Temperature dependence of the resonance and low-energy spin excitations in superconducting FeTe$_{0.6}$Se$_{0.4}$

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We use inelastic neutron scattering to study the temperature dependence of the low-energy spin excitations in single crystals of superconducting FeTe$_{0.6}$Se$_{0.4}$ (T_c = 14 K). In the low-temperature superconducting state, the imaginary part of the dynamic susceptibility at the electron and hole Fermi-surfaces nesting wave vector $Q = (0.5,0.5)$, $\chi''(Q,\omega)$, has a small spin gap, a two-dimensional neutron spin resonance above the spin gap, and increases linearly with increasing $\hbar\omega$ for energies above the resonance. While the intensity of the resonance decreases like an order parameter with increasing temperature and disappears at temperature slightly above T_c, the energy of the mode is weakly temperature dependent and vanishes concurrently above T_c. This suggests that in spite of its similarities with the resonance in electron-doped superconducting BaFe$_{2-x}$(Co,Ni)$_x$As$_2$, the mode in FeTe$_{0.6}$Se$_{0.4}$ is not directly associated with the superconducting electronic gap.

I. INTRODUCTION

Soon after the discovery of high-temperature (high-T_c) superconductivity in Fe-based materials,[1–4] neutron-scattering studies revealed that the parent compounds of these superconductors have an antiferromagnetic (AFM) ground state[5–7] similar to those of unconventional heavy Fermions and copper oxide superconductors.[8] This observation has inspired many theories to postulate that spin fluctuations in these materials may be responsible for electron pairing and superconductivity.[9–18] In one of the leading theories, superconductivity arises from quasiparticle excitations between the electron and hole pockets near M and Γ points of the Brillouin zone, respectively. One of the consequences of opening up electronic gaps in the superconducting state is that there should be a neutron spin resonance. The energy of the resonance should be coupled to the addition of the hole and electron superconducting gap energies [$\hbar\omega = |\Delta(k + Q)| + |\Delta(k)|$], and the intensity of the mode should follow the superconducting order parameter.[15–17] Indeed, the discovery of the neutron spin resonance in electron- and hole-doped iron pnictide BaFe$_2$As$_2$ at the AFM wave vector $Q = (0.5,0.5,L)$ in the tetragonal unit-cell notation ($a = b = 3.963$ and $c = 12.77$ Å)[19–26] suggests that superconductivity arises from quasiparticle excitations between the signed reversed electron and hole pockets. This notion is further confirmed by the temperature[27] and magnetic field[28] dependence of the mode energy, which is directly coupled to the superconducting electronic gap energy. For iron chalcogenide Fe$_{1+\delta}$Te$_{1-x}$Se$_x$, previous inelastic neutron-scattering experiments[29–40] have also established the presence of a resonance at the electron-hole Fermi-surface nesting wave vector, which is the same as the AFM ordering wave vector for iron pnictides,[20–25] and the intensity of the resonance increases below T_c just like it does for iron pnictides. Therefore it appears that the neutron spin resonance is ubiquitous for different families of iron-based superconductors and directly correlated with superconducting electronic gaps.[41]

In this paper, we report inelastic neutron-scattering studies of superconducting FeTe$_{0.6}$Se$_{0.4}$ ($T_c = 14$ K). Although there are extensive neutron-scattering measurements on non-superconducting and superconducting Fe$_{1+\delta}$Te$_{1-x}$Se$_x$,[29–40] our detailed wave-vector and energy-dependent studies of the neutron spin resonance provide interesting information concerning the nature of the mode and its relationship to the superconducting electronic gap. First, we confirm the earlier work[30] that the mode is purely two dimensional and dispersionless for wave vectors along the c axis, which is different from the dispersive nature of the resonance in electron-doped BaFe$_{2-x}$(Co,Ni)$_x$As$_2$.[21–23] Second, we extend the earlier work[30] on the temperature dependence of the mode. By carrying out systematic series of energy scans very close and above the superconducting transition temperature T_c, we find that the energy of the mode is essentially temperature independent and collapses at a temperature slightly above T_c, and does not follow the temperature dependence of the superconducting electronic gap as determined from Andreev reflection measurements.[42] Finally, we show that the intensity gain of the resonance is approximately compensated by spectral weight loss at energies below it, and there is a spin gap opening for low-energy spin excitations below T_c. These results suggest that the neutron spin resonance in the FeTe$_{0.6}$Se$_{0.4}$ system may not be directly coupled to the superconducting electronic gap as those for BaFe$_{2-x}$(Co,Ni)$_x$As$_2$.[27,28] We discuss possible microscopic origins for this phenomenon.

