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Corrigendum

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The authors originally listed in the above article (C Pfleiderer, A Neubauer, S Mühlbauer, F Jonietz, M Janoschek, S Legl, R Ritz, W Münzer, C Franz, P G Niklowitz, T Keller, R Georgii, P Böni, B Binz, A Rosch, U K Rößler and A N Bogdanov) is incorrect.

The correct author list is C Pfleiderer, A Neubauer, S Mühlbauer, F Jonietz, M Janoschek, S Legl, R Ritz, W Münzer, C Franz, P G Niklowitz, T Keller, R Georgii, P Böni, B Binz, A Rosch.

Due to a miscommunication amongst the authors, Ulrich K Rößler and Alexei N Bogdanov became aware of this paper only when it was published. They did not take part in the writing of this text, and do not bear any responsibility for its contents.

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Quantum order in the chiral magnet MnSi

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Abstract
Systems lacking inversion symmetry, such as selected three-dimensional compounds, multilayers and surfaces support Dzyaloshinsky–Moriya (DM) spin–orbit interactions. In recent years DM interactions have attracted great interest, because they may stabilize magnetic structures with a unique chirality and non-trivial topology. The inherent coupling between the various properties provided by DM interactions is potentially relevant for a variety of applications including, for instance, multiferroic and spintronic devices. The, perhaps, most extensively studied material in which DM interactions are important is the cubic B20 compound MnSi. We review the magnetic field and pressure dependence of the magnetic properties of MnSi. At ambient pressure this material displays helical order. Under hydrostatic pressure a non-Fermi liquid state emerges, where a partial magnetic order, reminiscent of liquid crystals, is observed in a small pocket. Recent experiments strongly suggest that the non-Fermi liquid state is not due to quantum criticality. Instead it may be the signature of spin textures and spin excitations with a non-trivial topology.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In recent years the systematic synthesis of new materials and the equally systematic screening of their physical properties have led to the discovery of new forms of electronic order. Well known examples include unconventional forms of superconductivity [1], nematic electron liquids [2] or peculiar forms of non-Fermi liquid behavior [3]. As part of these studies there is growing interest in the coupling between different order parameters and complex forms of order. All of these examples are driven by many-body interactions, where the microscopic nature of these states is far from being understood experimentally or theoretically.

A materials class of special interest are systems lacking inversion symmetry, because in general they support Dzyaloshinsky–Moriya (DM) spin–orbit interactions. On the one hand DM interactions play an important role in the coupling of different ferroic properties in many multiferroics [4]. On the other hand, it has been suspected for a while that DM interactions may stabilize non-trivial magnetic structures [5–10]. Simple examples for systems with DM interactions are selected multilayers and surfaces. However, three-dimensional compounds lacking inversion symmetry are also quite common; amongst the 230 crystallographic space groups 65 are chiral. This suggests that phenomena related to a lack of inversion symmetry, in principle, are rather common.

In the following we review the properties of the, perhaps, best studied three-dimensional compound lacking inversion symmetry, the itinerant-electron magnet MnSi [11]. This compound may be considered a model system that allows one to explore how weak DM interactions may profoundly change the bulk and transport properties of a material. Recent experiments have motivated speculation that these changes may be the result of non-trivial spin structures. The paper proceeds as follows. In section 2 we introduce the ambient pressure properties of MnSi. Section 3 is dedicated to a review of the pressure versus temperature phase diagram at zero field, which displays an abrupt transition of the electrical resistivity from a Fermi liquid to a non-Fermi liquid temperature dependence. The observation of partial magnetic order in the NFL regime, reviewed in section 4, strongly suggested that the DM interactions are somehow essential for an understanding of the NFL resistivity. We finally review the most recent work, which suggests the formation of a genuinely new state at high pressures as seen by Larmor diffraction and the extrapolated magnetic field versus pressure phase diagram in small angle neutron scattering in section 5. In section 6 we give a short account of theoretical speculation that this new state in MnSi at high pressures may be related to complex spin textures with a non-trivial topology. We conclude our review in section 7 with a summary of open issues that are currently under investigation.

