Inelastic neutron and x-ray scattering from incommensurate magnetic systems

Peter Böni1, Bertrand Roessli2 and Klaudia Hradil3

1 Physik Department E21, Technische Universität München, D-85748 Garching, Germany
2 Laboratory for Neutron Scattering, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland
3 X-Ray Center, Vienna University of Technology, A-1060 Vienna, Austria

Received 16 January 2011
Published 8 June 2011
Online at stacks.iop.org/JPhysCM/23/254209

Abstract

Neutrons and x-rays are powerful probes for studying magnetic and lattice excitations in strongly correlated materials over very wide ranges of momentum and energy transfers. In the focus of the present work are the incommensurate magnetic systems MnSi and Cr. Under application of a magnetic field, helically ordered MnSi transforms into a weak itinerant ferromagnet. Using polarized neutrons we demonstrate that the Stoner excitations are spin-flip excitations. The amplitude (longitudinal) fluctuations associated with the magnon modes are already strong far away from $T_c$. Interestingly, even the non-spin-flip excitations associated with the Stoner modes are observable. In Cr, we have observed Kohn anomalies in the phonon spectrum at those incommensurate positions in reciprocal space, where the spin density wave is observed. The corresponding phonon and magnon modes are not coupled. In addition, an anomalous softening of a transverse phonon branch along the N–H zone boundary line is observed that is caused by strong electron–phonon coupling. High resolution neutron scattering indicates that the low energy Fincher–Burke excitations may rather correspond to localized modes in momentum and energy and not to propagating collective modes. Finally, we demonstrate that in the near future it may become feasible to investigate excitations in very small samples, thus allowing us to measure the dynamics of strongly correlated materials under extreme conditions and in the vicinity of quantum phase transitions.

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

1. Introduction

Effects related to incommensurate magnetic or charge order have revealed many interesting effects in condensed matter physics. Recent examples include (i) high-$T_c$ superconductors, where antiferromagnetic fluctuations may be responsible for the pairing of the electrons [1], (ii) multiferroic compounds such as the manganites RMnO$_3$ (R: lanthanide, alkaline metals) [2] or borates [3], where a coupling between magnetic spiral-like order and the lattice or magneto-elastic coupling may lead to ferro-electric coupling, or (iii) itinerant magnets such as MnSi, where a skyrmion lattice has recently been identified [4]. By measuring the collective excitations in these materials it is possible to determine the energy scales that are responsible for the competing interactions, which are the origin for the novel incommensurate orderings.

Two prototypical magnetic systems are in the focus of our present interest, namely MnSi and Cr. MnSi serves as a prototype system for a weak itinerant magnet exhibiting modulated magnetically ordered phases [4] due to the competition between the Dzyaloshinskii–Moriya and the exchange interaction [5, 6]. The incommensurate antiferromagnet Cr is brought into focus due to the fact that the excitation spectrum shows striking similarities with the magnetic excitations in high $T_c$ superconductors [7]. In order to access the important energy ($E$) scales in these systems, various scattering techniques are to be used.

With the ongoing efforts to improve the $E$ resolution of inelastic x-ray beamlines approaching 1 meV, photons have become a valuable tool for investigating the lattice dynamics in solid state physics. In contrast to neutron scattering, the relative change of the energy of the photons during scattering is small, i.e. the scattering triangle remains essentially isosceles. Therefore, the phonon dispersions can be measured efficiently over large regions in momentum ($Q$) space using area detectors. Compared to investigations with photons,
Figure 1. The spin split bands are separated by the exchange splitting $\Delta$ as shown in (a). (b) shows the continuum of spin-flip excitations between the two bands, i.e. the Stoner continuum. The spin wave (SW) branch extends from $q = 0$ towards the Stoner continuum. (c) shows the continuum of non-spin-flip excitations within a band.

Figure 2. (a) Reciprocal lattice of helimagnetic MnSi showing the position of the magnetic satellites close to the nuclear Bragg reflections. (b) depicts the reciprocal lattice of Cr that was prepared in a single $Q$ state with $Q$ along the [100] direction. Therefore, magnetic satellites that are allowed by symmetry along the [010] and [001] directions are silent. The incommensurabilities are exaggerated by factors of 10 and 3 for MnSi and Cr, respectively.

the strengths of neutron scattering are a widely tunable $E$ resolution and the large interaction with the magnetic degrees of freedom. To take the benefit of the individual probes, we have applied both techniques to investigate the dynamics in Cr and MnSi as described below.

