Spin anisotropy of the resonance peak in superconducting FeSe$_{0.5}$Te$_{0.5}$

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We have used polarized-neutron inelastic scattering to resolve the spin fluctuations in superconducting FeSe$_{0.5}$Te$_{0.5}$ into components parallel and perpendicular to the layers. A spin resonance at an energy of 6.5 meV is observed to develop below $T_c$ in both fluctuation components. The resonance peak is anisotropic, with the in-plane component slightly larger than the out-of-plane component. Away from the resonance peak, the magnetic fluctuations are isotropic in the energy range studied. The results are consistent with a dominant singlet pairing state with $s^\pm$ symmetry, with a possible minority component of different symmetry.

The discovery of superconductivity in iron pnictides and chalcogenides with transition temperatures $T_c$ up to 55 K has prompted comparisons with the high-$T_c$ copper-oxide superconductors. In common with the cuprates, the phase diagram of the Fe-based superconductors shows a suppression of static magnetic order and the emergence of superconductivity with doping. Also, as in the cuprates, a spin resonance is observed to develop below $T_c$ in the magnetic spectrum of the Fe-based superconductors as measured by inelastic neutron scattering. The existence of a superconductivity-induced spin resonance peak has been shown to relate to the superconducting pairing state and gap symmetry.

Among the Fe-based superconductors, the iron chalcogenides Fe$_x$Se$_y$Te$_{1-x}$ have the simplest crystal structure (space group $P4/nmm$, room-temperature lattice parameters $a = b \approx 3.8$ Å, $c \approx 6.1$ Å). This, together with the availability of large single crystals, makes them attractive for fundamental studies. Antiferromagnetic order characteristic of the parent compound FeTe persists up to $x \approx 0.1$, after which short-range magnetic order and partial superconductivity coexist for concentrations $x \leq 0.5$. Bulk superconductivity is reported for $x > 0.4$ with a maximum $T_c \approx 15$ K at $x \approx 0.5$.

Inelastic neutron scattering experiments have shown that the spin fluctuations in Fe$_x$Se$_y$Te$_{1-x}$ extend up to 250 meV. A spin resonance is observed to develop below $T_c$ at an energy of 6.5 meV, centered on wave vectors of the form $Q_0 = (0.5,0.5,0)$. The resonance peak is quasi-two-dimensional, which means that it varies only weakly with the out-of-plane wave-vector component $c^*$. The position of the resonance peak in momentum space carries information about the symmetry of the superconducting state. For example, for singlet pairing, the BCS coherence factor enhances the neutron response function when the superconducting gap changes sign between the points on the Fermi surface connected by $Q_0$. In iron-based superconductors, the singlet $s^\pm$ pairing state is consistent with many experimental results, including the existence of a spin resonance at $Q_0$. However, a spin resonance at $Q_0$ is not particular to $s^\pm$. It is also predicted, for example, for certain triplet $p$-wave states.

Until now, inelastic neutron scattering measurements on FeSe$_{0.5}$Te$_{0.5}$ were performed with an unpolarized neutron beam. However, certain superconducting gap functions can result in anisotropic spin susceptibilities at the resonance energy. In this Rapid Communication, we report the results of polarized-neutron inelastic scattering measurements on FeSe$_{0.5}$Te$_{0.5}$, which determine the anisotropy of the imaginary part of the dynamical susceptibility $\chi''(Q,\omega)$. We find that the resonance peak exhibits a spin anisotropy such that the in-plane component $\chi''(Q_x,\omega)$ is larger by about 20% than the out-of-plane component $\chi''(Q_{x'},\omega)$. This is consistent with a dominant singlet superconducting ground state with $s^\pm$ symmetry, and contrasts with a recent polarized-neutron scattering study of BaFe$_{1.9}$Ni$_{0.1}$As$_2$, which revealed a highly anisotropic spin-resonance peak appearing only in the in-plane response.

The single-crystal sample of nominal composition of FeSe$_{0.5}$Te$_{0.5}$ was grown by the modified Bridgman method. Analysis of pulverized crystals from the same batch by x-ray powder diffraction revealed a composition Fe$_{1.045}$Se$_{0.406}$Te$_{0.594}$ with traces of Fe$_2$Se$_3$ (5% volume fraction) and Fe ($\leq 1\%$) as impurity phases. In a previous study, we performed magnetometry measurements on a piece of the same crystal and found bulk superconductivity below $T_c = 14$ K. The neutron scattering sample was rod shaped and had a mass of approximately 5 g. The mosaic spread in the ab plane was found to be 1.5° (full width at half-maximum).

