FAST TRACK COMMUNICATION

Magnetic excitations of Fe$_{1+y}$Se$_x$Te$_{1-x}$ in magnetic and superconductive phases

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Abstract

We have used inelastic neutron scattering and muon-spin rotation to compare the low energy magnetic excitations in single crystals of superconducting Fe$_{1+y}$Se$_0.5$Te$_0.5$ and non-superconducting Fe$_{1.10}$Se$_0.25$Te$_0.75$. We confirm the existence of a spin resonance in the superconducting phase of Fe$_{1+y}$Se$_0.5$Te$_0.5$, at an energy of 7 meV and a wavevector of $(1/2, 1/2, 0)$. The non-superconducting sample exhibits two incommensurate magnetic excitations at $(1/2, 1/2, 0) \pm (0.18, -0.18, 0)$ which rise steeply in energy, but no resonance is observed at low energies. A strongly dispersive low energy magnetic excitation is also observed in Fe$_{1+y}$Se$_0.25$Te$_0.75$ close to the commensurate antiferromagnetic ordering wavevector $(1/2 - \delta, 0, 1/2)$, where $\delta \approx 0.03$. The magnetic correlations in both samples are found to be quasi-two-dimensional in character and persist well above the magnetic (Fe$_{1.10}$Se$_0.25$Te$_0.75$) and superconducting (Fe$_{1+y}$Se$_0.5$Te$_0.5$) transition temperatures.

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

1. Introduction

Considerable effort has been devoted over the past two years to investigating the basic properties of the Fe-based family of superconductors [1–3]. A central question is whether magnetism plays an important role in the formation of the superconducting state. A useful strategy for tackling this problem is combining neutron scattering and muon-spin rotation (\(\mu\)SR) measurements on one and the same sample. Neutron scattering provides information on magnetic correlations and on the nature of the magnetic excitations, while \(\mu\)SR can determine whether static magnetic order and/or bulk superconductivity exists.

The Fe$_{1+y}$Se$_x$Te$_{1-x}$ system is a convenient one to study with this methodology as large high quality crystals can be grown [4, 5] and the tetragonal crystallographic structure is relatively simple to analyse and model. Single crystals are easier to grow if there is a small excess of Fe (i.e. $y > 0$), especially for small $x$ [6–9].

The pure FeSe compound is a superconductor with a transition temperature $T_c \sim 8$ K [6]. The $T_c$ can be increased by partial substitution of Te for Se such that $T_c \sim 14$ K for $0.4 \lesssim x \lesssim 0.8$ and $y \approx 0$ [10, 11]. The application of pressure has also been found to raise $T_c$, with values as high as 37 K observed for FeSe [12–15]. Compounds with $x \lesssim 0.4$ do not exhibit bulk superconductivity but order magnetically below a temperature which has a maximum of 67 K at $x = 0$ and which decreases with $x$ and vanishes at $x \sim 0.4$. We recently found evidence for coexistence of incommensurate magnetic order and partial superconductivity for $x \sim 0.25$ [11].
In this study, we present neutron scattering and μSR measurements on two single-crystal samples: (i) Fe$_{1.0}$Se$_{0.5}$Te$_{0.5}$, a bulk superconductor, and (ii) Fe$_{1.0}$Se$_{0.25}$Te$_{0.75}$, a non-superconducting sample which exhibits magnetic order. For superconducting Fe$_{1.0}$Se$_{0.5}$Te$_{0.5}$ we observe a resonant magnetic excitation consistent with that reported previously for compounds with similar compositions [16–19]. For Fe$_{1.0}$Se$_{0.25}$Te$_{0.75}$ we observe strongly dispersive, low energy magnetic excitations associated with the magnetic ordering wavevector (1/2 − δ, 0, 1/2), δ ≈ 0.03, and also with the wavevectors (1/2, 1/2, 0) ± (0.18, −0.18, 0). We find no evidence for a resonance peak in the excitation spectrum of Fe$_{1.0}$Se$_{0.25}$Te$_{0.75}$.