2. Magnetic scattering from itinerant ferromagnets

First we describe inelastic neutron scattering experiments that were performed with polarized neutrons in MnSi, thus providing a direct means to observe single-particle excitations in itinerant ferromagnets. According to the most simple model for ferromagnetism in delocalized systems, Stoner [8] assumed that, within a single band model, the interaction between the spin-up and spin-down electrons leads to a separation of the bands by an exchange splitting $\Delta$ (figure 1(a)). The energy gain is partly compensated by an increase of the kinetic energy of the conduction electrons. In this picture, long range ferromagnetic order is destroyed by the thermal excitation of electrons between the spin-split bands. Figure 1(b) depicts the continuum of single-particle excitations between the spin-up and spin-down bands and (c) shows the continuum of excitations within the bands. Also indicated in (b) is the spin wave branch of the collective excitations (magnons) that proceeds from $q = 0$ (Goldstone mode) towards the Stoner continuum.

Extensive measurements of the spin fluctuations in the ordered phase of MnSi using unpolarized neutrons have been performed by Ishikawa et al many years ago [9]. These experiments were not sensitive enough to distinguish between spin-flip (sf) and non-spin-flip (nsf) contributions. In order to separate the contributions, we have investigated the magnetic excitation spectrum using inelastic neutron scattering with longitudinal polarization analysis.

Due to the lack of a symmetry center in the cubic crystal structure of MnSi ($P\bar{2}13$), a left-handed magnetic spiral with a long period $\Lambda \approx 185 \, \text{Å}$ is observed leading to magnetic satellite peaks close to the nuclear reflections (figure 2) [10]. Weak crystal electric fields pin the spirals along the $\langle 111 \rangle$ directions. Under application of a field $B \approx 0.6 \, \text{T}$ a ferromagnetic state is induced. Close to $T_c$ and in a field $B \approx 0.2 \, \text{T}$, a skyrmion lattice develops that is stabilized by low energy fluctuations [4]. Apart from these modulated phases, the magnetic properties are considered to be those of a ferromagnet.

Figure 3 shows contour maps of the sf and nsf excitations of MnSi as measured in the field-induced ferromagnetic state ($B = 0.7 \, \text{T}$) at $T = 26 \, \text{K}$ (0.88$T_c$). The experiments were performed on the triple-axis spectrometer IN20 at the Institut Laue-Langevin (ILL) around the (110) Bragg peak in a (110) plane using longitudinal polarization analysis (figure 2).

The sf data in figure 3(a) clearly shows the spin wave branch emerging from the (110) Bragg peak becoming very steep near $\zeta = 0.8$. One can clearly distinguish two different regimes: (i) at low $E$ transfer $E < 2.5 \, \text{meV} \ (0.9 < \zeta < 1.0)$ a ferromagnetic spin wave dispersion is observed given by
Figure 3. Contour maps of MnSi measured along the [110] direction at $T = 26$ K and in a field $B = 0.7$ T. The sf data (a) clearly indicates the spin wave branch merging into the Stoner continuum. The contours of the nsf scattering (b) show the steep phonon branch as well as the longitudinal fluctuations, which extend to high energy transfers.

$E_q = Dq^2$ with $D = 23.5 \pm 3.0$ meV $\AA^2$ [11] and (ii) at large $E$ transfers the excitations can be directly identified in terms of Stoner excitations as depicted in figure 1(b). A close inspection of the data shows that only the spin waves renormalize with increasing $T$, while the Stoner excitations do not change significantly [12].

The nsf data (figure 3(b)) shows, at low energy transfers, large cross sections. They are identified as longitudinal magnetic fluctuations, which have also been observed in Ni [13] and EuS [14]. However, due to the strong electronic correlations in MnSi leading to a large magnetic correlation length, the longitudinal modes are already strong much further away from $T_c$, as the present measurements have been conducted. These modes diverge near $T_c$ due to the increasing magnon–magnon interactions [15]. The longitudinal modes extend into the regime of single-particle excitations. The distribution of their spectral weight looks qualitatively similar as the simple model shown in figure 1(c).