2. Chiral magnetic order in MnSi

Following the pioneering work by Lifshitz, Dzyaloshinsky [12] and Moriya [13] in the 1960s, the first material in which a homochiral spin spiral could be identified was the transition metal compound MnSi [14]. In the cubic B20 crystal structure of MnSi the atoms are slightly rotated away from centrosymmetric positions, thus breaking inversion symmetry. In other words the crystal structure of MnSi is already chiral. As an empirical observation it is interesting to note that the crystal structure of all MnSi samples studied to date have the same handedness.

MnSi exhibits strongly exchange enhanced paramagnetism down to a magnetic phase transition at $T_c = 29.5$ K. Due to the lack of inversion symmetry, the spin–orbit coupling in MnSi assumes the Dzyaloshinsky–Moriya form, $\vec{m} \cdot \nabla \times \vec{m}$, favoring the perpendicular spin orientation. In combination with ferromagnetic exchange the DM interaction stabilizes a helical spin spiral below T_c. By comparison to the typical atomic distances or the lattice constant the wavelength of this spin spiral is very long, $\lambda_h \approx 180$ Å. At ambient field the propagation axis of the helix is very weakly pinned to the cubic space diagonal $\langle 111 \rangle$.

Shown in figure 1 is the magnetic phase diagram of MnSi. Well below T_c an applied magnetic field, $\vec{B}$, unpins the helical order and aligns its wavevector $\vec{Q}$ parallel to the field, $\vec{Q} \parallel \vec{B}$, for a field exceeding $B_{c1} \approx 0.1$ T [14, 16–21]. This state is referred to as the conical phase. For a magnetic field exceeding $B_{c2} \approx 0.6$ T the effects of the DM interaction are suppressed, giving way to a spin-aligned (ferromagnetic) state. For temperatures just below T_c an additional phase, referred to as the A-phase, is stabilized in a finite field interval as shown in figure 1 for $\vec{B} \parallel \langle 100 \rangle$ [14–24]. It was believed for a long time that the A-phase could be explained by a single-$\vec{Q}$ helix, where $\vec{Q}$ is perpendicular to the applied field [17, 20, 21]. Recently we have established that this view is incomplete and that the A-phase, in fact, represents a skyrmion lattice [15, 25].

For a long time the magnetic properties of MnSi received most attention as a model system exhibiting itinerant-electron spin fluctuations. Pioneering inelastic neutron scattering studies established the spectrum of spin fluctuations for the first time in an itinerant-electron magnet, essentially across the entire Brillouin zone [26]. Remarkably, the spin fluctuations in the paramagnetic state are well described by the double Lorentzian line form of an over-damped harmonic oscillator, also referred to as Moriya–Kawabata fluctuations [27]. Because the helical modulation in the ordered state occupies only a small fraction of the volume (<0.1%) of the Brillouin zone it was initially ignored. The question of the low-lying magnetic excitations was only recently revisited theoretically [28, 29] and experimentally. These studies reveal the existence of helimagnons [30, 31], i.e., Goldstone modes characteristic of long-wavelength helimagnets.

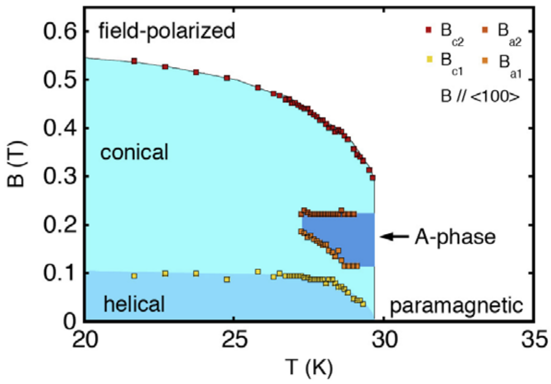

Figure 1. Magnetic phase diagram of MnSi as reported in [15, 16]. For $B = 0$ helimagnetic order develops below $T_c = 29.5$ K. Under a magnetic field the helical order unpins and aligns along the field above B_{c1}; above B_{c2} the helical modulation collapses. In the conical phase the helix is aligned parallel to the magnetic field. The A-phase was recently identified as a skyrmion lattice [15]. The transition fields shown here have been inferred from the AC susceptibility, where the DC and AC field were parallel $\langle 100 \rangle$ [16].