2. Experimental details

Single crystals of Fe$_{1+x}$Se$_x$Te$_{1−x}$ were grown by a modified Bridgman method as reported by [4]. Neutron scattering measurements were carried out on the triple-axis spectrometer TASP [20] at the SINQ [21] spallation source (Paul Scherrer Institut, Switzerland). Bragg reflections from a pyrolytic graphite PG(002) monochromator and analyser were used at a fixed final wavevector of 2.66 Å$^{-1}$. A PG filter was placed after the sample to reduce contamination from higher order harmonics in the beam and the instrument was set up in the open–open–open–open collimation with the analyser focusing in the horizontal plane. The crystals were single rods with masses of approximately 4 g. The Fe$_{1.0}$Se$_{0.25}$Te$_{0.75}$ sample was orientated in two settings to give access to (h, 0, l) and (h, k, 0) planes in reciprocal space. Measurements on Fe$_{1.0}$Se$_{0.5}$Te$_{0.5}$ were made in the (h, k, 0) plane only. In this report we index the reciprocal lattice with respect to the tetragonal unit cell described by the P4/nnm space group with unit cell parameters $a ≈ 3.8$ Å and $c ≈ 6.1$ Å along lines joining the nearest neighbour Fe atoms.

Zero-field-cooled magnetization measurements were performed on a Quantum Design MPMS magnetometer with a measuring field μ0H = 0.3 mT using the direct current method. To reduce the effects of demagnetization, thin plate-like pieces of Fe$_{1+y}$Se$_x$Te$_{1−x}$, cleaved from the main single crystals, were oriented with the flat surface (ab plane) parallel to the applied field.

Zero-field (ZF) and transverse-field (TF) muon-spin polarization (μSR) experiments were performed on the πM3 beam line at SμS (Paul Scherrer Institut, Switzerland). In TF experiments a magnetic field of 11.8 mT was applied parallel to the crystallographic ab plane of the crystal and perpendicular to the muon-spin polarization.

3. Results

3.1. Magnetization and μSR measurements

Zero-field-cooled magnetization data normalized to the ideal 1/4π value ($M_{\text{norm}}$) are shown in figure 1(a). The
Fe$_{1.10}$Se$_{0.50}$Te$_{0.50}$ sample is seen to be a bulk superconductor with the onset of the transition $T_{c}^{\text{onset}} \simeq 14.0$ K and $M_{\text{norm}} \simeq 0.8$ at $T \simeq 2$ K. The Fe$_{1.10}$Se$_{0.25}$Te$_{0.75}$ sample also exhibits superconductivity ($T_{c}^{\text{onset}} \simeq 8.6$ K) but has a small superconducting fraction of order 10% at low temperature.

For the $x = 0.5$ sample, the ZF time spectra measured at $T = 1.7$ and 20 K are almost identical, thus suggesting that the magnetic state of this sample is the same above and below the superconducting transition temperature. The solid lines correspond to a fit with the function $A_{ZF}(t) = A_{0}^{ZF}e^{-\Delta t^{2}/\Lambda^{2}}$, where $A_{0}^{ZF}$ is the initial asymmetry and $\Delta^{ZF}$ is the exponential relaxation rate. Such behaviour is consistent with dilute Fe moments as observed recently for FeSe$_{1−x}$ [22].

The TF data for $x = 0.5$ fit well to the function $A_{TF}(t) = A_{0}^{TF}e^{-(\Lambda t^{2}+\sigma^{2})}cos(\gamma_{\mu}Bt+\phi)$. Here, $\gamma_{\mu}/2\pi = 135.5$ MHz $T^{-1}$ is the muon gyromagnetic ratio, $\phi$ is the initial phase of the muon-spin ensemble, and $\sigma$ is the Gaussian relaxation rate. Figure 1(b) shows that the TF asymmetry $A_{TF}^{\phi}$ is almost temperature independent. The slightly stronger relaxation of the muon-spin polarization at 1.7 K relative to that at 20 K is due to the formation of the vortex lattice at $T < T_{c}$.

