Tunable magnetic helicity in Mn$_{1-x}$Fe$_x$Ge: A Monte Carlo simulation

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Motivated by recent experiments on B20 compound Mn$_{1-x}$Fe$_x$Ge, we explore the variations of spin structures and magnetic helicity $\gamma_m$ in Mn$_{1-x}$Fe$_x$Ge as a function of concentration $x$, using Monte Carlo simulation. We propose a simple spin model including hybrid Dzyaloshinskii-Moriya (DM) interactions with different signs in FeGe and MnGe. The results reveal a series of spin structures with helicity $\gamma_m$ varying continuously from negative values to positive values upon decreasing $x$ ($0.0 \leq x \leq 1.0$). The simulated $x$-dependence of the spin structures is consistent with experimental observations, suggesting the validity of the present hybrid model. This study sheds light on control of magnetic helicity in helimagnets, by mixing crystals with different DM interactions. © 2015 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4918632]

Chirality plays an important role in condensed matters. One example of chiral objects is the non-centrosymmetric B20-type helimagnets with space group $P2_13$, such as MnSi, Fe$_{1-x}$Co$_x$Si, and FeGe. Among these helimagnets, MnSi and Fe$_{1-x}$Co$_x$Si show a helical (proper screw) spin structure at low temperature ($T$), as a result of the competition between ferromagnetic (FM) exchange interactions and antisymmetric Dzyaloshinsky-Moriya (DM) interaction. On the other hand, a perpendicular external magnetic field ($H$) may transform a single twisted helical to an energetically favorable magnetic skyrmion (vortex-like spin-swirling structure). This intriguing topological structure has been receiving tremendous interest due to its unusual magnetic and transport properties, especially the topological Hall effect.

Handedness of the atomic configuration (crystalline chirality $\Gamma_c$) in these B20-type helimagnets has been examined long ago by convergent beam electron diffraction. These compounds exhibit the left- and right-handed chiral configurations, both of which carry the same formation energy. Regarding the helical and skyrmion structures, there exists an unique spin swirling direction defined as the spin helicity $\gamma_m$. Systematic studies showed that the handedness of crystalline chirality $\Gamma_c$ is closely related to that of $\gamma_m$ via different signs of the DM interaction. Specifically, the Mn-based compounds (Mn$_{1-x}$Fe$_x$Si and Mn$_{1-x}$Co$_x$Si) show the positive $\Gamma_c \times \gamma_m$ (+) correlation, while the Fe-based ones (Fe$_{1-x}$Co$_x$Si) exhibit the opposite $\Gamma_c \times \gamma_m$ (−) correlation (where we define the left and right handedness of $\Gamma_c$ or $\gamma_m$ as − and +, respectively). It means that Mn and Fe based compounds possess different signs of the DM interaction for $\Gamma_c$ with the same handedness. Motivated by this interesting characteristic, earlier studies on Mn$_{1-x}$Fe$_x$Si family target at tuning $\gamma_m$ by varying the Mn/Fe ratio. However, unfortunately, the pure FeSi is not magnetically ordered, so this material does not possess a good controllable capability of $\gamma_m$. Recently, the B20 compound Mn$_{1-x}$Fe$_x$Ge provides a good platform to track the variation of $\gamma_m$. Both the pure compounds of FeGe and MnGe were previously demonstrated to be magnetically ordered, and experimental evidence for $x$-dependence of spin structure in Mn$_{1-x}$Fe$_x$Ge compounds was recently reported, where $\gamma_m$ can be altered in response to variation of the Mn/Fe ratio. Also, it was observed in Mn$_{1-x}$Fe$_x$Ge that the skyrmion size and its helicity are dependent of $x$.

On the other hand, recent experiments on B20-type helimagnet MnGe showed that the field dependence of the topological Hall effect is very distinct from that observed in other B20-type compounds like MnSi and Fe$_{1-x}$Co$_x$Si, suggesting an alternative structure of skyrmion lattice as the zero-field ground state. This state is very different from the often observed hexagonal skyrmion crystal (SkX) in MnSi and Fe$_{1-x}$Co$_x$Si. Small-angle neutron scattering (SANS) experiments revealed a strong spin-orbit coupling (SOC) accompanying with an easy axis of magnetization along the (100) direction. Considering these experimental evidences, an alternating half-skyrmion (HSk) and half-anti-skyrmion crystal (HASK) structure on a square lattice, as a candidate for the zero-field ground state of MnGe, is suggested. In this case, a compass-type anisotropy arising naturally from the strong SOC plays a key role in determining this exotic spin structure.