The inelastic neutron scattering measurements were carried out on the IN22 triple-axis spectrometer at the Institut Laue-Langevin. The crystal was aligned with the $c$ axis perpendicular to the scattering plane and mounted in an ILL-type orange cryostat. The spectrometer was operated with a fixed final wave vector of $k_f = 2.66$ Å$^{-1}$ and without collimation. A graphite filter was installed in the scattered beam to suppress contamination by higher-order wavelengths. The analyzer was horizontally focused to increase intensity. The corresponding energy resolution with this setup is approximately 0.8 meV at the elastic position. Longitudinal polarization analysis was performed with the Cryopad device. Cryopad is designed...
Longitudinal polarization analysis allows a complete separation of the magnetic fluctuations perpendicular to the incident neutron polarization from spin fluctuations parallel to the neutron polarization before and after the scattering. Solid lines show least-squares fits to the spectra assuming a Gaussian lineshape. Data in both (a) and (b) were recorded at a temperature of 2 K.

\[
\sigma(y, y) \propto \chi_{ab}'' + N + B_{NSF}^{SF}, \\
\sigma(z, z) \propto \chi_{c}'' + N + B_{NSF}^{SF}, \\
\sigma(x, x) \propto N + B_{NSF}^{SF},
\]

where \( N \) refers to the coherent nuclear cross section and \( B_{SF} \) and \( B_{NSF} \) are the SF and NSF backgrounds. To simplify the notation, we omit the explicit dependence on \( Q \) and \( \omega \) from now on. These scattering processes are represented in Fig. 1. The background was found to be independent of the polarization in the SF cross-sections to within experimental error from measurements at \( Q \approx (0.1, 0.9, 0) \) and \( E \approx 6 \) meV.

Figure 2(a) shows energy scans performed at \( Q_0 = (0.5, 0.5, 0) \) in the three SF channels and in the \( \sigma(x, x) \) NSF channel. The intensity in the \( \sigma(x, x) \) channel is significant, highlighting the importance of using polarized-neutron scattering to separate the nuclear contribution from the magnetic signal. From Eq. (1), the \( \sigma(x, -x) \) cross section contains the total magnetic scattering. The scattering in this channel contains a peak at \( h_0b_0 \approx 6.5 \) meV, corresponding to the spin resonance previously reported by unpolarized-neutron inelastic scattering measurements in compounds of similar composition.\(^7\text{-}^{11}\)

\[\text{Intensity (Counts)}\]

\[\begin{array}{c}
\text{Energy (meV)}\\
\text{Intensity (Counts)}
\end{array}\]

\[\begin{array}{c}
0 & 15 \text{ meV}\\
0 & 500 \text{ Counts}
\end{array}\]

\[\text{Intensity (Counts)}\]

\[\begin{array}{c}
(h, 1 - h, 0) \text{ (r.l.u.)}\\
\text{Intensity (Counts)}
\end{array}\]

\[\begin{array}{c}
0.1 & 1 \text{ meV}\\
0.1 & 300 \text{ Counts}
\end{array}\]
FIG. 3. (Color online) (a) Comparison of the scattering from in-plane ($\chi''_{ab}$) and out-of-plane ($\chi''_c$) magnetic fluctuations in FeSe$_{0.5}$Te$_{0.5}$. Solid lines through the data points are guides to the eye. (b) and (c) Intensity maps showing the cross sections $\sigma(y, -y)$ and $\sigma(z, -z)$, which contain $\chi''_{ab}$ and $\chi''_c$, respectively. All the data in this figure were recorded at $T = 2$ K.

Figure 2(b) shows the $\sigma(x, -x)$ cross section in wave-vector scans along $(h, 1 - h, 0)$ at selected energies. At 3 meV, only a flat background is evident. Above the resonance energy, steeply rising incommensurate magnetic excitations are observed. Our results are consistent with unpolarized-neutron scattering measurements on FeSe$_{0.5}$Te$_{0.5}$ (see Refs. 9 and 10).

The $\sigma(y, -y)$ and $\sigma(z, -z)$ SF channels, shown in Fig. 2(a), contain the magnetic scattering from out-of- and in-plane fluctuations, respectively [see Eq. (1)]. The signal in these channels is very similar throughout the energy range measured, with both channels having a peak at the resonance energy. A small but statistically significant difference is observed between $\sigma(y, -y)$ and $\sigma(z, -z)$ on the resonance peak itself. By using Eq. (1), we can eliminate the background contribution and separate the in- and out-of-plane components of magnetic scattering: $\chi''_{ab} \propto \sigma(x, -x) - \sigma(y, -y)$ and $\chi''_c \propto \sigma(x, -x) - \sigma(z, -z)$. Figure 3(a) shows the result of this procedure. The resonance peak appears at the same energy to within an experimental error of 1 meV in both $\chi''_{ab}$ and $\chi''_c$. The peak is slightly larger in $\chi''_{ab}$. On either side of the spin resonance energy, the intensity is approximately the same for both channels.

