Article

The Helical Magnet MnSi: Skyrmions and Magnons

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Abstract: MnSi has played a major role since the late 1970s in developing the theory of helical magnets in non-centro symmetric materials showing Dzyaloshinsky-Moriya interaction (DMI). With a long helimagnetic pitch of 175 Å as compared to the lattice d-spacing of 4.55Å, it was ideal for performing neutron studies especially as large single crystals could be grown. In these studies under the application of a field of 180 mT perpendicular to Q, a so-called A-phase in the B-T phase diagram was found and interpreted as a rotation of the alignment of the magnetic helix away from the pinning axis. After the surprising discovery of the skyrmion lattice in the A-phase in 2009 much interest arose as it could be shown that the magnetic skyrmion lattice is topologically protected. Due to the rigidity of the skyrmionic lattice it is only loosely bound to the crystal lattice and therefore only relatively small current densities can already induce a motion of this lattice. Another very interesting aspect are the excitations in the spin system of MnSi. As the helimagnetic state is characterized by a long pitch of about 175 Å, the associated characteristic excitations form a band structure due to Umklapp scattering and can only be observed at very small q with energies below 1 meV. We have investigated the the magnons in MnSi in the whole (B,T)-phase diagram starting in the single-k helimagnetic state by applying a small magnetic field B = 100 mT. This way, the complexity of the magnon spectrum is significantly reduced allowing a detailed comparison of the data with theory resulting in a full theoretical understanding of the spin system of MnSi in all its different magnetic phases.

Keywords: Helimagnets, magnetic skyrmions, non-recpriocal dispersion relation

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1. What makes MnSi so interesting?

Helical magnets are currently of high interest because they serve as model systems for complex magnetic ordering yielding interesting properties such as multiferroicity and the appearance of stable magnetic vortices (i.e. skyrmions) to name a few. The interplay of the various energy scales often leads to magnetic ordering exhibiting a periodicity that is incommensurate with the lattice and to magnetic Brillouin zones that are much smaller the chemical ones.

A prominent example showing incommensurate helical ordering is MnSi being a non-centro symmetric itinerant magnet with a $P2_13$ structure and a lattice spacing of a = 4.55 Å. The (B-T)-phase diagram is shown on the left of Fig. 1. It exhibits a large variety of different phases such as single-handed helical and conical ordering, a field-polarized and paramagnetic state, as well as a non-trivial topological spin state (A-phase), i.e. a skyrmion phase.