In the mid 1980s Lonzarich and co-workers showed that a self-consistent phenomenological framework accounts correctly for the transition temperature and the enhanced ratio of the fluctuating Curie–Weiss moment to the small ordered moment in MnSi and Ni_3Al [32, 33]. Perhaps most remarkably, the effects of interactions causing non-linear behavior and magnetic order in this model may still be accounted for by a linear response that is self-consistently renormalized. This renormalization can be made *quantitatively* consistent with the magnetic transition temperature.

The excellent quantitative account of the important energy scales in MnSi and related ferromagnetic compounds motivated a research program into the nature of the zero temperature phase transitions of itinerant-electron ferromagnets. The purpose of these studies was the identification of a ferromagnetic quantum critical point. For early results see [34, 35].

3. Emergence of an extended non-Fermi liquid regime

The question of a 'ferromagnetic' quantum critical point was first studied in MnSi [34, 35]. Based on the excellent agreement with ferromagnetic spin fluctuation theory any additional complexities due to the Dzyaloshinsky–Moriya spin–orbit coupling were initially ignored. The first studies of the electrical resistivity appeared to be consistent with a QCP

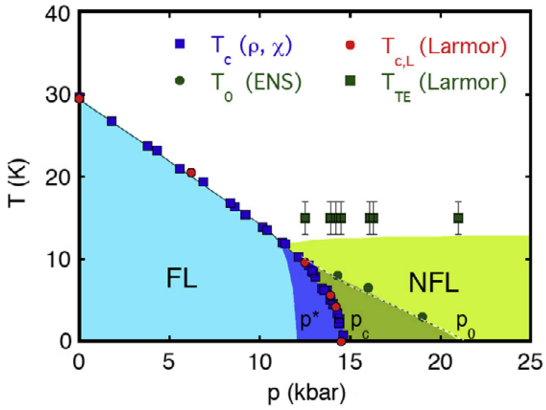

Figure 2. Phase diagram of MnSi as a function of pressure reported in [39]. Data for T_c are based on the resistivity and the susceptibility reported in [38]. T_0 is based on elastic neutron scattering reported in [40]. In the regime of Fermi liquid (FL) resistivity, dark shading shows the regime of phase segregation seen in μ-SR [41]. In the regime of non-Fermi liquid (NFL) resistivity, dark shading indicates the regime of partial order [40]. The transition temperature $T_{c,L}$ observed in the lattice constant by Larmor diffraction is in excellent agreement with previous work. The cross-over temperature T_{TE} represents the appearance of lattice contraction as measured by Larmor diffraction.

at p_c [35] (figure 2). Below p_c the electrical resistivity follows the quadratic temperature dependence of a Fermi liquid, where the coefficient of the quadratic temperature dependence was found to diverge for $p \to p_c$ [35, 36].

However, the AC susceptibility, as measured in very pure single crystals, showed that the magnetic phase transition at T_c for increasing pressure changes from second order to first order at $p^* \approx 12$ kbar [37, 38]. Further support for the absence of a ferromagnetic QCP at p_c was subsequently obtained in terms of the magnetic field dependence of the susceptibility. Here itinerant metamagnetism confirmed the presence of a local minimum in the free energy [16, 38].

Based on the first order behavior in the susceptibility and the spectrum of spin fluctuations expected at high pressures as inferred from the measured uniform susceptibility, it was anticipated that the temperature dependence of the electrical resistivity above the critical pressure should be that of a Fermi liquid. In contrast, subsequent studies of the temperature dependence of the electrical resistivity in MnSi for $p > p_c$ established a stable, extended non-Fermi liquid temperature dependence of the electrical resistivity $\Delta\rho \propto T^{3/2}$ [42–44].