To obtain a more quantitative interpretation of the data we show in figure 4 cuts through the contours of figure 3 at $E = 5$ meV. The Stoner excitation in the sf channel is clearly visible near $(0.8, 0.8, 0)$. The solid line is a fit to equation (1), the magnetic nsf scattering is barely visible. The solid and the broken lines are fits to equation (1), assuming a diffusive or a propagating mode, respectively, and a cross section for the acoustic phonon at $\zeta = 0.95$.

function:

$$S(q, E) = \frac{q^2_a}{\kappa^2 + q^2} \frac{1}{2 \pi} \frac{E \Gamma}{(E - E_q)^2 + \Gamma^2} (n + 1)$$

Figure 4. Constant energy scans for $E = 5$ meV at $T = 26$ K and $B = 0.7$ T. (a) shows a Stoner excitation near $(0.8, 0.8, 0)$. The solid line is a fit to equation (1). (b) The magnetic nsf scattering is barely visible. The solid and the broken lines are fits to equation (1), assuming a diffusive or a propagating mode, respectively, and a cross section for the acoustic phonon at $\zeta = 0.95$.

We point out that the measurements shown here have been conducted at very large $|q|$ \( \gg 2\pi/\Lambda \), where the helical correlations are expected to be of no relevance. Indeed, the contour plot for $E = 0$ (figure 5) demonstrates that the magnetic satellites are very close to the nuclear zone center.
Driven by the discovery of a non-Fermi liquid state under high pressure and a skyrmion lattice for $B = 0.2$ T, the magnon spectrum in the helical phase has recently been investigated with high resolution triple-axis spectroscopy using cold neutrons. Around the magnetic satellites, a rich spectrum of spin excitations are observed that are identified as helimagnons [17]. Using a model [18] based on only three parameters, namely the pitch of the helix, the spin wave stiffness and an overall amplitude of the signal, one can account for all spectra, demonstrating that helimagnons are a universal characteristics of systems with weak chiral interactions.

In conclusion, we have shown that inelastic neutron scattering with polarized neutrons allows us to study the single-particle excitations in weak itinerant magnets. The results prove directly that the Stoner excitations are sf excitations. Unexpectedly, the nsf fluctuations show significant cross sections even at high energy transfers. To disentangle the complicated spectra and to determine the helicity of the helimagnons it would be of great value to extend the polarized beam measurements to small momentum transfers using high resolution spectroscopy. Indeed, recently it was shown that the paramagnetic [19] and helimagnetic [20] excitations in MnSi have a chiral contribution, which is large near $T_C$.

3. Magnon and phonon excitations in antiferromagnetic Cr

In metals with Fermi surfaces, nesting enhances the number of transitions at the nesting wavevectors $Q_n$ when compared to other wavevectors. The nesting greatly increases the number of possible electronic transitions at $Q_n$, which may lead to the formation of spin density waves (SDWs) [21, 22] and/or may soften and broaden phonons [23]. Both effects are observed in strongly correlated electron systems, i.e. in the copper oxide superconductors [24, 26] and in elemental Cr [25]. Both materials show a long history of intensive investigations.

In contrast to MnSi, where the magnetic ordering is caused by the non-centrosymmetric crystal structure leading to a pronounced Dzyaloshinskii–Moriya (DM) interaction [5, 6], the incommensurate magnetic ordering in Cr is the result of the nesting properties of the electron and hole Fermi surface [22] as shown in figure 6.

Cr undergoes a transition from the paramagnetic phase to an SDW phase at $T_N = 311$ K characterized by propagation vectors $Q^\pm = (1 \pm \delta, 0, 0)$ with $\delta = 0.048$. The corresponding magnetic satellite peaks are visible near the forbidden Bragg reflections of the bcc structure, i.e. $h+k+l = \text{odd}$ (figure 2). In the transverse spin density wave (TSDW) phase $T_{sd} < T < T_N$, the magnetic moments are aligned perpendicular to $Q^\pm$. At $T_{sd} = 121$ K the magnetic structure undergoes a first-order phase transition to a longitudinal spin density wave (LSDW) phase with the spins aligned along $Q^\pm$.

Due to the SDW, there is a distortion of the lattice with twice $\delta$. Indeed, charge density waves (CDW) were observed using both neutron and x-ray diffraction [30, 31]. Recently, even pressure measurements were performed [32]. The CDW can be induced either by Fermi surface nesting or by a strain wave induced by magneto-elastic coupling to the SDW.