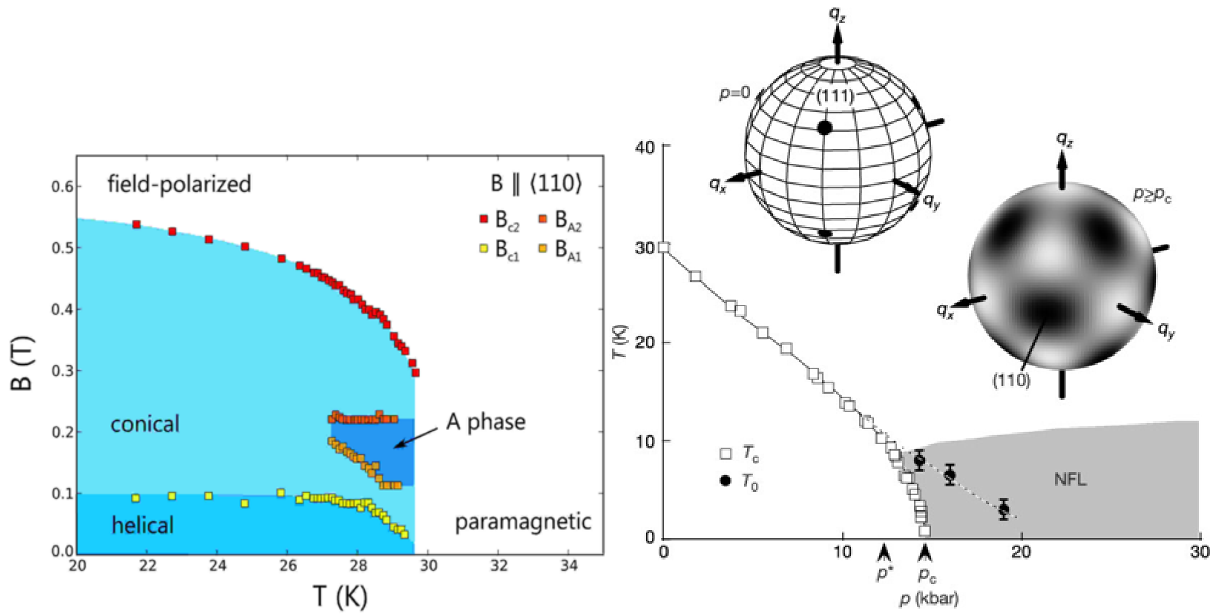

Figure 1. (Left) The (B-T)-phase diagram of MnSi. The A-phase is a skyrmion phase (reproduced from [1], (c) by the American Association for the Advancement of Science (AAAS). **(Right)** The (T-P)-phase diagram of MnSi. The grey shaded area designates the non-Fermi-liquid phase (NFL). The spheres show directions of strong elastic neutron intensity in reciprocal space (reproduced from [2], (c) by Springer Nature.)

In this article we show how the concept of a skyrmionic crystal developed from the neutron scattering experiments and the measurement of the anomalous Hall effect. Furthermore we will show the behavior of magnetic excitations (magnons) in all parts of the phase diagram of MnSi. Together with a mean-field theory of the dynamics in MnSi this leads to a very detailed understanding of the experimentally observed spectra.

2. How everything began

Somewhat surprisingly, despite the discovery of the helimagnetic state in the 1970s [3,4], initial research focused on the investigation of the magnetic excitations in the field-polarized phase of MnSi (see Fig. 1) because MnSi represents beside Ni_3Al one of the best realizations of a weak itinerant ferromagnet due to the large difference between the ordered and the paramagnetic moments. In his seminal works, Ishikawa et al. succeeded to map out the spectrum of the spin waves, the Stoner excitations, as well as the paramagnetic scattering up to room temperature [4].

It was Shirane et al. who investigated for the first time the effect of the single-handed helimagnetic phase in MnSi on the cross section of polarized neutrons [5] followed by work by Roessli et al. who demonstrated the appearance of chiral correlations due to the single-handedness of the helical ordering in the paramagnetic phase [6].

In 2004, Pfleiderer et al. discovered the appearance of a non-Fermi-liquid phase under the application of a pressure above 14.6 kbar [2]. Most interestingly, the intensity of the magnetic satellite peaks due to the magnetic ordering vanished and scattering in the {110} directions in small angle neutron scattering appeared. The origin of these peaks and the excitations stabilizing or destabilizing the NFL phase are not known at present. At this time, the phase diagram of MnSi the (T-p)-regime was established too (see Fig. 1).

Theoretical explanation were based on the early theory of Bak and Jensen [7], who proposed an explanation of the phase diagram of MnSi based on the Dzyaloshinsky-Moriya interaction (DMI) originating from the non-centro symmetry. We now understand the appearance of most of the

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magnetic phases in MnSi to be a generic property of the hierarchy of the various energy scales appearing in these B20 compounds, as for the helical ground state: The ferromagnetic exchange interaction corresponds to the strongest scale and leads to a preference for parallel spin alignment. The much weaker Dzyaloshinsky-Moriya interaction favors a perpendicular alignment of the spins. The competition between the two leads to the formation of a spin spiral with a large pitch of $\lambda_h = 175$ Å. The DMI is the leading term (2nd order) in the spin-orbit coupling (SOC) which is a relativistic correction. It is a consequence of the crystal structure ($P2_13$), which lacks inversion symmetry. Finally, very weak crystal field effects (4th order SOC) representing the lowest energy scale pin the wave vectors k_h of the helix to point along the space diagonals of the cubic unit cell (a = 4.558 Å) of MnSi with $k_h = 2\pi/\lambda_h = 0.036$Å^{-1}.

There is an interesting feature in the (B-T) phase diagram, the so-called A-phase. The small angle scattering experiments, at that time performed only with a field perpendicular to Q, showed a disappearance of the magnetic intensity when entering this small pocket in the phase diagram. This was interpreted as a re-orientation of the magnetic helix out of the scattering plane.

3. From the neutron small angle scattering results to the skyrmion interpretation

In 2009, extensive small angle neutron scattering experiments had been performed in all phases of MnSi [1,8]. Most of these experiments were performed with field perpendicular to Q, but some of these measurements were also performed along Q. Apart from the A-Phase the results confirmed the picture described above with the hierarchy of the three interactions. But in the A-Phase, when applying B parallel to Q, a six-fold geometry showed up independent of orientation of the underlying crystal. This results has been explained as the formation of a skyrmion lattice. Theoretically it can be described by a 3-fold q-structure with Q=0, which has a local minimum of the free energy inside the A-phase. Due to thermal fluctuations this structure is being stabilized in a global minimum in the A-phase. This leads to the picture of a hexagonal vortex like skyrmion lattice or crystal in MnSi.

