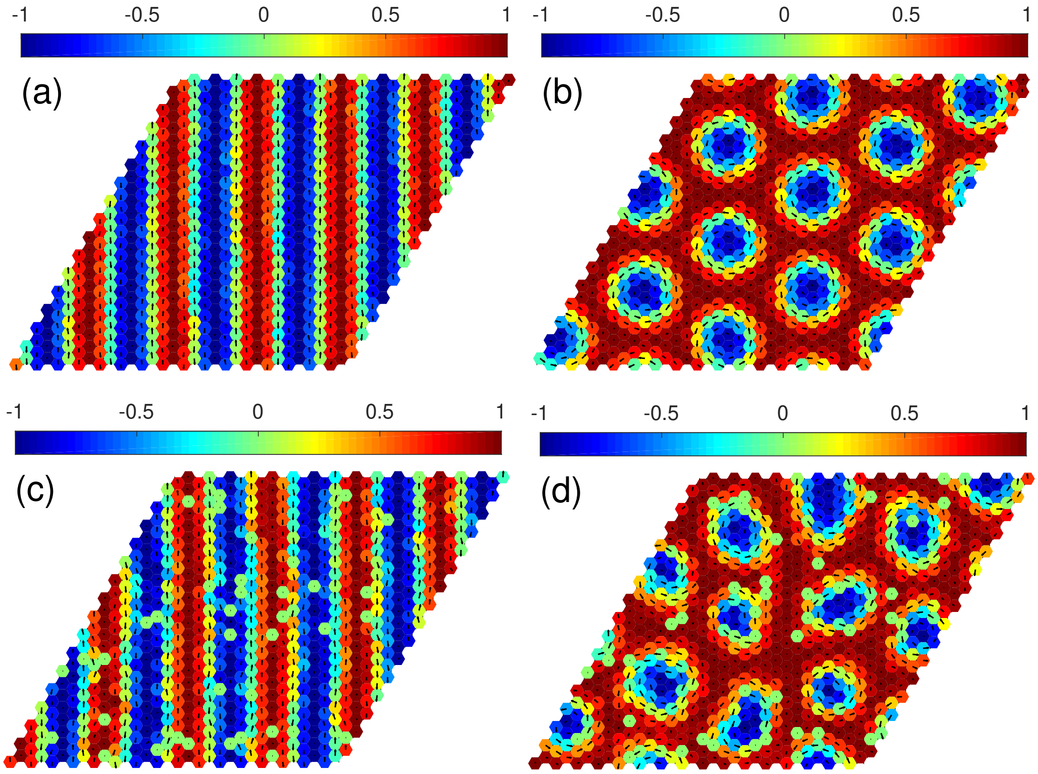

Fig. 2. (a,c) HL states at $h = 1.6$ and (b,d) SkX states at $h = 2.4$ for (a,b) pure system and (c,d) with $p = 5\%$ of vacancies. The remaining parameters are $T = 0.01$, $D = 0.5$, and $L = 48$. The green circles in (c,d) represent the vacancy locations. The snapshots are taken for one of the replicas.

computational demands the simulations are carried out on General Purpose Graphical Processing Units (GPGPU) using CUDA, which allows for the massive parallelization of the calculations.

## RESULTS

### Pure model

The frustrated Heisenberg AFM with DMI has been previously intensively studied in a wide parameter space (Rosales et al. 2015; Mohylna and Žukovič 2020; Mohylna et al. 2021). The phase diagram of a pure isotropic model with a moderate DMI ($0.2 < D < 1$) consists of three distinct phases: the helical (HL) phase with chiral stripes rotating in the $x-y$ plane; the skyrmion lattice (SkX) phase, which consists of three interpenetrating skyrmion lattices on each of the sublattices and the V-like (VL) phase (Mohylna et al. 2021). The typical snapshots of the HL and the SkX phases on one of the sublattices in the case of no impurities are depicted in Fig. 2 (a) and (b), respectively. The colours represent the values of the $z-$component: the red spins are those pointing along the external magnetic field and the blue ones pointing opposite to it. The arrows show the projections of the spins to the $x-y$ plane. The representative phase diagram for the model without impurities with $D = 0.5$ is shown in Fig. 3 in black circles. The SkX phase occupies a relatively big part of the $T-h$ plane and emerges at the fields around $h \approx 0.24$ for the lowest temperature, which is reflected in a sharp increase of the chirality signaling the first-order phase transition (Rosales et al. 2015), as also shown in Fig. 4(a). The increase of temperature results in some distortion of the skyrmions' profile and consequently it leads to smoothing of the chirality and the change of the transition type to the second-order one (Mohylna et al. 2021).

