Incommensurate itinerant antiferromagnetic excitations and spin resonance in the FeTe$_{0.6}$Se$_{0.4}$ superconductor

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We report on inelastic neutron scattering measurements that find incommensurate itinerant like magnetic excitations in the normal state of superconducting FeTe$_{0.6}$Se$_{0.4}$ ($T_c=14$K) at wave-vector $Q_{inc} = (1/2 \pm \epsilon, 1/2 \mp \epsilon)$ with $\epsilon=0.09(1)$. In the superconducting state only the lower energy part of the spectrum shows significant changes by the formation of a gap and a magnetic resonance that follows the dispersion of the normal state excitations. We use a four band model to describe the Fermi surface topology of iron-based superconductors with the extended $s(\pm)$ symmetry and find that it qualitatively captures the salient features of these data.

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Magnetism is a key ingredient in the formation of Cooper pairs in unconventional high temperature superconductors. A consequence of magnetism and the symmetry of the superconducting order parameter is a magnetic resonance that has been detected through inelastic neutron scattering for a wide range of magnetic superconductors ranging from the heavy fermion systems, to cuprates, and more recently in the iron based superconductors. In each class of materials the features of the resonance in $S(Q, \omega)$ are different and are directly related to the electronic degrees of freedom, providing vital clues to the role of magnetic fluctuations in each case. We report inelastic neutron scattering measurements showing that the magnetic excitations in FeTe$_{0.6}$Se$_{0.4}$ are incommensurate and itinerant-like, and that upon entering the superconducting state only the lower energy part of the spectrum shows significant changes by the formation of a gap and a magnetic resonance that follows the dispersion of the normal state excitations. Using a four band model that describes the Fermi surface topology of iron-based superconductors and the extended $s(\pm)$ symmetry, we can qualitatively describe the salient features of the data. The good agreement between theory and experiment found here may provide clues to a better understanding of the magnetic resonance in high-$T_c$ cuprate superconductors.

In cuprate superconductors a magnetic resonance, whose energy scales with the superconducting energy gap, is a saddle point of a broader spectrum of magnetic excitations. The interpretation of the magnetic resonance and its relationship to superconductivity is complicated in the cuprates due to the occurrence of charge stripes and the pseudogap phase that lead to hotly debated models such as quantum excitations from charge stripes, or spin excitons from a particle-hole bound state of a $d$-wave superconductor.

A far clearer picture has emerged in the iron based superconductors partly due to their more itinerant nature. It was realized early on that the nesting between hole and electron Fermi surfaces related by the antiferromagnetic wavevector $Q_{AF} = (\pi, \pi)$ in the undoped iron superconductors, might also be responsible for unconventional superconductivity of the so-called extended $s(\pm)$-wave symmetry. The remarkable feature of this superconducting order parameter is the different sign of the superconducting gap on bands that are separated by $Q_{AF}$ can yield a magnetic resonance in the form of a spin exciton at the same wavevector.

This picture suggests that the continuum of magnetic excitations will be gapped in the superconducting state up to twice the energy of the superconducting gap, $2\Delta$ and that an additional feature, the magnetic resonance will occur at an energy $\hbar \omega_{res} < 2\Delta$. The latter is a consequence of both the unconventional symmetry of the superconducting order parameter which enhances the continuum at $2\Delta$, and the residual interaction between the quasiparticles that shifts the pole in the total susceptibility to lower energies. While so far the magnetic resonance has been observed in iron based superconductors...
FIG. 1: Typical INS transverse scans through crystal structure (tetragonal P4/ nmm unit cell) compared to other iron based superconductors make this system attractive for further examination. In this work we describe the measurements in momentum space in units of $Q=(\pi/a,\pi/a)$ while we omit reference to the $c$-axis direction for clarity as $Q_c=0$ throughout. The portion of reciprocal space probed is shown in Fig. 1(e), which was not explored in a previous study.