The topological nature of the skyrmions and their interaction with electrons are described in terms of the (fictitious) emergent electromagnetic fields [9–12] E and B. Taken together B and E account for the Berry phase that the spin of a conduction electron accumulates when following the magnetic texture adiabatically. As the integral of $B/\hbar$ over a surface describes the solid angle ds, the emergent magnetic flux of each skyrmion is exactly given by one (negative) flux quantum [9]

$$\int Bds = -\phi_0. \tag{1}$$

Interestingly the skyrmions are therefore characterized by a non-zero winding number indicating a topical different structure from classical magnetic systems. Experimental consequences have been detected directly in terms of an additional (topological) contribution to the Hall signal and an emergent electric field, providing evidence of the motion of the skyrmions [9,13,14]. The measurements of this motion show that the skyrmions are only loosely bounded to the lattice of the underlying crystal and that with current densities 3 orders of magnitude smaller as in usual magnetic structures the lattice can start moving [14]. This is due to the rigidity of the involved lattices and the large periodicity, which leads to an efficient decoupling of the magnetic and atomic lattices.

As the topologically quantized winding number or, equivalently, the quantized magnetic flux has to change when two skyrmions merge a decay of a skyrmion is only possible via a monopole interaction and such this structures are strongly stabilized. Because of the topological nature of the winding number, this is in fact only possible by a singular field configuration for which the local magnetization vanishes at a point in space. Hence, when two ingoing skyrmions merge, there must be a singular field configuration, a hedgehog defect with winding number +1, which creates one quantum of emergent magnetic flux. The point of coalescence therefore carries a quantized emergent magnetic charge; i.e., it is an emergent magnetic monopole. Similarly, when an ingoing single skyrmion line splits into two, an anti-monopole with winding number -1 is located at the point of separation.

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The stability and the high mobility of the skyrmions made these structures ideal candidates for quantum computing storage devices and therefore since the discovery of the skyrmion crystal in MnSi several hundreds of publications have been dealt with such topological protected multi-q structures in metals, semimetals and insulators in bulk materials. They were found in thin films and isolators and even more complex systems are discussed.

4. Magnons in MnSi

So far we only dealt with the structure within the different magnetic phases of MnSi. There are also magnetic excitations of the spirals and the skyrmion lattice, which shed light on the dynamics of these spin systems. Exciting these modes requires a very small energy transfer even at relatively large q. It turns out that most of the interesting physics takes place below 1 meV excitation energy, thus missed by the early experiments and now reinvestigated with modern instruments with an adequate momentum and energy resolution.

As for the structure in each of the magnetic phases typical excitations can be found and we will explain their dynamics in the following subsections going with increasing magnetic field from the helical, over the conical into the ferromagnetic phase. We also measured the excitations of the skyrmion crystal assuming the skyrmion to be a 3-q phase-coupled superposition of helical spirals.

4.1. Helimagnons

The dispersion of the excitations of the magnetic spiral in the multi-domain helimagnetic phase was first mapped out by Janoschek et. al [15] in zero field where the magnetic domains align along the <111> directions. The resulting spectra are complex superpositions of the contributions of 4 chiral domains. Janoschek et al. succeeded to describe the dispersions in terms of a universal Ginzburg-Landau theory which depends only on known macroscopic material parameters. The helical ground state is obtained by a minimistaion of the free energy functional [16]

$$F(M) = \int d^3r \left(f_0 + f_{demagnetization} + f_{anisotropy} + f_{correction} \right) \tag{2}$$

with the magnetization $M(r)$ being the order parameter. The free energy f_0 consists of the exchange interaction J, favoring a parallel magnetization and the DMI D, favoring a perpendicular alignment, resulting in a spiral with the pitch length $k_h = D/J = 0.0036\text{Å}^{-1}$. The anisotropy terms then aligns the spiral along the <111> direction. Recently, Schwarze et al. [16] performed measurements at q = 0 for all magnetic phases in MnSi, $Fe_{1-x}Co_xSi$ and Cu_2OSeO_3 using microwaves. For an adequate description of the measurements, the theory had to be modified ($f_{correction}$) to include a dipolar magnetic term and a cubic anisotropy. The first term being essential for a realistic description of the spectra, the latter term giving a small correction.