II. EXPERIMENTAL DETAILS

We carried out neutron-scattering experiments on the HB-3 thermal triple axis spectrometer at the High Flux Isotope Reactor (HFIR), Oak Ridge National Laboratory. We used a pyrolytic graphite PG(002) monochromator and analyzer with a collimation of 48′-monochromator-60′-sample-80′-analyzer-240′-detector. The data were collected in fixed E_f mode at 14.7 meV with a PG filter placed between the sample and
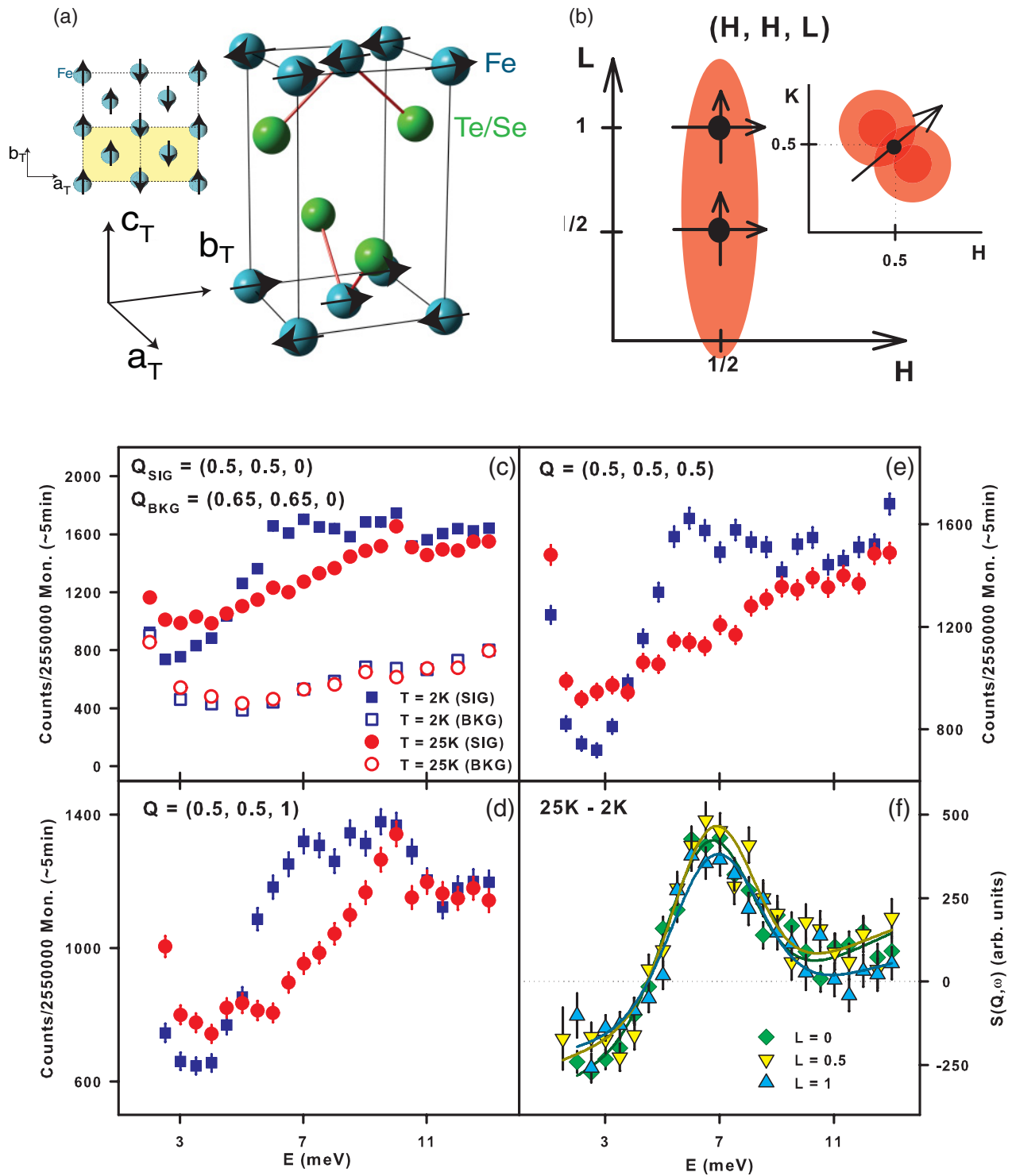
FIG. 1. (Color online) (a) Diagram of the Fe spin ordering with the shaded region defining the magnetic unit cell. (b) Cartoon of the scan directions though the $(1/2, 1/2, L)$ nesting vector. The inset illustrates the direction in the $[H, K]$ plane to which scans were confined. Excitations at $(1/2, 1/2, L)$ in $FeTe_{1-x}Se_x$ consist of two incommensurate peaks that spread away from one another in the transverse direction. The red circles in the inset depict these excitations with the radius of the circles equal to twice the FWHM of the $(1/2, 1/2, 0)$, 7.5 meV resonance peaks measured on crystals from the same batch on a different experiment. The separation of their centers is set to agree with the dispersion mapped out in this previous experiment (Ref. 34). (c)–(e) Energy scans about the 7 meV resonance position above and below T_c for $L = 0, 1/2, 1$. Clear intensity gain is observed inside the superconducting state. The background at $L = 0$ is plotted above and below T_c and is found to be identical, allowing direct temperature subtraction of the scans with no need for background correction. (f) Temperature subtraction of energy scans shown in panels (c)–(e) demonstrating no observable dispersion of the resonance energy along L.
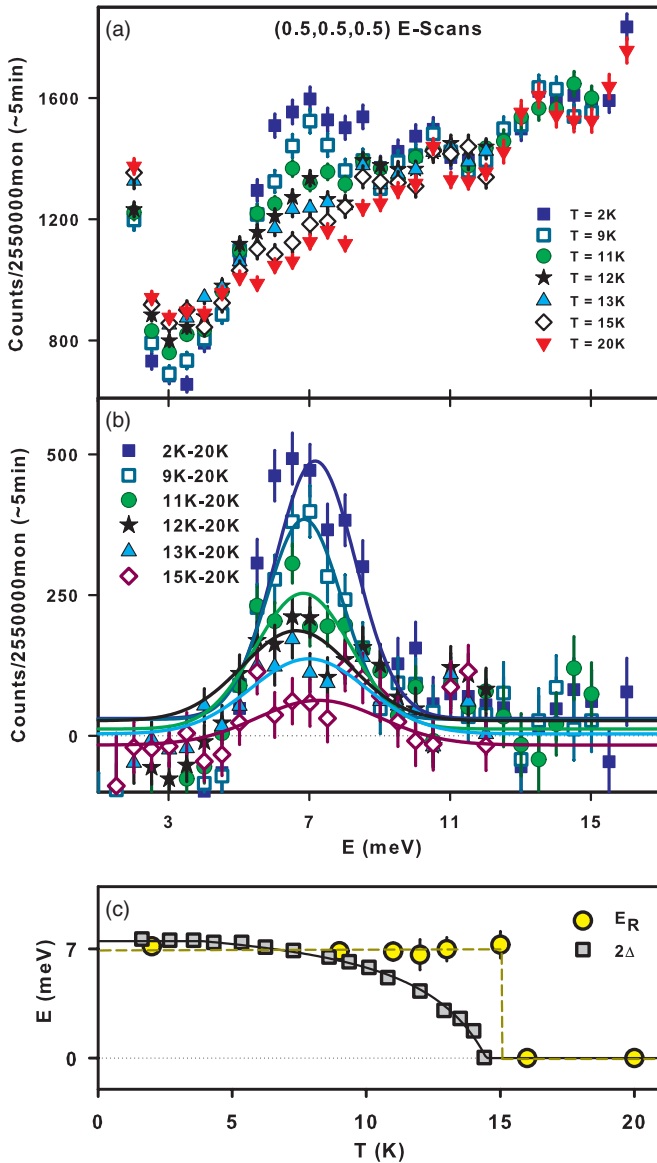