FeTe$_{0.6}$Se$_{0.4}$ crystallize in a primitive tetragonal structure P4/nmm, with $a=3.80$A and $c=6.02$A. High quality single crystal samples of this composition were prepared as described previously. Inelastic neutron scattering measurements were collected using the triple axis spectrometer IN8 operated by the Institut Laue-Langevin (ILL), with the sample mounted with the $c$-axis vertical giving us access to spin excitations within the basal $ab$-plane. The neutron optics were set to focus on a virtual source of 30 mm. Measurements were made with the (002) reflection of a pyrolytic graphite monochromator and analyzer with an open/open/open/open configuration. A graphite filter was placed after the sample position. Diaphragms were placed before and after the sample. Cooling of the sample was achieved by a standard orange cryostat. Data were collected around the $(1/2,1/2,0)$ and $(3/2,3/2,0)$ Brillouin zones using a fixed final wave vector $k_f$ of either 2.66 or 4.1 Å$^{-1}$.

In the normal state we find steeply dispersive magnetic excitations that we have measured up to 80 meV, Fig. 1(a,b). At higher energies, two excitations are clearly resolved in the transverse scans. At lower energies (2-6 meV), the two peaks merge to a broad response, far wider than the resolution function ($\Delta Q \sim 0.06$ r.l.u. measured on the (200) Bragg peak). These lower energy data at 20K are best modeled with two excitations as shown for example for the 6 meV data in Fig. 1(a). In Fig. 1(a) we show the lower portion of these transverse scans in the normal state as a color map where in addition we plot as black points the $Q$-position of the excitations determined by fitting Gaussian peaks to the data of Fig. 1. The dispersion of these excitations is linear up to 80 meV as shown in Fig. 1(a), and the points for the lower energy excitations fall on the same line confirming our above analysis. Extrapolating the centers of these excitations to zero energy, red lines in Fig. 1(a), it is clear that they do not converge to the commensurate $Q=(1/2,1/2)$ wavevector but to the incommensurate positions of $Q_{inc} = (1/2 \pm \epsilon, 1/2 \mp \epsilon)$ with $\epsilon=0.09(1)$. We note that for the FeTe end-compound with lower excess Fe, static antiferromagnetism appears at $Q=(1/2,0)$.

Longitudinal scans (not shown) between 2 and 80 meV through these spin excitations show that they are well defined isolated maxima and not parts of spin-wave cones as found in insulators, a conclusion concurred also in other recent INS measurement. Therefore, we argue that magnetic excitations in the normal state of the superconductor FeTe$_{0.6}$Se$_{0.4}$ consist of a single-lode excitation continuum from each incommensurate $Q_{inc}$. This behavior in $S(Q,\omega)$ is reminiscent of Fincher-Burke excitations reported for many itinerant magnetic materials that are...
close to an antiferromagnetic instability and indicates that an itinerant, rather than a local moment picture is relevant for this superconductor.

Cooling below $T_c$, we find substantial changes to the lower energy magnetic excitations, while our measurements indicate that the higher energy excitations remain largely unchanged when compared to the normal state measurements, see Fig. 2(b,d)\textsuperscript{12} The most pronounced changes are in the lower energy part of the magnetic excitation spectrum where the opening of a spin gap and the development of the magnetic resonance is observed as shown in Fig. 2(b). Here a spin gap opens below $\hbar\omega\approx 4$ meV, while there is an enhancement of the spectral weight above the spin gap that peaks at $\hbar\omega_{res}=6$ meV. At higher energies the spectral weight returns to approximately the same values as the data found in the normal state, see Fig. 1

Although the normal state magnetic excitations extend from incommensurate points it is important to establish if the magnetic resonance itself is centered on the commensurate wavevector $Q=(1/2,1/2)$ or if it consists of two excitations originating from the two incommensurate wavevectors (at $Q_{inc}$) found in the normal state. For a more detailed analysis of the Q-scans across the resonance we subtract the 20K from the 2K data in order to obtain only the magnetic scattering contributing to the resonance (see below). These data are shown in Fig. 3(b), while the centers of the resonant excitations are plotted in Fig. 2(b). Between $\hbar\omega=7$ and 12 meV, the data shows well separated resonant excitations that follow the normal state dispersions, while their intensity decreases with energy transfer giving only a minor superconducting enhancement at 12 meV. At the resonance, however, $\hbar\omega_{res}=6$ meV, a “flat top” excitation is evident that can be modeled with two incommensurate peaks at $Q_{res}=(\mathbf{q}+\epsilon,\frac{1}{2} \mp \epsilon)$ with $\epsilon=0.05(1)$, a value that departs slightly from the one found in the normal state.