In order to follow up the detailed dependence of the helimagons on momentum q and B-field, we induced a single-domain state which was achieved by application of a small magnetic field of approximately B_{c1} = 100 mT, (see Kugler et al. [17]). This procedure allowed to directly identify the individual bands of the theoretically proposed band structure (see Fig. 2) for finite q. We showed that the theory required further refinements by introducing a higher-order correction term $\Delta F = \rho_s A(\nabla^2 n)^2/(2k_h 2)$ in the free energy with the unit vector n pointing along the magnetization M and ρ_s the stiffness density [17]. Fitting the data yields for the dimensionless constant A the value $A = -0.0073 \pm 0.0004$ providing an excellent description of the data. MnSi can therefore be viewed as a 1-dimensional helimagnetic crystal. The low energy bands are basically equivalent to the physics of particles trapped in a one-dimensional potential. This result is highly relevant because the skyrmions in MnSi are related particles that are trapped by a similar potential and are stabilized on the energy scales of the helimagnetic state.

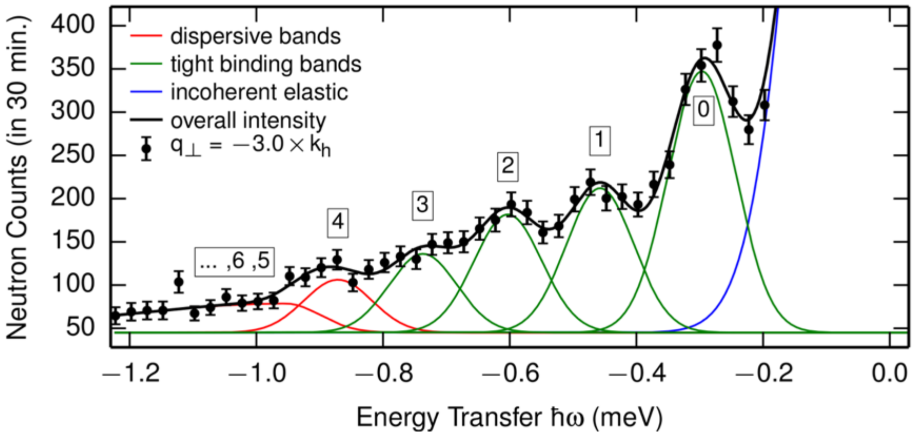

Figure 2. The figure (reproduced from [17], (c) by the American Physical Society, USA) shows an energy scan measured at T = 20 K and for a fixed momentum transfer q perpendicular to the wave vector k_h of the helix. Five bands are readily resolved and can be parametrized using a multi-Gaussian spectrum. The elastic peak appears due to incoherent scattering.

4.2. Conical and in the field-polarized phase

The dispersion branches in the field-polarized ferromagnetic phase were originally mapped out by Ishikawa et al. [4,18], however, only at momentum transfers $q \gg k_h$ as they performed experiments using thermal triple-axis spectroscopy, restricting the accessible energies to a range well above 1 meV. In a recent study, our group succeeded in quantitatively verifying the helimagnon model [19] under application of magnetic fields, starting from the helimagnetic-conical phase transition at B_{c1}, moving up to the second critical field B_{c2} [20] at much smaller momentum transfer q and energies. Observing the transition from the band structure in the helical and conical phases towards a single mode in the field-polarized ferromagnetic phase, we demonstrated an excellent agreement of the theoretically predicted helimagnon energies and their spectral weights.

Fig. 3 shows the field dependence of the helimagnons for momentum transfers parallel (left-hand side) and perpendicular (right-hand side) to the helix wave vector k_h. The perpendicular momentum transfer leads to the band structure, which condenses towards a single field-polarized magnon on increasing the magnetic field. A peculiarity resulting from the non-centro symmetric space group of MnSi shows for momentum transfers parallel to the helix. Here, the spectral weights begin to reshuffle in an asymmetric way with increasing field [20]: Magnons are created with a different weight factor than they are annihilated. In the field-polarised phase all the dispersion branches which lose spectral weight on increasing fields do not exist anymore. This leads to the extreme situation that field-polarized magnons are created with a different energy than they are annihilated [21].