Static (within the $\mu$SR time window) magnetism develops in Fe$_{1.10}$Se$_{0.25}$Te$_{0.75}$ as signalled by a fast drop of both $A_{ZF}$ and $A_{TF}$ within the first 100 ns (see the upper panel of figure 1(b)). The solid lines correspond to fits with $A_{ZF}(t) = A_{0}^{ZF}e^{-\Delta t^{2}/\Lambda^{2}}+A_{0}^{TF}e^{-(\Lambda t^{2}+\sigma^{2})}$ and $A_{TF}(t) = e^{-\gamma_{\mu}Bt+\phi}[A_{1}^{TF}e^{-(\Delta t^{2}/\Lambda^{2})}cos(\gamma_{\mu}Bt+\phi)+A_{2}^{TF}e^{-(\Delta t^{2}/\Lambda^{2})}cos(\gamma_{\mu}Bt+\phi)]$. Here, $A_{1}^{TF}(TF)$ and $A_{2}^{TF}(TF)$ are the initial ZF (TF) asymmetry and the exponential depolarization rate of the slow (fast) relaxing component, respectively. The temperature evolution of $A_{1}^{TF}$, shown in figure 1(b), reveals that below 20 K magnetism occupies more than 95% of the whole sample volume. The corresponding values of the onset and the mid-point of the magnetic transitions, determined as shown in the figure, are $T_{c}^{\text{onset}} \simeq 33.7$ K and $T_{c}^{\text{mid}} \simeq 27.6$ K. We note that although the magnetic order is shown to extend throughout virtually the entire volume of the sample, $\mu$SR cannot be used to determine whether the magnetic order is long range. The neutron diffraction data presented in the next section show that the magnetic order is in fact relatively short range.

### 3.2. Neutron scattering results

Elastic neutron scattering measurements on Fe$_{1.10}$Se$_{0.25}$Te$_{0.75}$ in the $(h, l)$ scattering plane at 2 K, as shown in figure 2(a), reveal a diffuse magnetic peak centred on $(1/2, \delta, 0, 1/2)$ with $\delta \approx 0.03$. The incommensurate peak is much broader than the resolution of the instrument. From $Q$ scans through the peak we obtain correlation lengths along the $a$ and $c$ axes of 11.4(6) $\AA$ and 7.5(4) $\AA$ respectively at 2 K. Figure 2(b) shows that the magnetic peak develops below $T_{N} \sim 50$ K. The correlation lengths did not change measurably upon warming through the $T_{N}$ (figure 2(b): inset). The magnetic propagation vector $q = (1/2 − \delta, 0, 1/2)$ is similar to that found previously for the similar composition Fe$_{1.10}$Se$_{0.25}$Te$_{0.75}$ compound. For the latter compound we confirmed that the peak described by $q$ is magnetic in character using neutron polarization analysis [11]. Our results are consistent with measurements on Fe$_{1.09}$Se$_{0.25}$Te$_{0.75}$ for which the incommensurability is found to be $\delta \approx 0.04$ [23].

The magnetic scattering cross-section is directly proportional to the magnetic response function $S(Q, E)$—the Fourier transform of the space- and time-dependent spin–spin correlation function. According to the fluctuation-dissipation theorem, $S(Q, E)$ is in turn related to the imaginary part of the dynamical susceptibility $\chi''(Q, E)$ by [24]

$$S(Q, E) = \frac{1}{\pi}[n(E, T) + 1]|\chi''(Q, E)|.$$  \hspace{1cm} (1)

The Bose–Einstein population factor $n(E, T) = \exp(E/k_{B}T) − 1)^{-1}$ (where $k_{B}$ is the Boltzmann constant) takes into account the increase in scattering from bosonic excitations due to the thermal population at temperatures $T > 0$. Correction for this factor allows the temperature dependence of $\chi''(Q, E)$ to be studied.
Figure 3. Inelastic neutron scattering from Fe$_{1.0}$Se$_{0.25}$Te$_{0.75}$ in the vicinity of the magnetic ordering wavevector $q = (0.47, 0, 0.5)$.

(a) Constant energy scans collected at 2, 4 and 6 meV and 2 K along $(h, 0, 0.6)$. The data have been shifted in $\chi''(Q, E)$ by arbitrary amounts for clarity. (b) Constant energy scans collected at 2 meV and the temperature of 2 K showing $\chi''(Q, E)$ along $(h, 0, 0.5)$ and $(h, 0, 0.7)$ and $(h, 0, 0.9)$. The plots have been displaced and the dashed lines show Gaussian peaks through the spectra. (c) Constant energy scans at 2 meV at temperatures of 2, 40, 150 and 300 K showing $\chi''(Q, E)$ along $(0.5, 0, l)$. Note that a linear background has been subtracted in all scans. (d) Diagram of the $(h, 0, l)$ plane to show scan directions denoted by roman numerals.