As explained above, the interaction of the conduction electrons with the lattice vibrations enhanced by anomalies of the Fermi surface leads to anomalous phonon dispersions. Using inelastic neutron scattering, four regions have been identified where transverse acoustic phonons show anomalous behavior that can be traced back to nesting [33]. The two most pronounced anomalies occur near the N and H point as shown in figure 7. Because of the appearance of an SDW near H, this point is of particular interest. However, because of the coarse $Q$ resolution of neutron scattering the phonon anomaly may be washed out. Here, the improved $Q$ resolution of synchrotron radiation may help to highlight if there is a correspondence between the phonon anomalies and Fermi surface nesting in Cr near H.

3.1. Magnetic excitations in Cr

In order to obtain an overview on the spectral distribution of the magnetic excitations in Cr we show in figure 8 a contour plot of the inelastic intensity in the TSDW phase at $136$ K that was measured with high energy and momentum resolution using cold neutrons ($E_l = 5.64$ meV). Most dominant are the very steep excitations that emerge from the

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**Figure 5.** Elastic neutron scattering around the (110) Bragg peak showing the magnetic satellite peaks along the [111] directions. The weak spots at $(1, 1, 0.985)$ and $(1, 1, 1.015)$ are the result of the out-of-plane satellites, which become visible due to the coarse vertical resolution.

**Figure 6.** Schematic cross section of the Fermi surface in the (110) plane of Cr. Nesting vectors $Q$ connect the electron with the hole surface.
Figure 7. Comparison of the phonon dispersion as calculated by means of the Born–von Kármán model along the high symmetry directions [ζζ0] and [ζ00]. The data points from [33] show strong anomalies near the N and H points as indicated by arrows. Our results show that the T2 branch along the whole zone boundary line N–H softens as indicated by three arrows.

Figure 8. Contour map of the excitation spectrum as measured in the transverse spin density wave phase of Cr at $T = 136$ K. The intense peak near 4.5 meV and $Q = (1, 0, 0)$ is the Fincher mode. Note the additional peak near $(1.02, 0, 0)$ and 3 meV that has no counterpart at $(0.98, 0, 0)$. The ellipsoids indicate the resolution of the triple-axis spectrometer TASP [38] at various $E$ transfers.

The goal of the experiments using synchrotron radiation was to determine the precise $q$ position of the anomalous softening of the phonons and to compare it with the nesting features of the Fermi surface of Cr. In order to improve in $Q$ resolution over the previous experiments using inelastic neutron scattering [33], we applied inelastic x-ray scattering. The experiment was performed at beamline ID-28 at ESRF using a Cr single crystal with the dimensions $2 \times 2 \times 2$ mm$^3$. The phonon dispersion was investigated near the H point and the measurements extended along the zone boundary to the N point [39]. In addition to previous measurements, the phonon dispersion along the line connecting the H point and the N point (figure 7) was explored for the first time.

Figure 10 shows a phonon at $Q = (0.5, 3.5, 0)$ that belongs to the acoustic [110] T2 branch. Obviously, the

Other important features are the low energy excitations at $E < 10$ meV, which occur only in the TSDW phase [28, 29]. Clearly visible are the modes at $E \approx 4.5$ meV and at 8 meV (figure 9) as already observed by Fincher et al demonstrating the correspondence of our data with the previous data [34]. In addition, two modes at $\pm 1.016 \, \AA^{-1}/\pm 3.8$ meV and $\pm 0.984 \, \AA^{-1}/\pm 6.8$ meV are visible, which have no counterparts at the symmetry-related positions $\pm 0.984$ and $\pm 1.016 \, \AA^{-1}$. The comparison with previous data measured with lower resolution [27, 29] raises the question if dispersing modes alone can explain the data or if local modes have to be included.

Moreover, it is not easily understandable why the measured modes have very similar intensities although the thermal population factor $\langle n + 1 \rangle$ would predict a significantly higher intensity for the 3.8 meV mode when compared with the 6.8 meV mode, provided that both modes belong to the same dispersion as alluded to in [27]. We may speculate that mode coupling has to be involved. To answer these questions, more precise measurements with better statistics have to be performed.

3.2. Phonon softening in Cr

The goal of the experiments using synchrotron radiation was to determine the precise q position of the anomalous softening of the phonons and to compare it with the nesting features of the Fermi surface of Cr. In order to improve in Q resolution over the previous experiments using inelastic neutron scattering [33], we applied inelastic x-ray scattering. The experiment was performed at beamline ID-28 at ESRF using a Cr single crystal with the dimensions $2 \times 2 \times 2$ mm$^3$. The phonon dispersion was investigated near the H point and the measurements extended along the zone boundary to the N point [39]. In addition to previous measurements, the phonon dispersion along the line connecting the H point and the N point (figure 7) was explored for the first time.

