PARTIAL ORDER IN AN ITINERANT-ELECTRON MAGNET

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The transition metal compound MnSi is perhaps the most extensively studied of the itinerant-electron magnets apart from the elemental metals iron, cobalt, nickel and chromium. It is a stable, congruently melting compound that can be produced at high purity and high crystalline perfection in the cubic B20 structure. A large body of information on MnSi is available that includes thermodynamic and transport data as a function of temperature, magnetic field and pressure, and microscopic data based on neutron scattering, nuclear magnetic resonance and quantum oscillations. Though relatively simple, the B20 structure is somewhat unusual in that it lacks inversion symmetry, so that the weak spin-orbit interactions have the Dzyaloshinsky-Moriya (DM) form. In turn, for the $P2_13$ space group of MnSi this causes a helical twist of wavelength λ? ˜ 170 Å of the small ordered magnetic moments $\boldsymbol{m}$ = 0.4 μ_B/f.u. (formula unit) that is locked to $\boldsymbol{Q}$ = <111>, where $\boldsymbol{m} \perp \boldsymbol{Q}$.

Hydrostatic pressure as a clean tuning technique that does not interfere with the crystalline perfection is well established to induce highly reproducible variations of the bulk magnetic properties of MnSi, notably the suppression of the bulk magnetic transition temperature above p_c = 14.6 kbar. [1,2]. This has motivated us to investigate the magnetic state of MnSi microscopically at high pressure using neutron diffraction. Experiments were carried out on the cold thermal neutron triple axis spectrometer 4F1 at the Laboratoire Léon Brillouin where we achieved a high resolution in energy better than 50 μeV. Use of a triple axis spectrometer allowed a significant reduction of the diffuse scattering by the miniature pressure cell.

For our study the helical modulation at ambient pressure proves to be instrumental, because the corresponding Bragg scattering at Q ˜ 0.037 $Å^{-1}$ is confined to a small volume of reciprocal space, making it easy to track the magnetic order at high pressure. Data were collected near the [110] lattice Bragg point, where the exceptional structural perfection of our sample, evident from a resolution limited mosaic spread h ? << 0.2°, allowed for unambiguous results. Shown in Fig. 1 is an illustration of our key results. In the temperature versus pressure plane the Curie temperature of MnSi, T_c ˜ 29.5 K, determined in resistivity and susceptibility measurements, falls monotonically with pressure, p [1]. The transition is second order up to p^* ˜ 12 kbar and weakly first order between p^* and p_c, where T_C vanishes. At ambient pressure (top left corner of Fig. 1) resolution limited magnetic Bragg reflections of a coherence length x ? > 1200 Å at $\boldsymbol{Q}$ = 0.037 $Å^{-1}$ (111) are observed, that are characteristic of conventional three-dimensional long-range order. This is in excellent agreement with previous studies [3,4]. At p = 14.3 kbar, where T_C ˜ 3.3 K, the lattice constant is reduced by only 0.4 % in general agreement with the compressibility. For this pressure we observe neutron scattering intensity that is only resolution limited in the radial direction of a sphere with Q = 0.0422 $Å^{-1}$. The intensity is elastic at the limit of our energy resolution of 50 μeV.

To explore the distribution of intensity on the sphere of radius Q further we have performed various longitudinal, transverse and vertical scans with respect to <111> and <110>. For longitudinal scans we observe resolution limited intensity corresponding to x? > 1200 Å as expected of conventional long range magnetic order. In striking contrast typical transverse scans show intensity over the full arc between <111> that is strongly enhanced at <110>. Scans vertical to the arc between <111> and the longitudinal direction at (110) indicate a gradual decrease along this direction as well. At 1.7 K and <111> we find that approximately 4% of the intensity of ambient pressure is left. A detailed account of the T dependence may be found elsewhere [5].

The total integrated scattering intensity over the sphere at 1.7 K is conserved to within 10 % of that at ambient pressure. This is consistent with measurements of the bulk magnetic moment as function of pressure in a polarising magnetic field of 0.6 T [2] and suggests that we detect within a small margin of error all of the ordered moment present at ambient pressure.