The non-Fermi liquid resistivity emerges below roughly ~10 K, when T_c has been suppressed sufficiently under pressure. Near p_c the non-Fermi liquid resistivity has been followed down to the low mK range, thus ranging over nearly three orders of magnitude in temperature [42]. Measurements down to about 0.05 K establish that the $T^{3/2}$ dependence persists at least up to 30 kbar (about two times p_c) [42–44]. Measurements down to 1.5 K suggest that the resistivity exponent increases from ~1.5 above a pressure of the order ~45 kbar [45].

There is currently no explanation for the pressure dependence of the electrical resistivity in MnSi in the low temperature limit. Empirically, the stability of the non-Fermi liquid temperature dependence raises the question if it is the signature of a novel metallic state. In the following we review recent neutron scattering studies that clearly point at a transition of the ground state from a Fermi liquid resistivity to a metallic state with a genuine non-Fermi liquid resistivity.

4. Partial magnetic order in the non-Fermi liquid state

As spin fluctuations at ambient pressure are quantitatively consistent with T_c, and T_c may be tracked very well as a function of pressure in the electrical resistivity and susceptibility, one naively expects the ordered magnetic moment to vanish at p_c, giving way to an abundance of quantum critical spin fluctuations that would somehow explain the NFL resistivity. However, several neutron scattering studies in the 1990s failed to identify the nature of the magnetic fluctuations at high pressures (e.g., [46]). Based on these studies we decided to track the magnetic order as a function of pressure all the way up to p_c as a prerequisite for further inelastic experiments.

To our surprise we found that the ordered magnetic moment, measured in elastic neutron scattering with an energy resolution $\Delta E \approx 0.05$ meV does not vanish at p_c [40]. Instead we observed a scattering intensity everywhere on the surface of a small sphere in reciprocal space. The integrated scattering intensity near p_c is still large and only several tens of percent reduced from its ambient pressure value. This suggests that T_c stops tracking the ordered magnetic moment in a simple manner in the vicinity of p_c.

As the second remarkable feature we found that the scattering intensity is smeared out over the surface of a small sphere with broad maxima in the ⟨110⟩ direction. Just below p_c the intensity in the ⟨110⟩ direction and the ⟨111⟩ directions have different temperature dependences, where the signal observed for ⟨111⟩ tracks T_c as measured in the resistivity and susceptibility. In contrast, the smeared out intensity that is strongest in the ⟨110⟩ direction appears below a cross-over temperature T_0, which extrapolates to zero at $p_0 \approx 21$ kbar.[4]

Detailed scans establish that the scattering intensity on the surface of the sphere in reciprocal space is resolution limited for longitudinal scans, while there is intensity everywhere on the surface of the sphere in transverse scans. Because the scattering intensity is located on the surface of the sphere in reciprocal space, we interpret the transverse scattering intensity as not being the signature of short-range order. Instead we argue that the system exhibits on local scales long-range order in all three spatial directions. We therefore refer to the magnetic properties seen in neutron scattering below T_0 as partial magnetic order.

[4] It has recently been noted [47] that the neutron intensity for an isotropic surface of scattering intensity in reciprocal space due to randomly oriented helices, when measured near a ⟨110⟩ nuclear Bragg peak, displays a maximum for the ⟨110⟩ direction due to a selection rule. However, the intensity would drop to half of its value for directions perpendicular to the ⟨110⟩ direction, while we observe a much faster decrease. This establishes, that the maxima for ⟨110⟩ are not an experimental artifact.

The observation of partial magnetic order raises the question of the underlying spin structure. In the simplest interpretation the observed behavior may be the result of either of two extremes. First, the helical structure may have broken up into a multi-domain state in which the helix ends abruptly between domains. This state would be stabilized by topological defects between the domains. In a second, and alternative, scenario the helix direction has unlocked from the ⟨111⟩ and no longer exhibits strict directional order. Such a state is said to exhibit so-called topological order.

