

Skyrmion Lattice Domains in $\text{Fe}_{1-x}\text{Co}_x\text{Si}$

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Abstract. The strongly doped semiconductor $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ displays a dome of helimagnetic order for $0.05 \leq x \leq 0.7$. We report small angle neutron scattering of the magnetic structure in the skyrmion lattice phase of $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ for $x = 0.2$ and magnetic field parallel to a crystallographic $\langle 100 \rangle$ direction. We observe twelve equally spaced maxima of scattering intensity on a ring, with an underlying six-fold symmetry of two sets of six spots. The intensity distribution suggests the formation of two degenerate skyrmion lattice domain populations with respect to the four-fold symmetry of the $\langle 100 \rangle$ directions in the scattering plane.

1. Introduction

Recently the A-phase of the B20 compound MnSi was identified as a skyrmion lattice, i.e., a new form of magnetic order composed of topologically stable knots of the spin structure as its fundamental building blocks [1]. The skyrmions may be viewed as a type of magnetic vortices that are aligned parallel to the applied magnetic field, assuming a hexagonal densest packing. In fact, the skyrmion lattice shares considerable similarities with the vortex lattice in the intermediate mixed state of type two superconductors (for a discussion of the analogy see Ref. [2]). A key aspect is thereby, that weak chiral spin-orbit interactions (so called Dzyaloshinsky-Moriya interactions) are much weaker than ferromagnetic exchange, and much stronger than crystal field interactions. This implies a clear separation of energy scales which cause the formation of long wavelength chiral modulations that decouple very efficiently from the crystal lattice.

Just like for superconducting vortex lattices the ideal microscopic probe to explore the properties of skyrmion lattices in chiral magnets is small angle neutron scattering. However, because the skyrmion lattices observed so far are similar to an ideal triple- k spin structure, consisting of the superposition of three helices enclosing angles of 120° in a plane that is perfectly perpendicular to the applied magnetic field, higher harmonics have been below the detection limit of the neutron scattering experiments. In order to prove the existence of the skyrmion lattice beyond doubt requires experimental evidence for a fixed phase relationship of the three fundamental helical modulations over a long spatial range. In MnSi this has recently been achieved by means of the Hall effect, which displays an additional topological contribution consistent with the proposed skyrmion lattice besides the usual normal and anomalous Hall contributions [3].

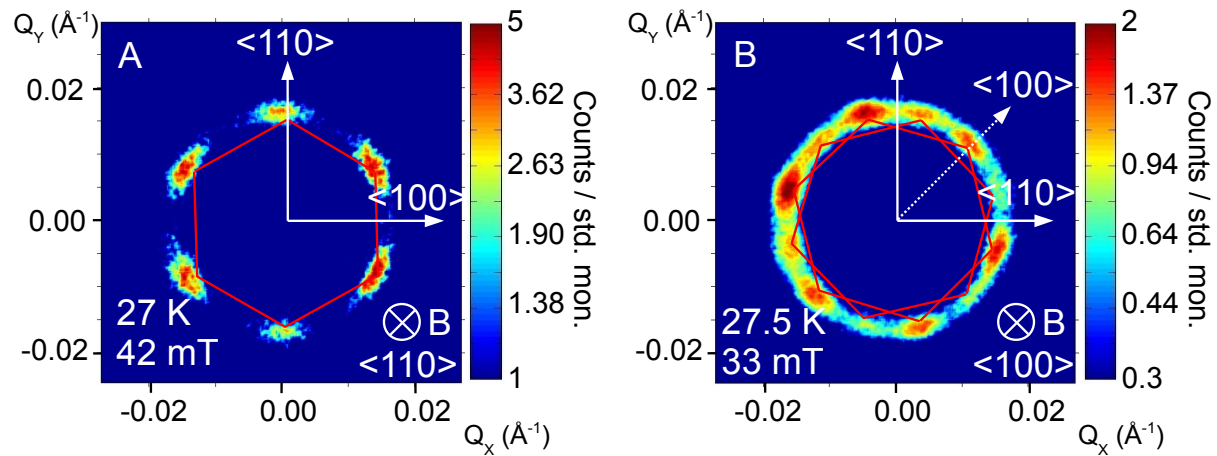


Figure 1. Typical scattering intensity in the A-phase of $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ ($x = 0.2$). Data were obtained after cooling in the magnetic field. Panel (a) displays the clear six-fold diffraction pattern characteristic of the formation of a skyrmion lattice. Panel (b) displays the diffraction pattern for magnetic field parallel to the $\langle 100 \rangle$ axis, when twelve equally spaced maxima on a circle are observed. This maxima are characteristic of two domain populations in a crystal field environment with two degenerate pinning potentials.

