Principal interactions in the helimagnetic system
Mn$_{1-y}$Fe$_y$Si

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Abstract. The magnetic structure of the noncentrosymmetrical cubic ferromagnets with Dzyaloshinskii-Moriya interaction Mn$_{1-y}$Fe$_y$Si with $y = 0, 0.06, 0.08,$ and $0.1$ has been studied by means of the small angle neutron diffraction and magnetization measurements. These compounds are shown to be ordered into the helix structure below the critical temperature $T_C$ that decreases linearly with the Fe doping and approaches zero at $y \approx 0.13$. We build the $H - T$ (magnetic field – temperature) phase diagrams for each compound and interpret them taking into account the Bak-Jensen hierarchical model of the principal interactions. Following the concentration evolution of these principal interactions (such as the spin wave stiffness $A$, the Dzyaloshinskii constant $D$, and the gap $\Delta$ in spin wave spectrum) we show that $A$ resembles behavior of $T_C$ approaching zero at $y \approx 0.15$. Thus, it is the isotropic ferromagnetic exchange interaction that plays a dominating role in determination of the critical temperature $T_C$ of the compounds under study. The Dzyaloshinskii constant $D$ has been found to be the same either in the system under study and in the related compounds Fe$_{1-x}$Co$_x$Si.

The magnetic and transport properties of MnSi, the noncentrosymmetrical cubic magnet with the space group P2$_1$3, have been a subject of intensive investigations during the last forty years due to their peculiar and intriguing properties. Its magnetic structure orders below $T_C = 29$ K in a left-handed spiral along the $\langle 111 \rangle$ directions with a propagation vector $k = 0.36$ nm$^{-1}$ at $T = 4$ K [1, 2, 3]. The spin helicity had been understood as a result of the equilibrium between the isotropic ferromagnetic (IF) exchange interaction and the antisymmetric Dzyaloshinskii-Moriya (DM) exchange interaction caused by lack of a center of symmetry in Mn atomic arrangement [4, 5, 6, 7]. These two interactions are considered isotropic themselves but another weak anisotropic exchange (AE) interaction fixes a direction of the helix along one of the cube diagonals [5].

The magnetic structure under an applied magnetic field for MnSi [8, 9] and the similar system Fe$_{1-x}$Co$_x$Si [10, 11] was recently studied and interpreted within the theory [12] which takes into account the interactions mentioned above. The neutron diffraction experiments enable to provide an important parameters of the magnetic system: the wavevector of the structure $k$, the critical field of the transformation to the ferromagnetic phase $H_{C2}$ and appearence of the A-phase in the $H - T$ phase diagram of this compound. (A-phase is the phenomenon of the 90°-rotation of the
helix wave vector $k$ from the direction parallel to the field axis to the direction perpendicular to it in a small pocket of the $H - T$ phase diagram near $T_C$ in the narrow range of the fields $g \mu_B H_{\text{gap}} \sim \sqrt{2} \Delta$. This so-called $k$-flop phase will be minutely considered below in the text).

The Author of Ref. [12] developed the microscopic theory for the cubic magnets without center of symmetry under magnetic field. The ground state energy and the spin wave spectrum were evaluated showing, in accord with Ref. [5], that the system finds its equilibrium in the helical ordering with the helix wave vector

$$k = \frac{SD}{A}. \quad (1)$$

Here $D$ is the strength of DM interaction, $A$ is the spin wave stiffness at a distance much shorter than the helix period (it is a quantitative characteristic of the IF interaction), and $S$ is the average spin per a unit cell. The structure was also shown to be intrinsically unstable with respect to the small magnetic field applied perpendicular to $k$ unless it is stabilized via a small gap in the spin wave spectrum $\Delta$ which is caused by the DM interaction. Luckily for these systems, the spin wave interaction in presence of the DM interaction guarantees enhancement of a small but positive gap of order of $\Delta^2 \sim (D^2/2A)^2$. The existence of this gap can also provide an explanation for the appearance of A-phase [8]. Vise versa, the existence of the A-phase can be considered as an indirect proof for existence of the spin wave gap predicted by the theory [12].