The salient point of these measurements is that in the normal state the magnetic excitations show a behavior consistent with an itinerant antiferromagnet. The onset of superconductivity affects only the lower energy magnetic excitations and spectral weight is shifted upwards to follow the normal-state incommensurate wave-vector dependence except at the spin resonance energy.

Our INS results are qualitatively consistent with the extended $s(\pm)$ model of the superconducting order parameter as we indeed observe the opening of a spin gap and the enhanced peak just above the gap as expected. However it is less clear if the magnetic resonance and its spectral weight in $S(Q,\omega)$ is consistent with this model. To clarify this we compute the magnetic susceptibility within the four-band model used previously to describe the Fermi surface topology in iron-based superconductors\textsuperscript{27} and use the conventional random phase approximation (RPA) which describes the enhancement of the spin response of a metal in presence of the moderately strong (interband and intra-band) repulsive interactions. In Fig. 3(c,d) we show the results for the total physical RPA susceptibility, $\chi_{RPA}(\mathbf{q},\omega_{m})$, calculated for the Fermi surface topology that is consistent with ARPES results on Fe\textsubscript{1.03}Te\textsubscript{0.7}Se\textsubscript{0.3} as a function of the momentum along the transverse direction (1/2+e,1/2-e) and $\hbar\omega$ in the normal (c) and superconducting (d) states (details of these calculations are presented in the Supplementary information).

In order to simulate the experimental resolution we have convoluted the results by the experimental Gaussian with $\Delta q = 0.06$r.l.u. and $\Delta \omega = 1$meV and the original calculated figures are shown in the Appendix. Due to the nesting condition of one of the
The imaginary part of the bare interband susceptibility shows the discontinuous jump implied by the Bogolyubov coherence factor (1 + $\Delta_k$, where $\Delta$ is the superconducting gap.

Above $\Omega_c$, the imaginary part of the bare interband susceptibility shows the discontinuous jump implied by the Bogolyubov coherence factor (1 + $\Delta_k$, where $\Delta$ is the superconducting gap. Thus, the condition $\Delta_k = -\Delta_k + Q_{AF}$ is true for the $s^\pm$-wave symmetry of the superconducting order parameter. Correspondingly, the real part of the bare susceptibility is positive, diverges logarithmically at $\Omega_c = 2\Delta_0$, and scales as $\omega^2$ at small frequencies. Then, switching on the electron-electron interactions, we find within RPA that for any positive value of the interband interaction the total (RPA) spin susceptibility acquires a pole or spin exciton below $\Omega_c$ with infinitely small damping. Therefore, other than the gapping of the spin spectrum, the most salient point of our analysis is that it predicts a spin exciton at $\hbar\omega_{res} \approx 6\text{meV}$ in the superconducting state that is located at the same incommensurate wavevectors as magnons in the normal state, a feature that is clearly found in our INS data. We stress here that the spin exciton is a property of the superconducting state and requires opposite sign of the superconducting order parameter at the corresponding wave vector, i.e., $\Delta_k = -\Delta_k + Q_{AF}$. We remark that the actual spin gap in the neutron spectrum is smaller than $2\Delta_0$, which is again a consequence of the spin exciton formation in the superconducting state.