First evidence that the skyrmion lattice in MnSi is not just a material specific property but a more general phenomenon was observed in studies of the B20 compound $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ for $x = 0.2$ and $x = 0.25$ [4] and MnSi under Fe and Co doping [5–7]. In contrast to MnSi, which forms in a pure metallic state, the helimagnetic properties of $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ emerge in a strongly doped semiconductor, located between the nonmagnetic semiconductor FeSi and the diamagnetic metal CoSi [8, 9]. In addition the relationship between the lack of inversion symmetry and the chirality of the spin-orbit interactions is opposite to MnSi, i.e., the Dzyaloshinsky-Moriya vector has opposite sign.

The key signature of the skyrmion lattice in MnSi and $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ that was first noticed in neutron scattering for applied magnetic field aligned parallel to the incident neutron beam is an intensity pattern with a sixfold symmetry. In MnSi the scattering pattern is not hysteretic while it is highly hysteretic in $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ as described in [4]. In general the six-fold scattering pattern of the skyrmion lattice is pinned along a $\langle 110 \rangle$ direction or the closest direction to it. In contrast, for magnetic field parallel to a $\langle 100 \rangle$ direction the pinning terms for the skyrmion lattice favor an alignment for either of the two $\langle 100 \rangle$ directions. This raises the question for the possible existence of different skyrmion lattice domains.

2. Experimental Techniques

Single crystals of $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ were grown by optical float-zoning under a purified Ar atmosphere with an ultra-high-vacuum (UHV) compatible image furnace. The polycrystalline feed-rods for the optical float-zoning were prepared in a UHV-compatible rod casting furnace using a bespoke Hukin crucible and radio-frequency heating. The large single crystals grown, were characterized by means of Laue x-ray diffraction and neutron single crystal diffraction. The neutron diffraction was carried out on the diffractometer RESI at FRM II. The characterization of the crystal structure established excellent properties with no evidence for short range disorder or the formation of a superlattice. The sample were characterized further by means of comprehensive measurements of the magnetization and the electrical resistivity as a function of temperature

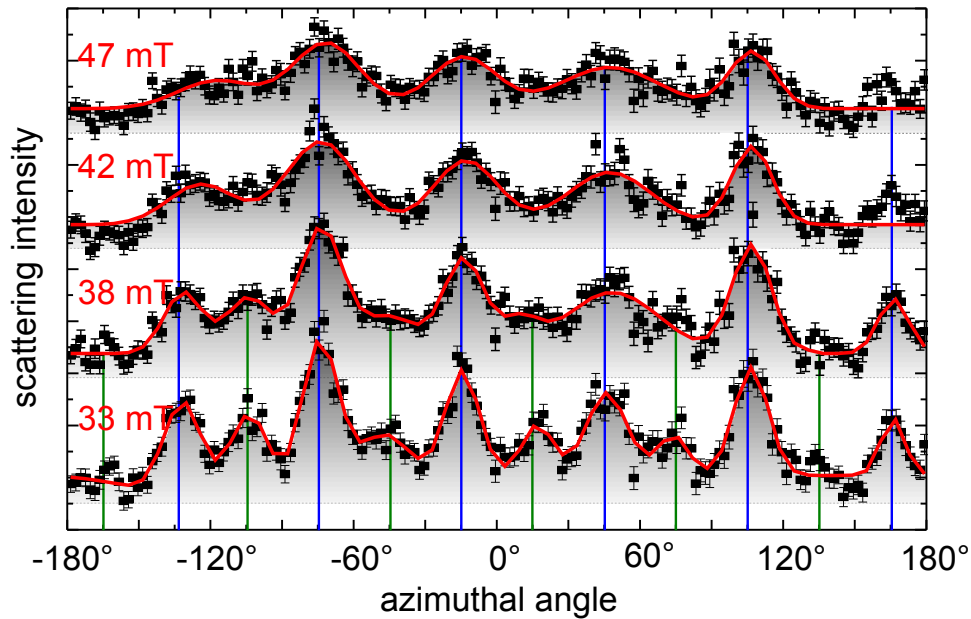


Figure 2. Azimuthal intensity variation in the A-phase of $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ for magnetic field parallel to the crystallographic $\langle 100 \rangle$ direction. Zero degrees corresponds to the top of the vertical direction. For clarity curves have been shifted vertically. Two sets of six equally spaced maxima are observed as marked by the green and the blue vertical lines. With increasing magnetic field the maxima at the green lines vanish, while the maxima at the blue lines increase, broaden and eventually vanish for the highest field measured. This behavior is characteristic for domain populations of skyrmion lattices.

and magnetic. In combination all of the data establish an excellent quality of our samples of $\text{Fe}_{1-x}\text{Co}_x\text{Si}$.