The theory [12] is able to relate the spin wave stiffness $A$ and the spin wave gap $\Delta$ to the critical fields $H_{C2}$ and $H_{\text{gap}}$, respectively, through the following expressions

$$g \mu_B H_{C2} = Ak^2 \quad (2)$$

$$\Delta \simeq g \mu_B H_{\text{gap}} / \sqrt{2}. \quad (3)$$

Combining these expression with the definition of $k$ (Eq. (1)) one is able to estimate the principal interactions of these systems from the parameters of the magnetic structure measured in the experiment ($k$, $H_{\text{gap}}$, and $H_{C2}$).

The present paper is aimed to follow the change of the principal interactions in the $\text{Mn}_{1-y}\text{Fe}_y\text{Si}$ compounds. The single crystals with the concentrations $y = 0, 0.06, 0.08$, and 0.1 were chosen for the study. These samples were the discs with a thickness of 3 mm and a diameter of 30 mm. The single crystals were grown using the Bridgmann method at the Forschungszentrum Karlsruhe in Germany. The crystal mosaic is about 0.5 degree. The structure and the homogeneity of the crystals were controlled by the x-ray Laue diffraction at Helmholtz-Zentrum Berlin. All these compounds are found to have the same structure of the space group $P2_13$.

The polarized SANS experiments were carried out at the SANS-2 scattering facility of the FRG-1 research reactor in Geesthacht (Germany). A polarized neutron beam with an initial polarization of $P_0 = 0.93$, the neutron wavelength $\lambda = 0.58$ nm ($\Delta \lambda/\lambda = 0.1$), and a divergence of 2.5 mrad was used. The scattered neutrons were detected by a position sensitive detector with $256 \times 256$ pixels. The detector-sample distance was set such that the $q$ range was covered from $6 \times 10^{-2}$ to 1 nm$^{-1}$ with a step of 0.01 nm$^{-1}$. We used the magnetic field which was ranged from 0 to 800 mT and directed along [111] axis. The scattering intensity was measured in the temperature range $8 \leq T \leq 60$ K with an accuracy better than 0.05 K. The magnetization was measured with the Quantum Design physical properties measurement system at HZB.

The SANS map for the sample with $y = 0.06$ at $T = 8$ K (that is well below $T_C$) for the neutron polarization $P_0$ along ("up") the guide field is shown in Fig. 1. This map demonstrates a typical magnetic Bragg scattering from the helical spin structure. The peaks in a range of small-angle scattering are visible, proved the fact that the Bragg condition is fulfilled:
2d\sin(\Theta_B/2) = \lambda \text{ where } d \gg \lambda, \text{ with } \lambda \text{ as the period of the helix and } \Theta_B \text{ as the scattering angle. The helix wave vector } \mathbf{k} \text{ of the doped samples has an ill-defined direction since the Bragg peaks are smeared around either } \langle 111 \rangle \text{ or } \langle 100 \rangle \text{ axes. This ill-defined orientation of the wave vector } \mathbf{k} \text{ is caused by a disorder induced by Fe doping into the pure MnSi compound. The doping can be supposed to induce randomness in the local anisotropy and, therefore, unpinns partially the directions of spirals. The helix wavevector of the pure MnSi is clearly oriented along the } \langle 111 \rangle \text{ axes only with the magnetic mosaic of order of } 2\text{–}3 \text{ degrees} [9].

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The transformations of the magnetic system under applied magnetic field are studied and have found to be very typical for all compounds. Figure 2 shows magnetization and susceptibility curves of the sample Mn\(_{0.94}\)Fe\(_{0.06}\)Si taken at \( T = 2 \) K. The magnetization changes the slope from \( \gamma_1 \) to \( \gamma_2 \) in the range of small field at \( H_{C1} \). Further on, the magnetization demonstrates the linear dependence in the range from \( H_{C1} \) to \( H_{C2} \). It saturates above the critical field \( H_{C2} \) indicating the field-induced phase transition from the conical to the ferromagnetic state. The susceptibility shows clearly the features at these two critical fields \( H_{C1} \) and \( H_{C2} \).