The similarity between the $\chi''_{ab}$ and $\chi''_c$ components is emphasized in the color maps shown in Figs. 3(b) and 3(c), which show the intensity distribution as a function of energy and wave vector along $(h, 1 - h, 0)$. The data plotted in these maps are the $\sigma(y, -y)$ and $\sigma(z, -z)$ cross sections, which contain the $\chi''_c$ and $\chi''_{ab}$ fluctuations, respectively. The overall conclusion from all the $T = 2$ K data is that the low-energy spin fluctuations in FeSe$_{0.5}$Te$_{0.5}$ are isotropic ($\chi''_{ab} \approx \chi''_c$) to within experimental error, except on the resonance peak itself, where $\chi''_{ab}$ is approximately 20% larger than $\chi''_c$.

Figure 4 presents the results of measurements of the temperature dependence of the magnetic fluctuations at $Q_0 = (0.5, 0.5, 0)$ and $Q_0 = (0.5, 0.5, 0)$ in FeSe$_{0.5}$Te$_{0.5}$. Here, we show data obtained from the $\sigma(x, -x)$ cross section, which, from Eq. (1), is proportional to $\chi''_{ab}(Q_0, \omega) + \chi''_c(Q_0, \omega)$. Because the measured intensity is proportional to $\chi''(Q, \omega)/(1 - \exp(-\hbar\omega/k_B T))$, we have multiplied the intensity by $1 - \exp(-\hbar\omega/k_B T)$ to compare susceptibilities at different temperatures. We see that the resonance peak disappears above $T_c = 14$ K, while, at higher energies, the susceptibility remains essentially unchanged. At 16 K, we also observe an increased response below the spin gap. Figure 4(b) shows the temperature evolution of the $\sigma(x, -x)$ cross section for temperatures from 2 to 13 K. A scan measured at 16 K (above $T_c$) was subtracted to isolate the spin-resonance contribution. Upon warming, the intensity of the spin resonance shows little change up to 9 K. When the temperature approaches $T_c$, the spectral weight diminishes and the spin gap is gradually filled. Another notable feature is that the spin resonance does not shift to lower energies with increasing temperature, as one might
expect if the spin resonance were simply caused by a gap that closes at $T_c$ with temperature. From our measurements, we conclude that the position and the energy width of the spin resonance are temperature independent up to at least $\sim 0.8T_c$. The lack of softening of the resonance energy with increasing temperature has also been found in FeSe$_{0.1}$Te$_{0.9}$. Figure 4(c) shows the evolution of the integrated intensity of the spin resonance, which behaves as an order parameter of the superconducting phase. In the vicinity of $T_c$, measurements with higher precision are needed to obtain a more quantitative estimate of the renormalization of the inelastic intensity than is available from the present experiment.

The polarized-neutron data presented here go beyond what has hitherto been possible with unpolarized-neutron scattering, and provide insight into the magnetic excitations of FeSe$_{0.5}$Te$_{0.5}$. A superconducting wave function with purely $s^\pm$ pairing state would result in an isotropic spin-resonance peak. Our data suggest a small anisotropy, in the sense $\chi''_{ab} > \chi''_{xx}$. This small anisotropy cannot readily be explained by the usual anisotropic terms in the spin Hamiltonian since the magnetic scattering is isotropic above and below the resonance peak. It is possible, therefore, that the superconducting pairing function contains a minority component with a different symmetry. For example, a spin-triplet with sign-reversed $p$-wave gap is predicted to give a resonance in $\chi''_{ab}$, but not in $\chi''_{xx}$.

The relatively small anisotropy in the spin resonance of FeSe$_{0.5}$Te$_{0.5}$, consistent with a dominant singlet-triplet excitation, is in stark contrast to the results of a study on BaFe$_{1.9}$Ni$_{0.1}$As$_2$, which revealed a highly anisotropic spin resonance with only the $\chi''_{ab}$ component nonzero. The results also differ from the spin-ladder system Sr$_{14}$Cu$_{24}$O$_{41}$, which also has a resonancelike coherent singlet-triplet excitation. First, the anisotropy is in the opposite sense (in Sr$_{14}$Cu$_{24}$O$_{41}$, the out-of-plane fluctuations are stronger than the in-plane fluctuations), and second, the anisotropy in Sr$_{14}$Cu$_{24}$O$_{41}$ is observed over a range of energies, not just on the peak.

Recently, polarized-neutron scattering measurements have been performed on YBa$_2$Cu$_3$O$_{6+\delta}$. The spin resonance in YBa$_2$Cu$_3$O$_{6+\delta}$ at 40 meV, corresponding to the odd-parity mode, was found to be quasi-isotropic to within the precision of the measurements. This implies that the resonance peak is predominantly a singlet-triplet excitation in both FeSe$_{0.5}$Te$_{0.5}$ and YBa$_2$Cu$_3$O$_{6+\delta}$. Furthermore, the resonance peaks in these materials do not soften appreciably as the temperature is increased toward $T_c$ (see Fig. 4 and Ref. 29). These similarities suggest that the superconducting states in the cuprates and Fe-based superconductors have some general features in common.