The small angle neutron scattering data of $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ reported in the following were obtained at the cold diffractometer MIRA at FRM II, using neutrons with a wavelength $\lambda = 9.7 \text{ \AA} \pm 5\%$. Backgrounds were determined at high temperatures and subtracted accordingly. A Cd marker on the sample support confirmed that the sample was always oriented correctly. The sample was cooled with a pulse-tube cooler. Magnetic fields up to 0.3 T were applied with a bespoke set of Helmholtz-coils. Data were recorded for a fixed sample orientation following rocking scans with respect to a vertical axis of typically $\pm 15^\circ$.

3. Results

Typical intensity patterns recorded after field-cooling into the A-phase of $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ are shown in Fig. 1. The characteristic six-fold symmetry is readily evident for magnetic field applied parallel to $\langle 110 \rangle$ as shown in Fig. 1 (A) and field parallel $\langle 111 \rangle$ (not shown). To guide the eye the peak maxima are connected by a thin red line. For both $\langle 110 \rangle$ and $\langle 111 \rangle$ the scattering pattern is aligned with a $\langle 110 \rangle$ direction in the scattering plane. The scattering pattern differs considerably for magnetic field applied parallel to the $\langle 100 \rangle$ direction as shown in Fig. 1 (B). Here a total of twelve spots are observed, comprised of two sets of six spots. Each set of six spots is aligned with one of the $\langle 100 \rangle$ directions in the scattering plane.

A more detailed analysis of the intensity distribution as a function of the azimuthal angle is shown Fig. 2 for various magnetic fields. Data were recorded after field cooling the sample. For the lowest magnetic field of 31 mT six pronounced maxima are observed every 60 degrees. The

location of these maxima is marked by thin blue lines. Between the large maxima we observe small maxima also spaced every 60 degrees, marked by a thin green line.

With increasing magnetic field the large maxima broaden and increase slightly in size until the small maxima have vanished at a field of 44 mT. At the highest field shown the large maxima are broadened even further while having decreased somewhat. It is thereby interesting to note that the location of the maxima is unchanged at all fields, i.e., the scattering pattern remains aligned with a crystallographic $\langle 100 \rangle$ direction.

4. Discussion

The key microscopic feature of the skyrmion lattice observed in small angle neutron scattering is an intensity pattern with six-fold symmetry. The theoretical framework presented in Ref. [1], readily explains the six-fold pattern including its orientation as well as the magnetic phase diagram as a skyrmion lattice. There is hence no doubt, that the A-phase in $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ represents a skyrmion lattice.

For field parallel $\langle 111 \rangle$ and $\langle 110 \rangle$ the scattering pattern in the A-phase aligns along a $\langle 110 \rangle$ direction consistent with an anisotropy term $\sum_{\vec{k}}(k_x^6 + k_y^6 + k_z^6)\vec{m}_{\vec{k}}\vec{m}_{-\vec{k}}$ with positive coefficient. In contrast, for field parallel $\langle 100 \rangle$ this term does not cause a dominant alignment, whereas terms such as $\sum_{\vec{k}}(k_x^4 k_y^2 + k_y^4 k_z^2 + k_z^4 k_x^2)\vec{m}_{\vec{k}}\vec{m}_{-\vec{k}}$ align the skyrmion lattice along one of the $\langle 100 \rangle$ directions depending on the sign of the prefactor. In our study we observe precisely such an alignment. Surprisingly, however, our small angle neutron scattering results on the A-phase of $\text{Fe}_{1-x}\text{Co}_x\text{Si}$ establish *both* skyrmion lattice orientations. This implies the unexpected formation of skyrmion lattice domain populations for field parallel $\langle 100 \rangle$. The presence of two domain populations may originate in at least two mechanisms. First, disorder-induced random prefactors of the relevant anisotropy term or, second, that one of the domain populations is metastable.

Acknowledgments

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