Figure 10 shows a phonon at $Q = (0.5, 3.5, 0)$ that belongs to the acoustic [110] T2 branch. Obviously, the
The position of the phonons can be measured with high precision. The solid line is a fit assuming a Lorentzian. Following this result, the dispersion of the phonons was determined in detail for $Q$ along [100] as well as along the N–H line [39]. The measured dispersions near $(\frac{1}{2}1\frac{1}{2}0)$ and (100) are shown in figure 11 (circles) and compared with the theoretical prediction based on a simple Born–von Kármán model (triangles). Along the [110] direction, the maximum softening is observed exactly at the zone boundary N. In contrast, the H phonon softens at the incommensurate position $\delta = 0.05$ where the SDW satellites occur and not at the zone boundary (100) as reported previously (figure 7) [33].

Similar measurements have been conducted at various $Q$ positions across the zone boundary line N–H. Figure 12 summarizes the softening of the measured phonons when compared with a Born–von Kármán model along the [100] direction (circles) and along the zone boundary N–P (triangles). In the [100] direction, the phonon softening has a distinct minimum at the nesting wavevector $Q^\parallel = (0.95, 0, 0)$. Surprisingly, a strong anomaly also appears along the entire zone boundary line N–P indicating that strong electron–phonon coupling limited to a small range of wavevectors can also result in strong phonon anomalies without invoking nesting. This observation implies that the phonon anomalies in copper oxide superconductors may also be explained by an enhanced electron–phonon coupling without invoking novel collective modes or some hidden nesting of the Fermi surface [39].

Additional measurements in the paramagnetic phase of Cr show a similar softening of the phonons close to the zone boundaries discussed above. In particular, the strong softening at $Q^\parallel = (0.95, 0, 0)$ persists, demonstrating that the magnetoelastic coupling is small. Therefore, the nesting of the Fermi surface is responsible for the Kohn anomaly in the phonon spectrum and the SDW below $T_N = 311$ K at $Q^\parallel$. They evolve independently with temperature.

In conclusion, we have shown that the incommensurate magnetic excitations in Cr can be interpreted in terms of electron–hole excitations at the Fermi surface. Strong electron–phonon coupling without invoking nesting leads to a pronounced softening of the T2 phonon branch along N–H, i.e. away from $Q^\parallel$. We have provided evidence that the low energy excitations in the TSDW phase may not be explained in terms of a mode having a dispersion as proposed in [27].
4. Small samples—extreme conditions

Studying quantum phase transitions by applying pressure, magnetic fields or by doping has proven to be a successful route to identify materials with novel properties. In MnSi, the application of pressure leads to the suppression of helical order near $p_c = 1.46$ GPa accompanied by the appearance of a partially ordered magnetic state and a non-Fermi liquid phase [40]. In Cr, antiferromagnetic order is suppressed around $p = 10$ GPa [32]. Neutron scattering as well as x-ray synchrotron scattering have provided valuable information about the vanishing of the order parameters: however, the present-day sensitivity of neutron spectrometers is not sufficient to also characterize the spectrum of the magnetic and lattice excitations close to the quantum phase transitions because the samples are usually very small [41].

Measurements of phonon dispersions may be conducted using inelastic x-ray scattering because only small samples are necessary and the $Q$ and $E$ resolutions are often sufficient as shown in section 3.2. Inelastic magnetic scattering by x-rays is, so far, impossible because the magnetic cross sections are very small. In the following we demonstrate the feasibility of inelastic neutron scattering on small samples using supermirror focusing guides [42], to generate very small but intense neutron beams at the sample position of the thermal triple-axis instrument PUMA at FRM II.

To perform the experiment, a focusing guide with a length of 500 mm as described in [43] has been installed between the monochromator and the sample position. In the first instance we used a neutron CCD camera to both correctly align the guide and to analyze the shape and intensity of the beam at the sample position. These measurements show that the beam has an FWHM of approximately 2 mm in the horizontal direction and 8 mm in the vertical direction at the sample position. Such a beam size is ideal for studying samples with millimeter dimensions, unlike the conventional PUMA profile where the primary beam has dimensions of roughly 25 mm in the horizontal direction and 28 mm in the vertical direction and any adjustment for sample size is done by adjustable, neutron absorbing slits. The CCD camera images also revealed that using the guide results in a very low background.