In the superconducting state, the magnetic susceptibility changes due to opening of the superconducting gap and the corresponding change of the quasiparticle excitations. Here the imaginary part of the bare interband susceptibility (i.e., without electron-electron interaction that yields RPA) is gapped for small frequencies and at the antiferromagnetic wavevector $Q_{AF}$, the value of this gap is determined as $\Omega_c = \min(\Delta_k, |\Delta_k + Q_{AF}|) \sim 2\Delta_0 \sim 8\text{meV}$, where $\Delta_0$ is the superconducting gap. Furthermore, above $\Omega_c$ the imaginary part of the bare interband susceptibility shows the discontinuous jump implied by the Bogolyubov coherence factor (1 + $\Delta_k$, where $\Delta$ is the superconducting gap. Thus, the condition $\Delta_k = -\Delta_k + Q_{AF}$ is true for the $s^\pm$-wave symmetry of the superconducting order parameter. Correspondingly, the real part of the bare susceptibility is positive, diverges logarithmically at $\Omega_c = 2\Delta_0$, and scales as $\omega^2$ at small frequencies. Then, switching on the electron-electron interactions, we find within RPA that for any positive value of the interband interaction the total (RPA) spin susceptibility acquires a pole or spin exciton below $\Omega_c$ with infinitely small damping. Therefore, other than the gapping of the spin spectrum, the most salient point of our analysis is that it predicts a spin exciton at $\hbar\omega_{res} \approx 6\text{meV}$ in the superconducting state that is located at the same incommensurate wavevectors as magnons in the normal state, a feature that is clearly found in our INS data. We stress here that the spin exciton is a property of the superconducting state and requires opposite sign of the superconducting order parameter at the corresponding wave vector, i.e., $\Delta_k = -\Delta_k + Q_{AF}$. We remark that the actual spin gap in the neutron spectrum is smaller than $2\Delta_0$, which is again a consequence of the spin exciton formation in the superconducting state.

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momentum-dependent strong superexchange interaction.

We note that there are intriguing similarities between this iron superconductor and the rare-earth- and actinide-based superconductors UPd$_2$Al$_3$[17-19], CeCu$_2$Si$_2$[20] and CeCoIn$_5$[21]. In all three compounds the magnetisation dynamics in the normal state are modulated in $S(Q,\omega)$. In the superconducting state (i) the low energy response is suppressed by the formation of a gap, (ii) a 'resonance' is formed at energies related to the superconducting gap energy and (iii) the higher energy dynamic response remains unchanged. In this iron superconductor $T_c$ is much higher than the rare-earth- and actinide-based superconductors making this behavior far clearer to observe.

To summarize, we find using inelastic neutron scattering that in the FeTe$_{0.6}$Se$_{0.4}$ superconductor the magnetic resonance appears superimposed at the incommensurate wavevector as paramagnon excitations found in the normal state. Our simple RPA model of the magnetic excitations on the basis of the $s(\pm)$ symmetry of the superconducting order parameters can qualitatively reproduce the main features of the neutron scattering data. The agreement supports an itinerant character of the spin excitations in iron-based systems and a spin spectrum determined by the single poles of $\text{Im}\chi$ with no extra contributions from localized moments. The success of this model stands in sharp contrast to the uncertainties still faced to fully understand the magnetic resonance in the cuprates.

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I. APENDIX

A. Theoretical Calculations

The band structure calculations as well as the ARPES measurements show that the Fermi surface of iron-based superconductors consists of two hole (h) pockets centered around the $\Gamma = (0,0)$ point and two electron (e) pockets centered around the $M = (\pi,\pi)$ point of the folded Brillouin Zone (BZ)[22]. We model the resulting band structure by using the following single-electron model Hamiltonian

\begin{equation}
H_0 = -\sum_{k,\alpha,\sigma} \epsilon^i_{\alpha} n_{k\alpha\sigma} - \sum_{k,\alpha,\sigma} t^i_{k} d^\dagger_{k\alpha\sigma} d_{k\alpha\sigma},
\end{equation}