### Effect of impurities

To simulate the presence of nonmagnetic impurities in our system we randomly replaced $p\%$ of the sites with vacancies and studied their effect on the HL and SkX states. In Fig. 2 we present the respective states in the pure system (a,b) and with $p = 5\%$ of nonmagnetic impurities (c,d) on one of the three interpenetrating sublattices in the HL phase (a,c) and the SkX phase (b,d). The snapshots for one of the running replicas are shown. We can observe that, compared to the pure systems, the presence of the vacancies naturally results in some distortion of the respective phases. Nevertheless, there are no signs of the formation of bimerons, as it was the case in the FM model (Silva et al. 2014). We believe that the present AFM system is more resilient against creation of bimerons than the FM one due to the fact that both HL and SkX textures are formed on each of the three interpenetrating sublattices. Therefore, for example the stripe-domain structures in the HL state are more robust and it is more difficult for nonmagnetic impurities to break them into pieces (bimerons) than in the FM system. In the SkX phase, one can notice that, besides the skyrmion distortion, the presence of vacancies also reduces their number. It is interesting to study their mutual interaction. It is known that isolated skyrmions in isotropic 2D Heisenberg magnets are attracted to the spin vacancies (Subbaraman et al. 1998; Pereira and Pires 2003), which allows an impurity to be at a skyrmion center. On the other hand, skyrmions in a skyrmion crystal do not generally appear centered at vacancies. In such

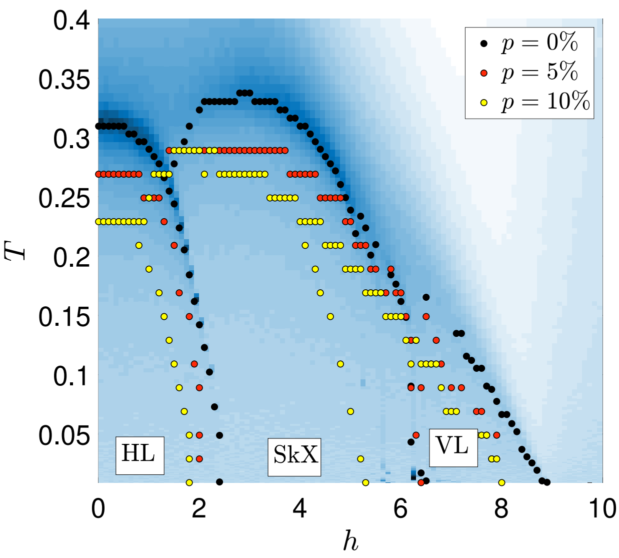

Fig. 3. Phase diagrams of the Heisenberg AFM model in $T - h$ plane with $D = 0.5$ and different $p$. The black, red and yellow circles represent the cases with $p = 0\%$, $5\%$ and $10\%$, respectively.

case it is energetically more favorable if configurations involve the whole crystal instead of isolated skyrmions and, therefore, the skyrmion lattice is formed in such a way that the spin vacancies become, in general, localized between skyrmions. This phenomenon can be also observed in the present case (see Fig. 2(d)) when the skyrmions have a tendency to rearrange in such way so that the vacancies stay at their outskirts.

In Fig. 4 we plot field and temperature dependencies of the chirality for selected values of the concentration of nonmagnetic vacancies $p$. The presence of impurities leads to distortion of the skyrmion profiles and consequently reduction of the magnitude of the chirality. At the same time it smears out its abrupt change, particularly at the HL-SkX phase transition, with smaller but finite values within the higher-field part of the HL phase (see Fig. 4(a)). The effect of the chirality reduction with the increasing concentration of the nonmagnetic impurities is also demonstrated in Fig. 4(b), in which the chirality is plotted versus temperature for a wide range of the concentration $p$. Similar effect of distortion of the skyrmion profiles and reduction of the chirality is also produced by thermal fluctuations. Our rough estimate of the critical threshold of the magnetic dilution below which no skyrmions can survive even at the lowest temperatures is $p_c \approx 35\%$. This value is in a good correspondence with the value estimated for the FM model on the square lattice (Silva et al. 2014).

The phase diagrams for $p = 5\%$ (red circles) and $p = 10\%$ (yellow circles) are presented in Fig. 3. Their overall topology remains unchanged as compared to the case of the pure model and consists of the HL phase in the low-field region, the VL phase in the high-field region and the SkX phase sandwiched between them. However, a tendency of the SkX phase to shrink and smear with the increasing number of impurities can be noticed. Nevertheless, the area of the SkX phase still occupies a big part of a phase diagram even for $p = 10\%$.