In this work, we explore the variations of magnetic helicity and spin structure in Mn$_{1-x}$Fe$_x$Ge compounds as a function of $x$, using Monte Carlo (MC) simulation. We propose a simple spin model with hybrid DM interaction taking account of the different DM interaction signs in FeGe and MnGe, respectively. The results indicate a series of spin states (e.g., helical and skyrmion states), with $\gamma_m$ varying continuously from negative values to positive ones, upon decreasing $x$. This study provides a theoretical guide to understand the control of magnetic helicity $\gamma_m$ and spin structure in helimagnets, by mixing crystals with different DM interactions, which can be generally considered as the crystal engineering of the spin structure and magnetic helicity.

We first consider a thin film of FeGe described by Hamiltonian including the FM exchange interaction ($J_{Fe}$).
and DM interaction (\(D_{\text{Fe}}\)) on a two-dimensional (2D) square lattice:

\[
\mathcal{H}_{\text{Fe}} = -J_{\text{Fe}} \sum_i S_i \cdot (S_i \hat{x} + S_{i+\hat{y}})
- D_{\text{Fe}} \sum_i (S_i \times S_{i+\hat{x}} \cdot \hat{x} + S_i \times S_{i+\hat{y}} \cdot \hat{y}),
\]

where \(S_i = (S^x_i, S^y_i, S^z_i)\) denotes a classical Heisenberg spin at site \(i\) and \((\hat{x}, \hat{y}, \hat{z})\) are the basis vectors of the simple cubic lattice.

For helimagnet MnGe, we consider that its outermost electrons are approximately localized in the Mott insulator regime, which can be described by an effective spin model including the FM exchange interaction (\(J_{\text{Mn}}\)), DM interaction (\(D_{\text{Mn}}\)), and compass anisotropy (\(A_{\text{Mn}}\)) following the previous studies:

\[
\mathcal{H}_{\text{Mn}} = -J_{\text{Mn}} \sum_i S_i \cdot (S_i \hat{x} + S_{i+\hat{y}})
- D_{\text{Mn}} \sum_i (S_i \times S_{i+\hat{x}} \cdot \hat{x} + S_i \times S_{i+\hat{y}} \cdot \hat{y})
- A_{\text{Mn}} \sum_i (S^x_i S^y_{i+\hat{x}} + S^y_i S^x_{i+\hat{y}}),
\]

In Eqs. (1) and (2), \(D_{\text{Fe}} < 0\) and \(D_{\text{Mn}} > 0\) yield the left-handed and right-handed helical structures, respectively, in FeGe and MnGe. Therefore, regarding \(\text{Mn}_1-x\text{Fe}_x\text{Ge}\) compounds, we propose a simple spin model on a 2D square lattice with hybrid DM interaction as

\[
\mathcal{H} = -J \sum_i S_i \cdot (S_i \hat{x} + S_{i+\hat{y}})
- D \sum_i (S_i \times S_{i+\hat{x}} \cdot \hat{x} + S_i \times S_{i+\hat{y}} \cdot \hat{y})
- A \sum_i (S^x_i S^y_{i+\hat{x}} + S^y_i S^x_{i+\hat{y}}) - H \sum_i S_i,
\]

In the simulations, we distribute the Mn and Fe species randomly on a 2D square lattice with a given Fe concentration \(x\). Here, \(H\) represents the magnetic field orienting along the z-axis, and parameters \(J, D,\) and \(A\) scale the effective strengths of the FM exchange interaction, DM interaction, and compass anisotropy between neighboring species, respectively. In addition, we assume that the effective \(J, D,\) and \(A\) between neighboring Mn-Fe pairs as \(J = (J_{\text{Mn}} + J_{\text{Fe}})/2, D = (D_{\text{Mn}} + D_{\text{Fe}})/2,\) and \(A = A_{\text{Mn}}/2\), respectively. Here, we must clarify that this treatment lacks rigorous theoretical foundation. However, from the phenomenological viewpoint, it seems reasonable that the effective \(J, D,\) and \(A\) may fall in between the corresponding values for Mn and Fe spins, although the actual mechanism could be more complex which is yet an open question. Moreover, we assume that the effective \(J, D,\) and \(A\) between neighboring Mn-Mn or Fe-Fe pairs remain identical to that set in the prototypical Hamiltonian for MnGe or FeGe compound.