We have performed successful test experiments on two different samples. Firstly, a small single crystal of Cr with dimensions $2 \times 2 \times 2$ mm$^3$ was investigated. Samples of this size can typically be used in a Paris–Edinburgh high pressure cell. Using the focusing guide, we were able to observe, amongst other things, the change in intensity of the magnetic excitations in the TSDW phase with increasing temperature (figure 13). Note that the counting time per point of 10 min corresponds to the typical counting time (13 min) used to collect the data in figure 3 of [34]. Clearly, because of the coarse $Q$ resolution due to the focusing, the fine structure of the spectrum is wiped out. Still, for the determination of the energy scale of the excitations versus pressure, inelastic measurements are feasible.

In a second experiment we measured phonons in two different samples of quartz ($\text{SiO}_2$). The small and the large samples have a volume of 8 mm$^3$ and 2000 mm$^3$, respectively (figure 14). The comparison allows a direct calibration between the conventional PUMA configuration and the configuration with a focusing guide. The results show that it is indeed possible to measure the acoustic transverse phonon at ($-0.9, 2.0, 0$) with high precision within a reasonable time. Actually the comparison of the two measurements demonstrates that the results with the focusing guide provide a much cleaner spectrum and a lower background. Of course, the performance can be further increased with a significant gain in
intensity if a second focusing guide is used between the sample and the analyzer [41], and if the critical angle of reflection of the focusing guide is increased from $m = 3$ to 7 [44].

In conclusion, we have shown that advanced neutron optics allows the investigation of magnons and phonons under extreme conditions using small crystals providing a good $E$ and $Q$ resolution. Focusing techniques for inelastic neutron scattering facilitate the exploration of new areas of science, where traditional experiments have been restricted due to the requirement of large samples.

5. Conclusions

Experiments using polarized neutrons allow the separation of phonon modes from magnon modes and furthermore between transverse and longitudinal fluctuations in magnetic systems. In MnSi it is observed that the Stoner excitations can be clearly identified in terms of spin-flip excitations of the conduction electrons. The non-spin-flip scattering provides direct information on the amplitude fluctuations in the single-particle regime. In addition, we found evidence that the low energy excitations in Cr (Fincher–Burke modes) may not be explained solely by assuming modes with a dispersion. Surprisingly the modes are asymmetric with respect to the (100) position [29]. The combination of inelastic x-ray and neutron scattering indicates that the Kohan anomaly near the H point occurs at the same position as the spin density wave, demonstrating that both effects are caused independently by the nesting properties of the Fermi surface of Cr. As a new feature, the x-ray results imply that the phonons are softened along the whole zone boundary line N–H due to electron–phonon interactions.

We have shown that the inelastic scattering of neutrons and x-rays provide complementary information on the lattice dynamics and the spectrum of the magnetic excitations in strongly correlated materials. While neutron scattering provides high energy resolution and moderate $Q$ resolution, x-rays provide sufficient $Q$ resolution, thus allowing us to map out the softening of phonons near the zone boundaries of transition metals like Cr.

Finally we have evaluated the possibilities of performing inelastic neutron scattering from small samples using advanced focusing techniques. By means of focusing guides the intensity of neutron beams at the sample position can be increased rather dramatically, thus allowing the investigation of samples with a volume of a few mm$^3$. Therefore, samples under extreme conditions can be investigated near quantum phase transitions. With the advancement of next-generation neutron sources using halo isomers that provide polarized neutron beams of very high brilliance [45], neutron scattering may make a big leap towards the investigation of excitations in samples that are much smaller than 1 mm$^3$.

Acknowledgments

We thank M Janoschek, D Lamago, R Mole, S Müllerbauer, P Niklowitz, C Pfleiderer, D Reznik and C Schanz for very useful discussions and assistance during the course of the experiments. This work is based on experiments performed at the FRM II in Garching, the Swiss Spallation Source SINQ, the HFR at the Institut Laue-Langevin and the ESRF in Grenoble. We gratefully acknowledge financial support from the research unit on Quantum Phase Transitions FOR960 of the German Science Foundation (DFG). Part of the work was supported by the Swiss National Science Foundation through MaNEP and by project 226507–NM3 within the seventh framework program FP7 of the EU.

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