Helimagnons in the Skyrmion Lattice of MnSi

M. Janoschek1,2, F. Jonietz2, P. Link3, C. Pfleiderer2, P. Böni2
1 II. Physikalisches Institut der Universität zu Köln, D-50937 Köln, Germany
2 Physik Department E21, Technische Universität München, D-85748 Garching, Germany
3 Forschungsneutronenquelle Heinz Maier-Leibniz (FRM II), Technische Universität München, D-85748 Garching, Germany
E-mail: marc.janoschek@frm2.tum.de

Abstract. In MnSi the application of a small magnetic field destabilizes the helimagnetic order in a narrow temperature interval just below the helimagnetic ordering temperature and stabilizes the formation of a hexagonal lattice of skyrmions, i.e., a lattice composed of a type of magnetic vortex lines. We have studied the skyrmion lattice in MnSi using a cold triple-axis spectrometer. Our data suggests that the skyrmion lattice represents a three-dimensional spin structure. The collective spin excitations of the skyrmion lattice are strongly reminiscent of the rich spectrum of helimagnon bands, recently shown to be a universal property of the helimagnetic state of MnSi in zero magnetic field.

1. Introduction

MnSi crystallizes in the non-centrosymmetric cubic space group P213 (a = 4.558 Å). The magnetic properties of MnSi result from a clear separation of energy scales in a metallic host. Below \( T_c = 29.5 \text{ K} \) and in zero magnetic field a long-wavelength spin spiral with the spins perpendicular to the propagation direction stabilizes. The competition of ferromagnetic exchange interactions, as the strongest scale, and Dzyaloshinskii-Moriya (DM) interactions, on an intermediate scale, result in a period of the helix of \( \lambda_h \approx 180 \text{ Å} \) [1, 2]. Here, the DM interactions are a manifestation of weak spin-orbit coupling in crystal structures without inversion center. The propagation direction of the helix is locked to the cubic space diagonal by the weakest energy scale, higher order spin-orbit interactions, also referred to as crystal field interactions.

As an important consequence, the hierarchical magnetic energy scales in MnSi result in a rich spectrum of helimagnon bands [3], recently observed in great detail in a comprehensive inelastic neutron scattering study [4]. Using a model based on only three parameters, namely the measured pitch of the helix, the measured spin wave stiffness in the ferromagnetic phase, and an overall amplitude of the signal as the only free parameter, one obtains a quantitatively precise and complete account of these helimagnon bands. In particular, the abundance of helimagnon bands turns out to be a new universal property of spin excitations that is driven by multiple, strong Umklapp scattering in small magnetic Brillouin zones.

Recently it was found that the application of a small magnetic field \( B \approx 150 \text{ mT} \) destabilizes the helimagnetic order and leads to a hexagonal lattice of skyrmions, a type of magnetic vortices [5]. The spin crystal is observed in a small phase pocket approximately 2 K wide just below \( T_c \), long known as the A-phase [6, 7]. The magnetic structure in the A phase may be derived from
a helical triple-\( k \)-structure where the propagation vectors of the three helices strictly reside in a plane perpendicular to the applied magnetic field assuming angles of 120° with respect to each other keeping a fixed phase relationship. Closer examination of the spin structure shows, that it is actually composed of topologically stable knots, the so-called skyrmions, as confirmed by the observation of a topological Hall effect [8].

As the skyrmions are akin to vortex lines in type II superconductors an important question concerns, if the spin structure in the skyrmion lattice represents a two- or a three-dimensional form of order. Moreover, as the skyrmion lattice represents a novel form of magnetic order composed of topologically stable knots an additional key question concerns, if the low-lying collective spin excitations in the A phase differ radically from those observed in the helical state. Understanding the spin excitations of the skyrmion lattice is also important in the context of the observation of partial magnetic order in the non-Fermi liquid regime of MnSi at high pressures [9, 10, 11]. We have studied these questions using a cold triple-axis neutron spectrometer. Our study establishes that the skyrmion lattice represents a three-dimensional spin structure and that the collective spin excitations are strongly reminiscent of the helimagnons observed in the helical state.

2. Experimental setup

Our experiments were carried out on the cold triple-axis spectrometer PANDA at the Forschungszentrum Jülich. A large single crystal of \( \sim 8 \) cm\(^3\) studied before (s. Ref. [12, 4]) was inserted in a standard FRM-II top-loading closed-cycle cryostat. The cryostat was hosted inside the room temperature bore of the cryogen free 7.5 T superconducting magnet. The sample was oriented with the [1, 1, 0] and [0, 0, 1] crystallographic directions in the scattering plane. The magnetic field was oriented perpendicular to the scattering plane, i.e., parallel to a crystallographic [1, 1, 0] direction. In order to avoid second order contamination of the neutron beam and to lower the background a beryllium filter was inserted between the monochromator and the sample. Additionally 60° Soller collimators were installed in the incident beam and in front of the detector, whereas 40° collimators were used in front of the sample and analyser.
3. Results
In a first series of measurements we performed elastic $Q$-scans as a function of temperature and magnetic field in order to determine the phase boundaries of the A-phase by tracking the magnetic intensities on the respective magnetic satellite reflections. The scans were performed with a final wave vector $k_f = 1.55 \, \text{Å}^{-1}$. The maximum magnetic intensity in the A-phase was found at $T = 27.7 \, \text{K}$ and $B = 170 \, \text{mT}$. All consecutive scans in the A-phase were performed at these values. Fig. 1 shows two maps around the $(1,1,0)$ nuclear Bragg reflection obtained by elastic $Q$-scans parallel to the $[1,1,0]$ direction in the helical phase ($B = 0 \, \text{mT}$) and the A-phase ($B = 170 \, \text{mT}$), respectively.