Further, we estimate the values of \(J_{\text{Fe}}, D_{\text{Fe}}, J_{\text{Mn}}, D_{\text{Mn}},\) and \(A_{\text{Mn}}\) according to experimental results. Earlier neutron scattering studies of FeGe showed that the helical spin propagates along the equivalent \(\langle 100 \rangle\) directions at temperature \(T_N = 278.7\) K under zero-magnetic field, while the propagation direction changes to the equivalent \(\langle 111 \rangle\) directions at \(T_{J_2} = 211\) K and \(T_{J_2} = 245\) K upon the temperature decreasing and increasing, respectively.\(^{11}\) For MnGe, the helical spin structure forms below transition point \(T_N = 170\) K.\(^{5}\) As the exchange constant is proportional to the \(T_N,\) we may estimate that \(J_{\text{Fe}}/J_{\text{Mn}} \sim 278\) K/170 K \(\sim 1.6.\) In addition, the helix wavelength is \(\lambda = 30-60\) Å, equivalent to \(8-12\) Å for MnGe, and \(\lambda = 683-700\) Å, equivalent to \(\sim 150\) Å for FeGe, where \(\lambda\) is lattice constant (\(\sim 4.7\) Å) for both MnGe and FeGe.\(^{5,13,14}\) We can estimate from the relation \(D/J \sim \sqrt{2}\tan(2\pi/\lambda)\) that \(D\) is one order of magnitude smaller than that of \(J\) for FeGe. However, we choose a big \(D/J (D_{\text{Mn}}/J_{\text{Mn}} = 0.6\) and \(D_{\text{Fe}}/J_{\text{Fe}} = -0.9)\) in the current simulation to reduce the helical wavelength and skyrmion radius, in order to save the calculation resources. For simplicity, we set \(J_{\text{Mn}} = 1, D_{\text{Mn}} = 0.6, J_{\text{Fe}} = 1.6,\) and \(D_{\text{Fe}} = -1.44\) in the calculation. Following the previous study,\(^{13}\) \(A_{\text{Fe}} = 0\) and \(A_{\text{Mn}} = 6.0\) are taken, which may generate an alternating HSk and HASk spin structure for MnGe under zero-magnetic field. In addition, the effective spin moments per atom in MnGe and FeGe are \(2.3\mu_B\) and \(1.0\mu_B,\) respectively, allowing \(|S_{\text{Mn}}| = 2.3\) and \(|S_{\text{Fe}}| = 1.0\) in the simulations. It is noted that setting \(|S_{\text{Mn}}| = |S_{\text{Fe}}| = 1.0\) or 2.3 does not change the conclusion qualitatively.

We perform MC simulations based on the standard single-flip Metropolis algorithm combined with the over-relaxation method.\(^{19}\) This clustering algorithm is believed to be effective in equilibrating the frustrated spin models with a large size.\(^{19,20}\) Our unit MC step consists of one Metropolis sweep and ten over-relaxation sweeps. Most calculations are carried out on a square \(32 \times 32\) lattice with periodic boundary conditions, and the results are further checked on a lattice with bigger size of \(64 \times 64\) to ensure the consistency. Meanwhile, all the calculations are carried out under a magnetic field \(H\) normal to the \(xy\)-plane of the 2D lattice, with a simulated annealing scheme: First, a paramagnetic lattice is chosen as the initial lattice and each simulation cycle starts from sufficiently high temperature. Then the lattice is cooled down gradually until a very low temperature \(T = 0.01,\) \(^{16,21}\) The as-obtained spin state is treated approximately as the ground state.