Figure 3(a) shows background corrected scans along the $(h, 0, 0.6)$ direction at energy transfers of 2, 4 and 6 meV for the Fe$_{1.10}$Se$_{0.25}$Te$_{0.75}$ crystal. A peak at $Q = q$ is present in each scan, indicating a strongly dispersing excitation. The broadening of the dispersion in $Q$ may be due to unresolvable splitting of the mode into two excitations at higher energies. The measured magnetic response at 2 meV parallel to $(1, 0, 0)$ for $l = 0.5, 0.7$ and 0.9, as shown in figure 3(b), reveals considerable broadening of $\chi''(Q, E)$ in the out-of-plane direction. Such broadening is characteristic of a quasi-two-dimensional system with weak interactions along $c$. Figure 3(c) shows that spin fluctuations persist up to at least 150 K, well into the paramagnetic state. At 40 K, i.e. close to the magnetic ordering temperature, $\chi''(Q, E)$ is almost the same as at 2 K.

We now turn to the low energy excitation spectrum in the vicinity of the wavevector $(1/2, 1/2, 0)$. Figures 4(a) and (b) show maps of $\chi''(Q, E)$ measured along $(h, 1 - h, 0)$ for Fe$_{1.10}$Se$_{0.25}$Te$_{0.75}$ at 2 and 40 K. The fluctuations measured at 2 K are consistent with the magnetic excitation spectrum at higher energies reported for Fe$_{1.03}$Se$_{0.27}$Te$_{0.73}$ [25]. The excitation spectrum at 2 K is characterized by steep incommensurate branches arising from $(1/2 \pm \epsilon, 1/2 \mp \epsilon, 0)$ where $\epsilon \approx 0.18$. The incommensurate excitations are still present at 40 K. The scans shown in figure 4(c) reveal that at $E = 7$ meV, the system response is nearly the same at 2 K as at 40 K. The background corrected $\chi''(Q, E)$ for the Fe$_{1.10}$Se$_{0.25}$Te$_{0.75}$ sample does not appear to change for energies in the 2–7 meV range measured at these temperatures. This is also the case for measurements along $(1/2, 0, l)$ in figure 3(c) that show $\chi''(Q, E)$ data at 2 meV to be similar at 2 and 40 K.

The results obtained for Fe$_{1.01}$Se$_{0.50}$Te$_{0.50}$ are in stark contrast to those for the non-superconducting Fe$_{1.10}$Se$_{0.25}$Te$_{0.75}$ sample just described. Figures 4(d) and (e) show maps of the magnetic spectrum as a function of wavevector along $(h, 1 - h, 0)$ for energies between 2 and 7 meV at 2 and 40 K. At 2 K we find a strong signal in $\chi''(Q, E)$ centred on $Q = (1/2, 1/2, 0)$ and $E \sim 7$ meV. This feature corresponds to the spin resonance reported previously in superconducting FeSe$_{0.4}$Te$_{0.6}$ [16], FeSe$_{0.46}$Te$_{0.54}$ [18] and FeSe$_{1.5}$Te$_{0.5}$ [19]. At higher energies, the excitations have been found to disperse away from $(1/2, 1/2, 0)$ along $(1, -1, 0)$ [18]. However, it is the low energy response of the system which shows the most dramatic change on transition into the superconducting state, as may be seen in figure 4(f). As the sample is cooled from 40 to 2 K, the integrated dynamical susceptibility of the peak at 7 meV increases by more than a factor of two and decreases in width along $(1, -1, 0)$ by $\sim 30\%$. Fluctuations continue to be observed well above $T_c$.

4. Discussion

In combination with earlier measurements, the results presented here establish that the low energy magnetic dynamics of Fe$_{1+y}$Se$_y$Te$_{1-y}$ vary strongly with $x$. The magnetic spectra of the magnetically ordered compound ($x = 0.25$) and the bulk superconductor ($x = 0.5$) both contain low energy magnetic fluctuations in the vicinity of the antiferromagnetic wavevector $(1/2, 1/2, 0)$. However, at $x = 0.25$ the
fluctuations are incommensurate with wavevector \((1/2 \pm \epsilon, 1/2 \mp \epsilon, 0)\), \(\epsilon \approx 0.18\), whereas at \(x = 0.5\) the strongest magnetic signal is commensurate. Moreover, at \(x = 0.5\) the magnetic spectrum has a gap of \(\sim 6\) meV and the size of the signal just above the gap increases strongly at low temperatures. This behaviour is consistent with the superconductivity-induced spin resonance reported recently in bulk superconducting samples of Fe\(_{1+x}\)Se\(_x\)Te\(_{1-x}\) of similar composition to ours \([16–19]\), and also in related Fe pnictide superconductors \([26–30]\).