The electrical resistivity and the susceptibility exhibit strong signatures at T_c and the metamagnetic transition. In contrast, both properties are featureless at the cross-over at T_0. As a possible explanation, we noted early on that the partial magnetic order in fact may fluctuate on the timescale relevant for transport, but appears quasi-static below T_0 for the energy resolution of 0.05 meV [40]. This conjecture is consistent with recent μ-SR measurements, which suggest a decreasing volume fraction of helical order between 12 kbar and p_c and that the partial order fluctuates slowly on a timescale between 10^{-11} and 10^{-10} s [41].

It appears reasonable to assume that the partial magnetic order is a key signature of the non-Fermi liquid resistivity. An unlocking of the helical order was neither anticipated experimentally nor theoretically. Possible scenarios included in particular: hidden forms of quantum criticality, reviewed in the following, or the stabilization of a new state with low-lying excitations that result in the non-Fermi liquid resistivity.

5. Non-Fermi liquid metal without quantum criticality

Several complementary studies have recently established that the non-Fermi liquid resistivity emerges under pressure without quantum criticality. These studies reported the pressure dependence of muon-spin-rotation (μ-SR) [41], the temperature dependence of the lattice constant using Larmor diffraction [39] and small angle neutron scattering under pressure and magnetic field [48].

Measurements of the AC susceptibility established, early on, that the transition changes from second to first order at 12 kbar [37, 38]. The first order behavior is consistent with the observation of itinerant-electron metamagnetism. The first evidence indicating phase separation was observed in neutron scattering, notably the distinctly different temperature dependence of scattering intensity on the surface of the sphere in reciprocal space, as mentioned above. Comprehensive evidence establishing phase separation was observed in a μ-SR study under pressure. In this study only the helical order could be observed. Above p^* the volume fraction was found to decrease until it vanished at p_c. The lack of evidence for the partial order suggests that the partial order is in fact slowly fluctuating and not static. This underscores our initial speculations that the cross-over line at T_0 is related to the fluctuation rate of the partial order as compared to the timescale of the neutron scattering.

To clarify if the non-Fermi liquid resistivity in MnSi is the property of a novel metallic state or related to a QCP one requires measurements of a variable that is conjugate to the control parameter. In the case of MnSi this control parameter is pressure, and the conjugate variable is the unit cell volume or, equivalently, the lattice constants. However, the changes of the size of the sample in pressure experiments cannot be measured with conventional capacitive dilatometry (at low temperatures the pressure medium is solid, even when using helium), and recent efforts using strain gauges glued to the sample are sensitive to strains due to local differences in compressibility. We have therefore used neutron diffraction as an elegant method to determine changes of lattice constant [39]. To clarify the evolution of the lattice constant in MnSi across p_c a resolution better than 10^{-5} is necessary. At the same time a high neutron intensity is necessary for a comprehensive study. This cannot be achieved routinely in conventional neutron diffraction. We have therefore used so-called neutron Larmor diffraction, a novel polarized neutron scattering technique [49].

At ambient pressure the lattice constant of MnSi decreases quadratically with temperature down to T_c. Below T_c a large spontaneous lattice expansion is observed. With increasing pressure the onset of this spontaneous lattice expansion tracks the pressure dependence of T_c as determined from the resistivity and susceptibility. When T_s is suppressed below about 15 K the lattice constant above T_c displays not only the quadratic temperature dependence. Below $T_{TE} \approx 15$ K an additional gradual volume contraction emerges. With respect to the quadratic temperature dependence, the lattice constant hence changes sign from lattice expansion to lattice contraction at T_{TE}; the thermal expansion changes sign from negative to positive. Such a change of sign is also expected at a QCP [50, 51]. However, for a QCP it occurs at $T = 0$ and not at a finite temperature as observed here. Our data in turn establish that the transition at p_c is first order.

Furthermore, at the onset temperature T_0 of the partial order the lattice constant is featureless. In contrast, if the partial order on local length scales was akin to the helical order observed at ambient pressure, we would expect a strong signature. This clearly suggests that the partial order represents a novel state.