In addition we carried out a rocking scan on the magnetic satellite reflection $(1,0.02,0.02,0)$ in the A-phase. For the scan we rocked the sample around the crystallographic $[0,0,1]$ axis that is parallel to the nominal scattering plane and perpendicular to the scattering vector $Q$ via the motorized sample goniometer of PANDA. This way the magnetic correlation length in the direction perpendicular to the scattering plane was probed. In order to improve the vertical $Q$-resolution of the spectrometer an additional vertical $15'$ Soller collimator was inserted in front of the analyzer and $k_f = 1.15 \, \text{Å}^{-1}$ was employed. During the rocking scan the orientation of the sample with respect to the magnetic field was kept fixed. The corresponding scan is shown in Fig. 2(a). For comparison corresponding vertical rocking scans were performed for the nuclear reflection $(1,1,0)$ and the magnetic satellite reflection $(1.016,1.016,0.016)$ in the helical phase. They are also given in Fig. 2(a). The correlation length was estimated by fitting a Gaussian profile to each of the peaks. The FWHM of all three peaks was found to be equal within the error and amounts to $1.3^\circ$. This correspond to $\Delta Q \approx 0.046 \, \text{Å}^{-1}$ and to a resolution limited magnetic correlation length $\xi > 139 \, \text{Å}$.

We also performed inelastic scans in the A-phase at a few selected positions along the $[1,1,0]$ directions, i.e., parallel to the propagation direction of one of the three helices in the A-phase (cf. small right panel of Fig. 2(b)). The constant-$Q$ scans are shown in the main panel of Fig. 2(b). The modes observed are broad and show shallow maxima similar to the helimagnon modes we reported in Ref. [4]. At first sight these scans may suggest the presence of two distinct maxima of tentative dispersive modes. However, in our previous work for the helical phase we have already demonstrated that this approach leads to misleading results. In fact, our data are strongly reminiscent of the helimagnon bands observed in the helical phase. Therefore, the solid lines in Fig. 2(b) are only guides to the eye.

4. Discussion
The elastic scans around the nuclear Bragg reflection $(1,1,0)$ establish that the magnetic satellite reflections of the skyrmion lattice are also present around nuclear reflections. Previous experiments [5] on the skyrmion lattice have been only performed in a small angle neutron scattering geometry, thus the reflections were only observed around the direct beam. This highlights an important difference between the skyrmion lattice and the Abrikosov lattice in type II superconductors. While the latter is NOT connected with the chemical crystal structure, this is not the case in the former case. In other words, the spin structure in the skyrmion lattice is intimately connected with the crystal lattice.

The rocking scans reported here moreover show, that the magnetic correlation length of the skyrmion lattice parallel to the magnetic field is comparable to the that observed in the helically ordered state. Keeping in mind that the resolution is not particularly high, this strongly suggests that the skyrmion lattice represents a true three-dimensional magnetic order, rather than a mere two dimensional lattice of particle-like objects.

Finally, the collective spin excitations are strongly reminiscent of the helimagnon modes observed previously in the helimagnetic state[4]. This shows that the helimagnons are preserved from the helical into the A-phase. Our previous work also shows that the helimagnons cannot
Figure 2: (a) Rocking curves of the nuclear \((1,1,0)\) Bragg reflection (black circles), the magnetic satellite \((1.016,0.016,0.016)\) of the helical phase (red squares) and the magnetic satellite \((1.02,1.02,0)\) of the skyrmion lattice (blue triangles). The rocking scans probe the magnetic correlation length in the direction perpendicular to the scattering plane (s. text for details). For clarity the intensity of the nuclear reflection is divided by a factor 50. (b) Four constant-Q-scans performed within the A-Phase at \(T = 27.7\ K\) and \(B = 170\ mT\). The scans are shifted by 100 counts for clarity. The solid lines are guides to the eye. The sharp peak (open data points) in the lowest graph at \(\bar{\omega} \approx 0.31\ meV\) is a spurious signal that arises from additional incoherent scattering of neutrons from the analyzer crystals of the triple axis spectrometer (cf Ref. [4]). The small panel on the right hand side shows the positions in the reciprocal space where the scans have been performed. The yellow circles represent the six magnetic reflections of the A-phase.

be fitted by simple approaches as this leads to misleading results. In turn our study raises as a new challenge to theory, whether the spin excitations under magnetic field, notably in the skyrmion lattice, are also as universal as the helimagnon bands observed in zero field.

Acknowledgments
We wish to thank S. Mühlbauer, B. Binz and A. Rosch for helpful discussions and D. Etzdorf for technical support. The neutron scattering experiments were carried out at the Forschungs-Neutronenquelle Heinz Maier-Leibnitz (FRM-II) of Technische Universität München at Garching in Germany.

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