A further difference is that the \(x = 0.25\) sample exhibits short-range, static (within the \(\mu\)SR time window) magnetic order with a characteristic wavevector \(q = (1/2 - \delta, 0, \pm 1/2)\), \(\delta \approx 0.03\), whereas according to our \(\mu\)SR data there is no static magnetic order in the bulk superconductor. The magnetic ordering wavevector \(q\) found at \(x = 0.25\) is the same as that in the parent phase Fe\(_{1+y}\)Te. The slight incommensurability is thought to be caused by the small excess of Fe accommodated in interstitial sites in the crystal structure \([8, 31, 32]\), although it is interesting that the incommensurability is the same to within experimental error at \(y = 0.10\) (the present sample) and at \(y = 0.03\) (the sample studied by us previously \([11]\)).

Our results suggest that there are two distinct magnetic ordering tendencies at \(x = 0.25\), one with wavevector \((1/2 - \delta, 0, \pm 1/2)\) and the other with wavevector \((1/2 \pm \epsilon, 1/2 \mp \epsilon, 0)\). The \(\mu\)SR data indicate that the volume fraction of magnetically ordered phase is close to 100%, but we cannot say whether the two characteristic magnetic correlations coexist on an atomic scale or whether the sample is magnetically inhomogeneous.

Finally, we comment on the fact that for the \(x = 0.25\) sample diffuse peaks are observed in the elastic (within energy resolution) channel below \(T \approx 50\) K by means of neutron scattering but static magnetic order is only detected below \(T \approx 35\) K by \(\mu\)SR. These observations can be reconciled by means of the difference in fluctuation rates observable by using muons (\(\sim \text{GHz}\)) and neutrons (\(\sim \text{THz}\)) below which spin freezing is measured. We infer from this that the characteristic fluctuations of the spin system lie between \(\sim \text{GHz}\) and \(\sim \text{THz}\) for \(35\) K \(\lesssim T \lesssim 50\) K. Such a gradual slowing down of the fluctuations could be a consequence of the quasi-two-dimensional nature of the spin system, which is also indicated by the persistence of spin correlations to temperatures well above the ordered phase. It is also interesting that the size of the magnetically ordered domains does not significantly increase with decreasing temperature, which suggests that the short-range order is never truly static but fluctuates down to the lowest temperature investigated. This picture is consistent with the recent observation of spin-glass behaviour in Fe\(_{1+x}\)Se\(_x\)Te\(_{1-x}\) for 0.05 < \(x < 0.55\) \([33]\).

5. Conclusion

We have observed a resonance-like peak at the antiferromagnetic wavevector \((1/2, 1/2, 0)\) in the low energy magnetic spectrum of Fe\(_{1.01}\)Se\(_{0.50}\)Te\(_{0.50}\), and shown that this feature is absent from the magnetic spectrum of Fe\(_{1.10}\)Se\(_{0.25}\)Te\(_{0.75}\) which instead shows incommensurate peaks flanking \((1/2, 1/2, 0)\). Our results reveal a clear distinction between the magnetic excitation spectra of Fe\(_{1+y}\)Se\(_x\)Te\(_{1-x}\) samples which are magnetically ordered and those which are bulk superconductors. We conclude that the existence of a resonance peak at the commensurate antiferromagnetic wavevector is a characteristic of bulk superconductivity in Fe\(_{1+y}\)Se\(_x\)Te\(_{1-x}\).

Figure 4. Variation of \(\chi' (Q, E)\) in the \((h, 1 - h, 0)\) direction for energies between 2 and 7 meV at temperatures of 2 and 40 K. Data in (a)–(c) are from Fe\(_{1.01}\)Se\(_{0.25}\)Te\(_{0.75}\) and data in (d)–(f) are from Fe\(_{1.01}\)Se\(_{0.50}\)Te\(_{0.50}\). Constant energy cuts at 7 meV along \((h, 1 - h, 0)\), measured at 2 and 40 K for Fe\(_{1.01}\)Se\(_{0.25}\)Te\(_{0.75}\) and Fe\(_{1.01}\)Se\(_{0.50}\)Te\(_{0.50}\) are shown in (c) and (f), respectively. A flat background has been subtracted in all scans and dashed lines through the data are fits with a Gaussian lineshape.
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