Finally, in the range $p^* < p < p_c$ the extrapolated spontaneous lattice expansion decreases and vanishes at p_c, consistent with a decreasing volume fraction of helical order inferred from μ-SR. This suggests a phase separation of the magnetic properties into a volume fraction with helical order and a corresponding volume fraction with the high pressure state.

Indirect evidence that the partial magnetic order represents not just meandering helices, or a single-Q complex multi-domain state, is also seen in the magnetic field dependence measured by means of small angle neutron scattering. For experimental details we refer to [48]. In our studies we measured the magnetic field dependence of the helical components of the spin structure as aligned parallel to a magnetic field that was oriented perpendicular to the incident neutron beam.

As its key result this study revealed a rather peculiar form of the magnetic field dependence of the helical components of the spin structure. Just above the critical pressure in the regime

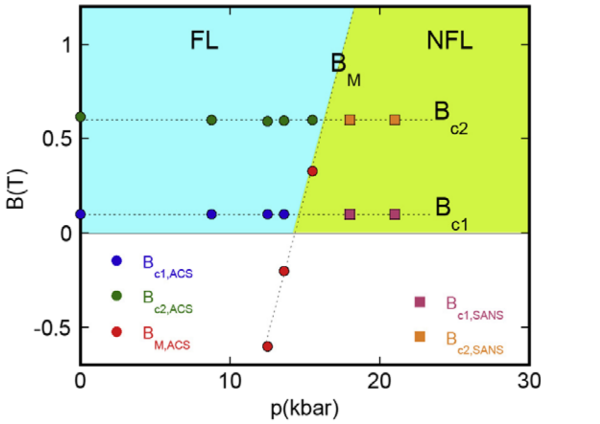

Figure 3. Extrapolated magnetic field versus pressure phase diagram for $T = 0$ as reported in [48]. Characteristic field values seen in the resistivity, susceptibility, and small angle neutron scattering. Blue and green shading indicate the regime of the FL and NFL resistivity, respectively, which are separated by the itinerant metamagnetic transition [16, 38]. Note that there is no significant pressure dependence B_{c1} and B_{c2} as seen in SANS, while data reported in [43] suggest that the NFL resistivity survives even above B_{c2}.

of the partially ordered state increasing the applied magnetic field in the zero field cooled state did not generate substantial alignment of the helical components. In contrast, when decreasing the magnetic field from a field value well above B_{c2} helices aligned spontaneously parallel to the applied field below B_{c2}. When further decreasing the field the scattering intensity vanished in a manner that suggested an intrinsic trend towards a different ground state.

Furthermore, when combining the small angle neutron scattering with the magnetic field, temperature and pressure dependence of the AC susceptibility and the resistivity, it is possible to construct the magnetic field versus pressure phase diagram shown in figure 3. While the characteristic field values of the appearance and suppression of the conical state, B_{c1} and B_{c2}, are only weakly affected by pressure, the non-Fermi liquid resistivity appears to survive to higher magnetic fields. This implies that the non-Fermi liquid resistivity is not simply the result of disordered helices. If it is connected to the DM interactions, the resulting state must be more stable, thus suggesting that it differs from simple helical order.

6. Search for non-trivial spin structures

The pressure dependence of MnSi provides compelling evidence of the formation of a novel form of order in which chiral Dzyaloshinsky–Moriya interactions may be a key ingredient. This raises the conceptual question what types of spin-order may be expected other than a plain unpinning of the helix or a multi-domain state. On a more general level these issues are relevant to all systems in which the Dzyaloshinsky–Moriya spin–orbit interaction may be expected. In the following we briefly summarize some of the main proposals.

Binz and Vishwanath have considered the stability of a multi-Q rather than the single-Q helix observed in MnSi at ambient pressure [8, 52]. For the cubic B20 environment of MnSi they find that several different multi-Q states may become stable. The partial magnetic order is explained in terms of a body-centered triple-Q state that is not perfectly locked to the crystal lattice. On a local scale the spin structures of the bcc spin crystal resemble, thereby, topologically stable knots as illustrated in figure 4. However, as there are lines in the spin structure where the magnetization vanishes, the spin crystals considered in these studies do not represent objects with a non-trivial topology in any strict sense. Instead they are stabilized by the basic symmetry properties of the bcc structure rather than their topology.