The small angle diffraction measurements show that in absence of the magnetic field the magnetic structure consists of spiral domains with ill-defined orientation of the spiral wavevector \( \mathbf{k} \). An external magnetic field provides the process of reorientation of spiral domain along the field axis. This process starts from the threshold field \( H_{C1} \) defined as a field which dominates over the local anisotropy and forms in the sample the single domain state with conical spirals.

Figure 1. SANS map of Mn\(_{0.94}\)Fe\(_{0.06}\)Si at \( T = 8 \) K.

Figure 2. Magnetic field dependence of the magnetic susceptibility \( \mu (\circ) \) and the magnetization (●) of Mn\(_{0.94}\)Fe\(_{0.06}\)Si at \( T = 2 \) K. Inset shows the crossover at \( H = H_{C1} \) in more details.
oriented along the field. In the scattering picture, it is seen as an accumulation of the different Bragg spots to the single one with \( k \) collinear to the magnetic field axis. Further increase of the magnetic field destroys the spiral order and forms the ferromagnetic state at \( H = H_{C2} \), where the Bragg spot vanishes. The system remains a stable conical structure in the range \( H_{C1} < H < H_{C2} \). The critical fields \( H_{C1} \) and \( H_{C2} \) are plotted as a function of temperature in the \( H - T \) phase diagram (Fig.3) for the representative compound with \( y = 0.06 \).

![Figure 3. \( H - T \) phase diagram of Mn\(_{0.94}\)Fe\(_{0.06}\)Si.](image)

The important feature of the \( H - T \) phase diagram (Fig. 3) is a \( k \)-flop of the helix wave vector that occurs in a certain field range close to \( T_C \). The \( k \)-flop is seen in the diffraction experiment as a 90° jump of the wavevector from \( k \parallel H \) to \( k \perp H \). Going deeper into the \( k \)-flop phase, the intensity of the Bragg reflection at \( q = k \parallel H \) decreases, while a new Bragg spot appears at \( k \perp H \). The field \( H_{\text{fl}} \) in Fig. 3 (triangles) shows the boundary of the new \( k \)-flop phase. The field value associated with the minimum of the intensity for \( k \parallel H \) is denoted as \( H_{\text{gap}} \).

The \( H - T \) phase diagram shown in Fig. 3 is typical for the cubic helimagnets including the ternary compounds under study Mn\(_{1-y}\)Fe\(_y\)Si and the compounds Fe\(_{1-x}\)Co\(_x\)Si studied in [10, 11]. The similarity of the \( H - T \) phase diagrams of different compounds suggests that the magnetic system of these compounds are governed by the same set of the interactions which interplay determines the values of the critical fields \( H_{C2} \) and \( H_{\text{gap}} \) as well as the value of the helix wavevector \( k \) (see Eqs. (1 – 3)). The concentration dependences of these three parameters are shown in Fig. 4. Here \( H_{C2} \) is determined as a value taken at the lowest measured temperature. As seen from Fig. 4 values of \( H_{C2} \) and \( H_{\text{gap}} \) demonstrate rather small alteration. The value of the helix wavevector \( |k| = k = 2\pi/d \) changes significantly showing a linear increase from 0.35 to 0.63 nm\(^{-1}\) with doping \( y \), i.e. the helix length shortens from 185 to 100 Å.

Taking into account the theory [12] and using Eqs. (1 - 3) we have calculated the changes of the major driving interactions of the magnetic system with doping \( y \): the spin wave stiffness \( A \), the Dzyaloshinskii constant \( SD \), and the spin wave gap \( \Delta \). This theory was successfully applied to the pure MnSi [9] and to the ternary compounds Fe\(_{1-x}\)Co\(_x\)Si [10].

The calculated energies of the principal interactions \( A/a^2 \), \( SD/a \), and \( \Delta \) are shown in Fig. 5 as a function of \( y \) (here \( a \) is the lattice parameter). The exchange energy \( A/a^2 \) decreases linearly from 2.4 to 1 meV. The extrapolation (red dotted line in Fig. 5) gives the critical concentration \( y \approx 0.15 \) where the exchange interaction becomes zero. The energy of DM interaction behaves similarly and has a moderated decrease from 0.38 to 0.25 meV. Opposite to these two, the spin wave gap \( \Delta \) linearly increases from 12 to 32 µeV (see the inset in Fig. 5).
Figure 4. Concentration dependences of the following principal parameters of Mn$_{1-y}$Fe$_y$Si: (■) the second critical field $H_{C2}$; (●) the k-flop field $H_{gap}$; (▲) the value of the helix wave vector $|\mathbf{k}|$.