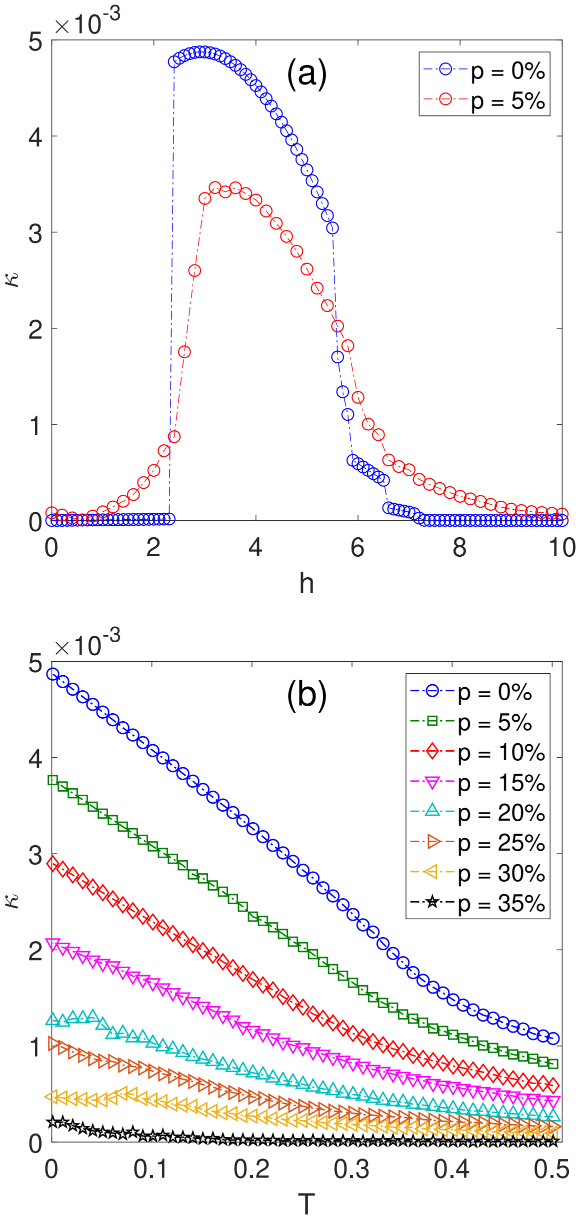

Fig. 4. (a) Field dependence of the chirality at $T = 0.01$ and (b) temperature dependence of the chirality at $h = 3.4$, for selected values of the concentration of nonmagnetic vacancies $p$.

## CONCLUSIONS

In this study we investigated the effect of the presence of nonmagnetic impurities in the frustrated Heisenberg AFM model with the DMI, hosting the SkX phase. The purpose was to confront the effects of impurities on the SkX phase in the present frustrated AFM model with that in the nonfrustrated FM model (Silva et al. 2014) and also to take into account a more realistic situation in the possible experimental realizations of the present model (Fang et al. 2021). We focused on the influence of impurities on the shape, size and position of individual skyrmions on the lattice as well as overall stability of the SkX phase. Our findings suggest, that both the HL and the SkX phases in the present frustrated AFM system are more robust to the distortion, induced by those impurities, than in the nonfrustrated FM counterpart, where the formation of bimerons occurs already at low concentrations of $p \approx 1\%$. In our case the skyrmion lattice is formed in such a way, that the vacancies become localized in-between the skyrmions. The effect caused by the presence of impurities is similar to the one caused by thermal fluctuations: the skyrmions shape is distorted and the chirality is both reduced and smeared out, in particular at the HL-SkX phase boundary. This tendency becomes more pronounced with the increasing percentage of the impurities. We estimate that the threshold below which no SkX phase can be stabilized is $p_c \approx 35\%$.

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## REFERENCES

Adam, G., Bashashin, M., Belyakov, D., Kirakosyan, M., Matveev, M., Podgainy, D., Sapozhnikova, T., Streltsova, O., Torosyan, S., Vala, M., et al. (2018). It-ecosystem of the hybrilit heterogeneous platform for high-performance computing and training of it-specialists. In *English, in CEUR Workshop Proceedings, V. Korenkov, A. Nechaevskiy, T. Zaikina, and E. Mazhitova, Eds*, volume 2267, pages 638–644.

Barker, J. and Tretiakov, O. A. (2016). Static and dynamical properties of antiferromagnetic skyrmions in the presence of applied current and temperature. *Physical review letters*, 116(14):147203.

Belavin, A. and Polyakov, A. (1975). Metastable states of two-dimensional isotropic ferromagnets. *JETP lett*, 22(10):245–248.

Berg, B. and Lüscher, M. (1981). Definition and statistical distributions of a topological number in the lattice o (3) $\sigma$-model. *Nuclear Physics B*, 190(2):412–424.

Bessarab, P., Yudin, D., Gulevich, D., Wadley, P., Titov, M., and Tretiakov, O. A. (2019). Stability and lifetime of antiferromagnetic skyrmions. *Physical Review B*, 99(14):140411.

Bogdanov, A. and Yablonskii, D. (1989). Thermodynamically stable "vortices" in magnetically ordered crystals. the mixed state of magnets. *Zh. Eksp. Teor. Fiz*, 95(1):182.