where $i = \alpha, \beta$, $\alpha, \beta$ refer to the band indices, $\epsilon^i_{\alpha}$ are the on-site single-electron energies, $t^i_{0,\alpha \sigma} = t^{\alpha_{1},\alpha_{2}}_{0} (\cos k_{x} + \cos k_{y}) + t^{\alpha_{1},\alpha_{2}}_{1,0} \cos k_{x} \cos k_{y}$ is the electronic dispersion that yields hole pockets centered around the $\Gamma$ point, and $t^\beta_{k} = t^{\beta_{1},\beta_{2}}_{1} (\cos k_{x} + \cos k_{y}) + t^{\beta_{1},\beta_{2}}_{2} \cos k_{x} \cos k_{y}$ is the dispersion that results in the electron pockets around the $M$ point. This model has been used previously to fit the available ARPES data[23] in optimally doped Ba$_{0.6}$K$_{0.4}$Fe$_2$As$_2$. It has been found recently[24] that the Fermi surface and the corresponding electronic structure of Fe$_1$0.6Te$_{0.7}$Se$_{0.3}$ is very similar to the 122 compounds except for the different level of nesting. To account for this change we use the following parameters $|\langle \epsilon^i, t^i_j, t^i_j \rangle| = (0.26, 0.16, 0.052)$ and $(0.18, 0.16, 0.052)$ for the $\alpha_{1}$ and $\alpha_{2}$ bands, respectively, and $(0.68, 0.38, 0.8)$ and $(0.68, 0.38, -0.8)$ for the $\beta_{1}$ and $\beta_{2}$ bands, correspondingly (all values are in eV).

Next we consider the one-loop contribution to the spin susceptibility that includes the intraband and the interband contributions:

\begin{equation}
\chi^{ij}_0(q, i\omega_n) = -\frac{T}{2N} \sum_{k,\omega_n} \text{Tr} \left[ G^i(k + q, i\omega_n + i\omega_n) G^j(k, i\omega_n) \right] + F^i(k + q, i\omega_n + i\omega_n) F^j(k, i\omega_n)
\end{equation}

where $i, j$ again refer to the different band indices. $G^i$ and $F^i$ are the normal and anomalous (superconducting) Green functions, respectively.

For the four-band model considered here the effective interaction will consist of the intraband and interband

![FIG. 4: Calculated Fermi surface topology for the four-band tight-binding Hamiltonian, Eq.(1). The circular pockets around the $\Gamma$-point refer to the hole $\alpha$-bands, and ellipses arise due to electron $\beta$-bands and are centered around the $M$-point.](image)
repulsion denoted by $U$ and $J$, respectively. Within RPA, the spin response can be written in a matrix form:

$$\hat{\chi}_{RPA}(\mathbf{q}, i\omega_n) = \left[ \mathbf{1} - \Gamma \hat{\chi}_0(\mathbf{q}, i\omega_n) \right]^{-1} \hat{\chi}_0(\mathbf{q}, i\omega_n)$$

(3)

where $\mathbf{1}$ is a unit matrix and $\hat{\chi}_0(\mathbf{q}, i\omega_n)$ is a $4 \times 4$ matrix formed by the interband and intraband bare susceptibilities determined by Eq. (2). The vertex is given by

$$\Gamma = \begin{bmatrix} U & J/2 & J/2 & J/2 \\ J/2 & U & J/2 & J/2 \\ J/2 & J/2 & U & J/2 \\ J/2 & J/2 & J/2 & U \end{bmatrix},$$

(4)

and we assume here $J = 0.05\,\text{eV}$ and $U \sim 0.13\,\text{eV}$. Note that the value of $U$ was chosen in order to stay in the paramagnetic phase.

The results are shown in Fig. 5, where one clearly finds the incommensurate response in both normal and superconducting state.

FIG. 5: Calculated $(\mathbf{q}, \omega)$ mesh of the physical spin susceptibility in the normal (a) and superconducting (b) state without taking into account the resolution function.

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We also note that the compositional dependence of these higher energy excitations appears also to be weak as shown in reference 31.

\[ \epsilon_\alpha^k = -\epsilon_\beta^{k+Q_A} \]