Figure 5. Concentration dependences of the following principal interactions of Mn$_{1-y}$Fe$_y$Si: (●) the spin wave stiffness energy $A/a^2$; (■) the Dzyaloshinskii energy. Inset shows the concentration dependence of the spin wave gap $\Delta$.

Figure 6. Critical temperature $T_C$ as a function of the Fe concentration $y$. Solid line is the linear extrapolation of the $T_C$ that approaches $T = 0$ at $y \approx 0.13$.

Remarkable though that this set of the principal interactions is able to explain the concentration dependence of the critical temperature $T_C$ plotted in Fig. 6. The critical temperature $T_C$ has been obtained from the neutron scattering measurements as the temperature, where the magnetic Bragg reflection transforms from Gaussian to Lorenzian. The critical temperature of this transformation was determined $T_C = 28.80(5)$, $16.50(5)$, $10.55(5)$, and $6.80(5)$ K for the samples with $y = 0.0, 0.06, 0.08, \text{and} 0.10$, respectively. $T_C$ is well seen to decrease linearly with the Fe concentration $y$ in agreement with the early studies (see Ref. [13] and references therein). Comparing concentration dependences of $T_C$ (Fig. 6) and $A$ (Fig. 5) one can see that the critical temperature resembles the behavior of the spin wave stiffness. This fact leads to the conclusion that the isotropic ferromagnetic exchange interaction governes the transition temperature in Mn$_{1-y}$Fe$_y$Si.

In our estimations using Eqs. (1 – 3) the value of the DM interaction is coupled to the average spin per unit cell $SD/a$. In fact the average spin changes significantly with the concentration.
The tendency to quantum phase transition at $T_\text{C}$ approaching 0 suggests that the Fe doping can lead the system $\text{Mn}_{1-y}\text{Fe}_y\text{Si}$ to the quantum phase transition at $y \approx 0.13$. Indeed, the increase of the non-thermal parameter, the concentration $y$, results in $T_\text{C}$ approaching 0, which is, by definition, the Quantum Critical Point (QCP). This concept can help to model the behavior of these systems close to the QPT using the Fe concentration as a non-thermal parameter. We believe that following the evolution of the system on the way to the QPT, using the changes of the principal interactions with doping $y$ estimated in this work, and combining all these data with a those of the critical behavior with $y$ up to 0.15 can shed a new light on the quantum phase transition in MnSi.

In this paper we have considered the concentration evolution of the principal interactions in the compound $\text{Mn}_{1-y}\text{Fe}_y\text{Si}$. These interactions have been calculated taking into account the theory [12] and combined data of the neutron diffraction and magnetization measurements. Firstly, we have established the Dzyaloshinskii constant $D$ is independent on the concentration of the dope in both cases $\text{Mn}_{1-y}\text{Fe}_y\text{Si}$ and $\text{Fe}_{1-x}\text{Co}_x\text{Si}$. Secondly, we have obtained that the transition temperature is governed by the energy of isotropic ferromagnetic exchange interaction. The tendency $T_\text{C} \rightarrow 0$ suggests that the Fe doping can lead the system $\text{Mn}_{1-y}\text{Fe}_y\text{Si}$ to the quantum phase transition at $y \approx 0.13$. Indeed, the increase of the non-thermal parameter, the concentration $y$, results in $T_\text{C}$ approaching 0, which is, by definition, the Quantum Critical Point (QCP). This concept can help to model the behavior of these systems close to the QPT using the Fe concentration as a non-thermal parameter. We believe that following the evolution of the system on the way to the QPT, using the changes of the principal interactions with doping $y$ estimated in this work, and combining all these data with a those of the critical behavior with $y$ up to 0.15 can shed a new light on the quantum phase transition in MnSi.

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