In another study the nucleation of helical order has been compared with a multi-dimensional modulation [5]. It was believed for a long time that two-dimensional modulations cannot form spontaneous ground states. The lack of stability can be traced to the cost of energy for double twisting at distances that are large compared with the nucleation point. In a coarse-graining continuum model describing the magnetization on intermediate scales a softened modulus of the magnetization may be expected. Using a non-analytic

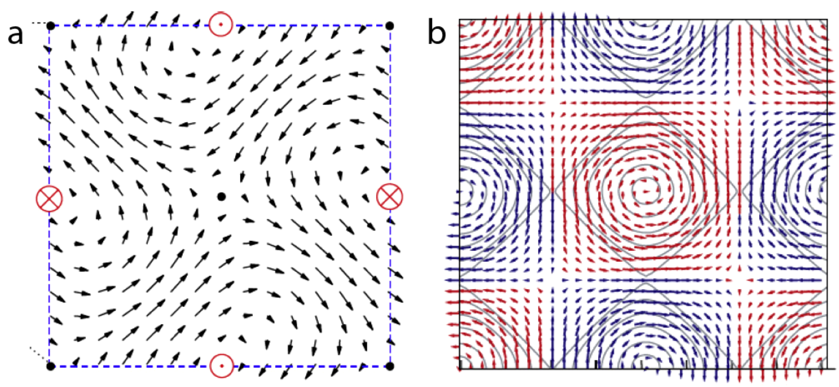

Figure 4. (a) Magnetization of a type 1 body-centered cubic spin crystal (a triple-Q state) described in [8, 52]. The line of sight is parallel to the ⟨100⟩ direction. The vectors denote the in-plane magnetization. The node lines are the centers of antivortices and the directed red lines are the centers of meron configurations. (b) Structure of a two-dimensional skyrmion lattice, derived as a minimum energy solution for the model described in [5]. Shown is the projection of the magnetization vectors, where red/blue signals a local magnetization direction with positive/negative components. The contour lines stand for constant magnetization modulus in the skyrmion cores. The result applies to thin magnetic films with broken inversion symmetry.

gradient term to describe the effects of a softened modulus in a Ginzburg–Landau model, it was possible to show theoretically that double-twist structures form spontaneous ground states. However, as a technical concern the question has been raised if this non-analytic term may lead to uncontrolled results in the presence of very strong fluctuations in the vicinity of T_c.

These studies along with models considering higher order DM interactions [9, 10] all arrive at the conclusion that one may expect non-trivial spin structures in MnSi. All of these spin structures are contenders as a possible explanation for the partial order and the non-Fermi liquid resistivity. Future research will have to clarify, if, and which of the proposed states may be stabilized as a function of temperature, magnetic field and pressure.

7. Open questions

The open challenges in MnSi as a model system for the rich physics of DM interactions may be summarized as follows. First of all it is necessary to advance the understanding of the nature of the partial magnetic order. For instance, it will be essential to determine the precise temperature, pressure and magnetic field dependence of the dynamical properties of the partially ordered. Second, there is currently no theoretical account for the NFL resistivity and how it may be related to the partial magnetic order. Third, and on a more general note, the theoretical work summarized above suggest that non-trivial spin structures may exist in many materials, where they include topologically stable spin structures such as merons or skyrmions. This raises the need for microscopic experimental evidence.

An entirely different line of work finally addresses possible analogies between ferromagnetic domain walls (Bloch walls) and helimagnetic order [53–56]. Here it has been suggested that so-called spin torque effects may be very strong indeed in MnSi and related compounds. Experimental studies of these spin torque effects may also pave the way to an understanding of the NFL resistivity. More importantly, perhaps, they may also establish a link between fundamental studies in itinerant helimagnets and issues relevant for technical applications.

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