Fig. 1 shows snapshots of several typical \(\text{Mn}_{1-x}\text{Fe}_x\text{Ge}\) spin lattices, under a small magnetic field \(H = 0.16.\) To illustrate the three-components of the spin structure, we use the color map to scale the spin component along the \(z\)-axis (out-of-plane) \(S^z_i,\) and use the spin vector (arrow) to describe the in-plane components \(S^{xy}_i.\) At \(x = 1.0\) [Fig. 1(a)], the helical state propagates along the \(\langle 11 \rangle\) direction, coinciding with experimental observations on FeGe. Although the Swiss-roll-like vortex texture was also observed by Lorentz electron microscopy, it is considered to be induced by the sample edge effect.\(^{3,22}\) Therefore, the helical spin structure is indeed the ground state of FeGe. On the other hand, for \(x = 0.0\) [Fig. 1(f)], corresponding to MnGe, the ground state is a spin crystal structure from the superposition of two orthogonal helices. We call it the \(\text{SC}_2\) state hereafter following Refs. 13 and 16. The \(\text{SC}_2\) spin structure can be generally written as \(^{12,13,23}
where $q$ is the wavenumber of the helical structure, $m$ is the magnetization induced by $H$, and $I(\vec{x})$ is a normalized factor. As a special SC$_2$ state, the zero-field ground state of MnGe (i.e., the SC$_2$ state at $H = 0$) is identified as an alternating HSK and HASk crystal structure on a square lattice. Now, we analyze the variation of the spin order in Mn$_{1-x}$Fe$_x$Ge by tuning $x$. The Mn doping in FeGe leads to a deformed helical order at $x = 0.9$ [Fig. 1(b)] and the appearance of a ferromagnetic-like state at $x = 0.7$ [Fig. 1(c)], with almost all the spins orientating along the $[11]$ direction. At $x < 0.7$, it is seen that the SC$_2$ state grows gradually, and the size of the vortex-like structure reduces remarkably with decreasing $x$ [see Figs. 1(d)−1(f)].

Subsequently, we look at magnetic helicity $\gamma_m = \sum_{i<n_i} \mathbf{n}_i \cdot \mathbf{\tilde{x}} + n_i \times \mathbf{n}_{i \pm \hat{y}}$, where $n_i = S_i/S$, and the sign of $\gamma_m$ is connected with that of the DM interaction. Specifically, a positive (negative) $\gamma_m$ reflects the dominant right- (left-) handed helix. In experiments, the equivalent $\gamma_m$ is evaluated using the polarized neutron scattering or Lorentz transmission electron microscopy. Fig. 2(a) shows the $x$-dependent behaviors of $\langle \gamma_m \rangle$ obtained from the MC simulations, where $\langle \ldots \rangle$ refers to the statistical configuration averaging at a given $T$. It is noted that $\langle \gamma_m \rangle$ is a monotonically decreasing function upon increasing $x$, with $\langle \gamma_m \rangle > 0$ at $x < 0.7$ and $\langle \gamma_m \rangle < 0$ at $x > 0.7$, while $\langle \gamma_m \rangle \sim 0$ at $x \sim 0.7$. The sign reversal of $\langle \gamma_m \rangle$ is also revealed in the $\langle \gamma_m \rangle$-$T$ curves, as plotted in Fig. 2(b). The $\langle \gamma_m \rangle$-$T$ curve for $x = 0.7$ fluctuates around $\langle \gamma_m \rangle \sim 0$ as $T$ varies, while the curves for $x > 0.7$ and $x < 0.7$ lie in the regions of $\langle \gamma_m \rangle < 0$ and $\langle \gamma_m \rangle > 0$, respectively. These behaviors are consistent with the $T$-dependent ac-susceptibility $\chi$ observed on Mn$_{1-x}$Fe$_x$Ge with a weak magnetic field $H = 50$ mT. Especially, experimental results showed a typical Curie-Weiss behavior for $x = 0.75$, suggesting the ferromagnetic nature. Actually, $\chi$ is proportional to the magnetization $M$ as $\chi = M/H$. To compare with experimental results, we examine the dependence of $\langle M \rangle$ on $T$ for different $x$, as plotted in Fig. 2(c), where $\langle M \rangle = \langle (M_x)^2 + (M_y)^2 + (M_z)^2 \rangle^{1/2}$, $M_x = \langle 1/N \rangle \sum_i \langle M_i \rangle$, $x = y, z$, and $N$ is the total spin number. The ferromagnetic-like phase occurs at $x = 0.7$, as indicated by the $\langle M \rangle$-$T$ curve in Fig. 2(c). In addition, the other $\langle M \rangle$-$T$ curves for $x = 0.0$, 0.1, and 1.0 exhibit various kink-like shapes, associating with the $T$-driven phase transitions from the paramagnetic state to helical state. These curves are in accordance with experimental observation. However, we note that some differences exist in terms of the $x$-dependent behavior of $T_N$. In fact, the variation of the spin states can be easily understood considering the competition between positive $D_{mn}$ and negative $D_{Fe}$, which results in vanishing of the average DM interaction constant $D$ at $x \sim 0.7$. Reasonably, the appearance of ferromagnetic-like phase can be understood. Therefore, it is also reasonable that the spin structures at $x > 0.7$ have $\langle \gamma_m \rangle < 0$ due to the dominant negative $D_{Fe}$, and those at $x < 0.7$ have $\langle \gamma_m \rangle > 0$ due to the dominant positive $D_{mn}$.