Our data imply a very shallow minimum of the free energy for <110> that is characteristic of a competition of interactions. To shed light on the nature of this competition we note that the conservation of scattering intensity implies that the partial order, i.e. departure from 3*d* order, must be the result of topological defects weakening the spin rigidity. The reduction of T_C is in particular not the result of soft dynamical modes. In the simplest case these defects are the walls of the magnetic domains. Because the weight of intensity near <110> is incompatible with the DM interaction for the space group of MnSi a modification of these topological defects (domain walls) appears likely. For instance, it has been shown theoretically [6], that the competition of the DM interaction with the ferromagnetic stiffness for the case of MnSi may stabilize the formation of magnetic vortices.

In conclusion, when changing the lattice constant of MnSi by just 0.4 % using hydrostatic pressure, the elastic magnetic neutron diffraction intensities display sharp Bragg reflections in the longitudinal direction only, akin of partial order in liquid crystals. The observation of mesoscopic partial order in high quality single crystals of one of the most extensively studied itinerant-electron magnets has deep implications in the search for novel electronic states of intermetallic compounds in general [7]. For MnSi the temperature dependence of the electrical resistivity of the normal state ($T > T_C$) for $p > p^* \sim 12$ kbar suggests a diffusive motion of the electrons resulting from the interactions among the itinerant electrons themselves [2]. It is tempting to associate such a diffusive motion with the only partial order for which we have presented direct microscopic evidence. Further, quasi-elastic neutron spectra of intermetallic materials near quantum critical transitions may this way be recognized as forms of partial itinerant electron magnetism that are driven by unstable topological defects of the magnetic rigidity. This could potentially resolve the controversy of local quantum criticality [8] versus low dimensional dynamics [9] as well as uniting itinerant with local moment magnetism.

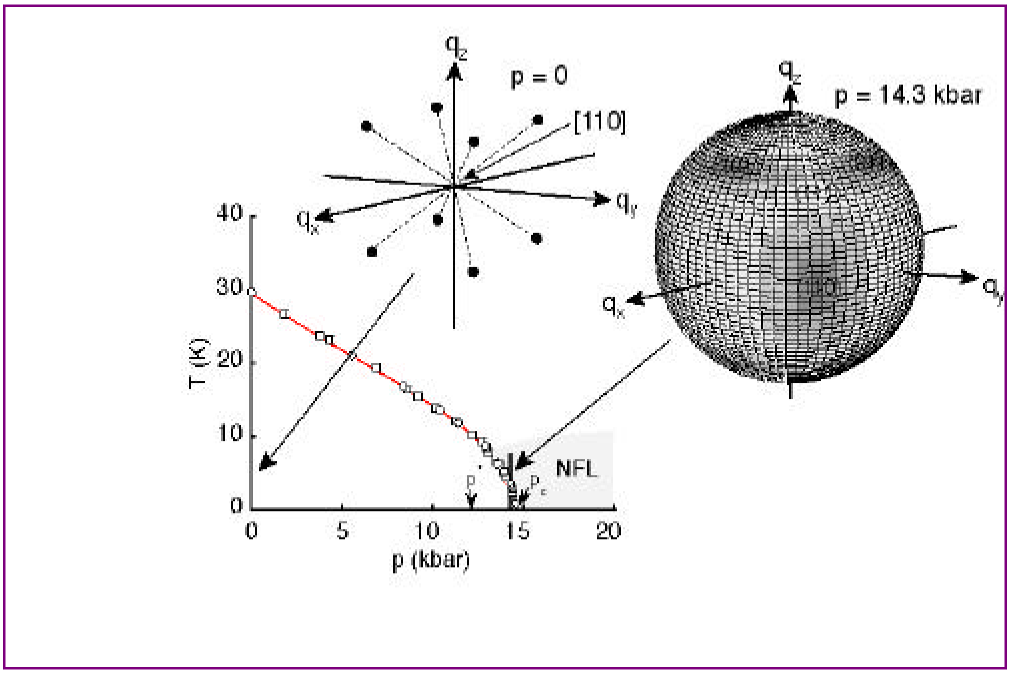

Figure 1. Schematic temperature *T* versus pressure *p* phase diagram of MnSi and qualitative illustration of the scattering intensity characteristic of the magnetic state. Data points of $T_C(p)$ are taken from reference [1]. Data were collected near the [110] lattice Bragg peak. At ambient pressure resolution limited magnetic satellites are observed at a distance Q=0.0375 Å^{-1}, characteristic of long range magnetic order. At high pressure intensity on a sphere of radius Q=0.0422 Å^{-1} is found with the highest intensity at <110> indicated by dark shading.

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