Creutz, M. (1987). Overrelaxation and monte carlo simulation. *Physical Review D*, 36(2):515.

Dzyaloshinsky, I. (1958). A thermodynamic theory of "weak" ferromagnetism of antiferromagnetics. *Journal of physics and chemistry of solids*, 4(4):241–255.

Fang, W., Raeliarijaona, A., Chang, P.-H., Kovalev, A. A., and Belashchenko, K. D. (2021). Spirals and skyrmions in antiferromagnetic triangular lattices. *Physical Review Materials*, 5(5):054401.

Finocchio, G., Ricci, M., Tomasello, R., Giordano, A., Lanuzza, M., Puliafito, V., Burrascano, P., Azzerboni, B., and Carpentieri, M. (2015). Skyrmion based microwave detectors and harvesting. *Applied Physics Letters*, 107(26):262401.

Liu, Z., dos Santos Dias, M., and Lounis, S. (2020). Theoretical investigation of antiferromagnetic skyrmions in a triangular monolayer. *Journal of Physics: Condensed Matter*, 32(42):425801.

Maryasin, V. and Zhitomirsky, M. (2013). Triangular antiferromagnet with nonmagnetic impurities. *Physical review letters*, 111(24):247201.

Mohylna, M., Buša Jr, J., and Žukovič, M. (2021). Formation and growth of skyrmion crystal phase in a frustrated heisenberg antiferromagnet with dzyaloshinskii-moriya interaction. *Journal of Magnetism and Magnetic Materials*, 527:167755.

Mohylna, M. and Žukovič, M. (2020). Emergence of a skyrmion phase in a frustrated heisenberg antiferromagnet with dzyaloshinskii-moriya interaction. *Acta Physica Polonica, A.*, 137(5).

Mohylna, M. and Žukovič, M. (2022). Stability of skyrmion crystal phase in antiferromagnetic triangular lattice with dmi and single-ion anisotropy. *Journal of Magnetism and Magnetic Materials*, 546:168840.

Moriya, T. (1960). Anisotropic superexchange interaction and weak ferromagnetism. *Physical review*, 120(1):91.

Mühlbauer, S., Binz, B., Jonietz, F., Pfleiderer, C., Rosch, A., Neubauer, A., Georgii, R., and Böni, P. (2009). Skyrmion lattice in a chiral magnet. *Science*, 323(5916):915–919.

Okubo, T., Chung, S., and Kawamura, H. (2012). Multiple-q states and the skyrmion lattice of the triangular-lattice heisenberg antiferromagnet under magnetic fields. *Physical review letters*, 108(1):017206.

Osorio, S. A., Rosales, H. D., Sturla, M. B., and Cabra, D. C. (2017). Composite spin crystal phase in antiferromagnetic chiral magnets. *Physical Review B*, 96(2):024404.

Pereira, A. and Pires, A. (2003). Solitons in the presence of a static spin vacancy on a 2d heisenberg antiferromagnet. *Journal of magnetism and magnetic materials*, 257(2-3):290–295.

Roessler, U. K., Bogdanov, A., and Pfleiderer, C. (2006). Spontaneous skyrmion ground states in magnetic metals. *Nature*, 442(7104):797–801.

Rosales, H. D., Cabra, D. C., and Pujol, P. (2015). Three-sublattice skyrmion crystal in the antiferromagnetic triangular lattice. *Physical Review B*, 92(21):214439.

Silva, R., Secchin, L., Moura-Melo, W., Pereira, A., and Stamps, R. (2014). Emergence of skyrmion lattices and bimerons in chiral magnetic thin films with nonmagnetic impurities. *Physical Review B*, 89(5):054434.

Subbaraman, K., Zaspel, C., and Drumheller, J. E. (1998). Impurity-pinned solitons in the two-dimensional antiferromagnet detected by electron paramagnetic resonance. *Physical review letters*, 80(10):2201.

Zhang, X., Ezawa, M., and Zhou, Y. (2015a). Magnetic skyrmion logic gates: conversion, duplication and merging of skyrmions. *Scientific reports*, 5(1):1–8.

Zhang, X., Zhao, G., Fangohr, H., Liu, J. P., Xia, W., Xia, J., and Morvan, F. (2015b). Skyrmion-skyrmion and skyrmion-edge repulsions in skyrmion-based racetrack memory. *Scientific reports*, 5(1):1–6.

Zhang, X., Zhou, Y., Song, K. M., Park, T.-E., Xia, J., Ezawa, M., Liu, X., Zhao, W., Zhao, G., and Woo, S. (2020). Skyrmion-electronics: writing, deleting, reading and processing magnetic skyrmions toward spintronic applications. *Journal of Physics: Condensed Matter*, 32(14):143001.

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