The suppression of the single helical spin state by field $H$ may generate the SKX structure with hexagonal arrangement, which can be approximately visualized by simple

\begin{equation}
S(x, y) = I|S_m| \left[ (\sin(qy), 0, \cos(qy)) + (0, \cos(qx), \sin(qx)) + (0, 0, m) \right]
= I|S_m| \left[ \sin(qy), \cos(qx), \cos(qy) + \sin(qx) + m \right],
I \equiv I(x, y) = \frac{1}{\sqrt{\sin^2(qy) + \cos^2(qx) + (\cos(qy) + \sin(qx) + m)^2}},
\end{equation}

FIG. 1. A series of typical Mn$_{1-x}$Fe$_x$Ge spin lattices with various concentration $x$ under $H = 0.16$. The blue (black) arrows denote spin components $S_i^a$ of Fe (Mn) atoms.

FIG. 2. Plot of the relations of $\langle \gamma_m \rangle$-$x$, $\langle \gamma_m \rangle$-$T$, and $\langle M \rangle$-$T$ in Mn$_{1-x}$Fe$_x$Ge for various concentration $x$, under $H = 0.16$. 
superposition of three helical states with equal pitch length but a relative angle of $120^\circ$ on the plane perpendicular to $H$ (i.e., $xy$-plane here). In Fig. 3(a), the $x$ dependence of $\langle \gamma_m \rangle$ under $H = 0.48$ is demonstrated. Obviously, the $\langle \gamma_m \rangle$ changes continuously from negative values to positive values with decreasing $x$. Fig. 3(b) shows the SKX state for $x = 1.0$ (i.e., FeGe). A low doping of Mn atom in FeGe makes the lattice hard to retain the SKX state, but some isolated skyrmions with irregular shapes emerge, such as the spin configuration at $x = 0.9$ [Fig. 3(c)]. The spin structure first undergoes a change from a SKX state to a ferromagnetic-like state, with increasing skyrmion size $a_{SL}$ [see Figs. 3(c)–3(e)]. Then, further decrease of $x$ leads to the appearance of some vortex-like structures [Fig. 3(f)]. Subsequently, the lattice evolves gradually into regular $C_4$ symmetric SC$_2$ state [Fig. 3(g)]. It is noted that these variations of spin structure are consistent with experimental report.9

Indeed, the underlying physical nature for the magnetic phenomena in Mn$_{1-x}$Fe$_x$Ge compounds is complex.10 This complexity arises from the fact that the crystalline chirality $\Gamma_{ch}$ may have potential relevance with the magnetic helicity $\gamma_m$ in determining the magnetic phenomena such as the spin transfer torque effect25 or the nontrivial Mn moment tilting.26 It is also suggested that the interplay between exchange interactions of several magnetic shells could reverse the sign of the effective constant of DM interaction ($D_{xy}$) even if the microscopic DM interaction does not change.27 However, based on experimental data and our simulated results, we consider that one essential ingredient for the tunable $\gamma_m$ in such mixed-crystal systems is the different DM interaction signs for the two components. The tunable magnetic helicity of the spin structure in Mn$_{1-x}$Fe$_x$Ge is mainly caused by the continuous variation in the magnitude of the DM interaction in accompanying with its sign reversal in the mixed-crystal system.9,10 Therefore, we believe that the inhomogeneous crystals play an important role in tuning the magnetic helicity of the spin structures.

In summary, we have proposed a simple spin model to study the spin structures in Mn$_{1-x}$Fe$_x$Ge compounds, by taking account of the different DM interactions in FeGe and MnGe. It has been demonstrated that the magnetic helicity $\gamma_m$ and sizes of the skyrmion patterns can be altered by varying the Mn to Fe ratio. The results are consistent with experimental observations and the simulation scheme captures the main physics for spin ordering sequence in Mn$_{1-x}$Fe$_x$Ge compounds. This study sheds light on control of magnetic helicity and spin structure in helimagnets, by mixing crystals with different DM interactions.

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