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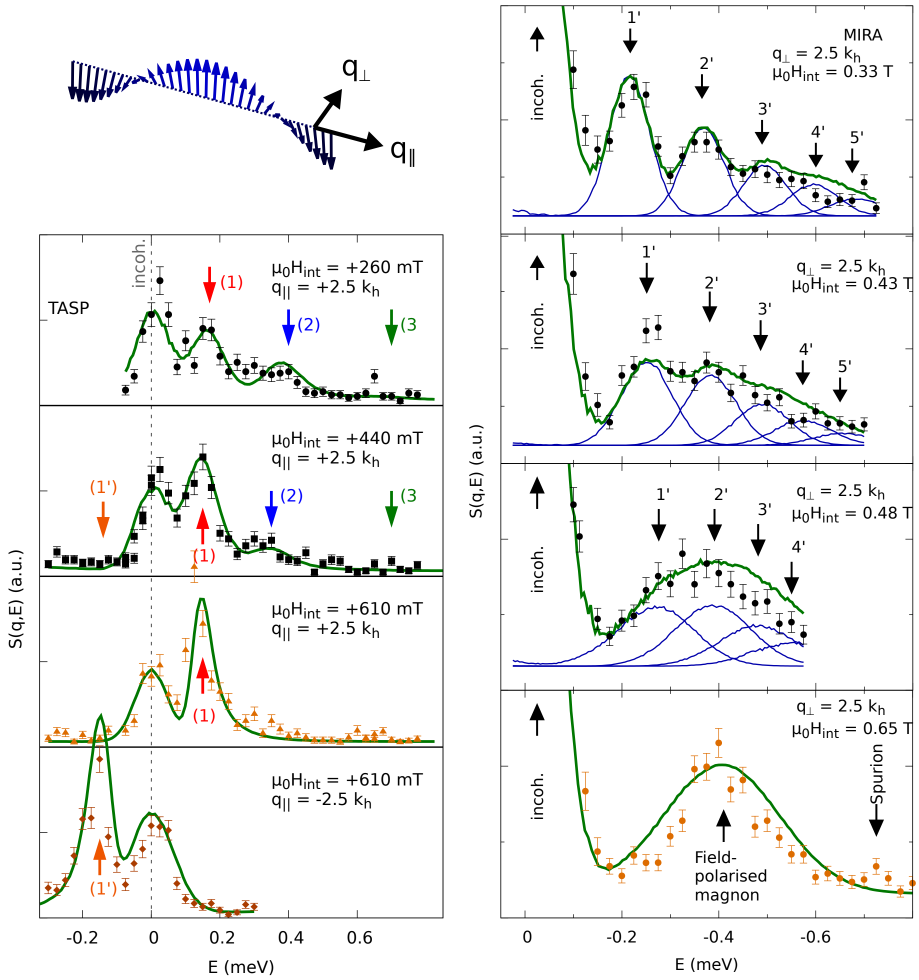

Figure 3. The figure (reproduced from [20], (c) by the American Physical Society, USA) shows the field-dependent evolution of the helimagnon excitations for momentum transfer parallel (left) and perpendicular (right) to the helix.

4.3. Skyrmion phase

The magnons in the skyrmion phase were first discovered by M. Janoschek [22] and are currently being mapped out and described theoretically [23]. Theoretically [19] the magnons comprise very complicated spectra which cannot be resolved individually using current spectrometers. Interestingly, the distribution of spectral weights imposes a dispersion-like pattern onto the densely-packed allowed branches and itself is within the limit of high-resolution neutron spectroscopy. At q=0, the spectral weights condense at three distinct energies which have been identified as a clockwise, a counter-clockwise and a breathing motion of the skyrmion vortex [16]. While these magnons at finite q [23] possess a still lower spin-wave stiffness than in the conical phase, we recently found that the magnons of both phases share the same asymmetric phenomena. Like the magnons in the conical phase, the spectral weight for creating a magnon in the skyrmion phase is markedly different than for annihilation as long as the momentum transfer is along the normal of the skyrmion plane [24].

5. Conclusions

Even though MnSi has now been measured for 50 years and several times altered our understanding of helimagnetic systems with long spiral pitches, it is still a very interesting material for learning more about the structure and dynamics of incommensurate magnetic structures. Here we have shown how MnSi became a prototype for helimagnetic systems and how the dynamics in this

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phase can be understood from the hierarchy of the 3 energy scales involved. We further reviewed the discovery of the skyrmions, which gave a huge boost in the understanding and use of topologically protected spin systems in the last decade. So far the magnetic system is only fully understood for ambient pressure. There is still a lot to learn for the development of the spin system with increasing pressure up to the critical pressure at 14.6 kbar where the long reaching interactions break down and only paramagnetic behavior survives. Especially interesting is the question of how much the low-pressure phases of the helimagnetic and skyrmionic phase contribute to the development of the non-Fermi liquid phase in MnSi.

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Abbreviations

The following abbreviations are used in this manuscript:

DMI	Dzyaloshinsky-Moriya interaction
SOC	Spin-orbit coupling

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