FIG. 2. (Color online) (a) Raw data for energy scans at $Q = (1/2, 1/2, 1/2)$ for multiple temperatures below T_c. At 2 K the 7 meV resonance is clearly present. A strong reduction in scattering for energies below 4 meV is also visible, indicating the opening of a gap in the system. Subsequent Q scans, however, show that this is not a true gap. As the temperature increases to T_c the resonance suppresses and the partial gap closes up. (b) Temperature subtraction of scans shown in panel (a). All of the data is fit with a Gaussian leaving the center energy as a free parameter to be determined. (c) Position of the resonance energy vs temperature as determined from the fits in panel (b); note that circles above $T = 15$ K are meant to indicate that the resonance has been completely suppressed. The temperature dependence of the superconducting gap (Ref. 42) is also graphed, explicitly demonstrating that the resonance does not shift in energy as a function of temperature so as to remain inside 2Δ as required by the spin exciton scenario.

analyzer to remove contamination from higher-order reflections. We coaligned two single crystals in the $[H,H,L]$ scattering plane and loaded them in a liquid-He orange cryostat. The total mass was $\sim$10 g with an in-plane and out-of-plane mosaic of $2.0°$ and $2.1°$ full width at half

maximum (FWHM), respectively. We defined the wave vector Q at (q_x, q_y, q_z) as $(H,K,L) = (q_x a/2\pi, q_y b/2\pi, q_z c/2\pi)$ reciprocal-lattice units (rlu) using the tetragonal unit cell (space group $P4/nmm$), where $a = 3.8$ Å, $b = 3.8$ Å, and $c = 6.0$ Å. In the parent compound, FeTe, the AFM Bragg peaks occur at the $(1/2, 0, 1/2)$ and equivalent wave vectors, corresponding to the crystallographic spin arrangement depicted in Fig. 1(a).[6,7] In the nonsuperconducting FeTe$_{1-x}$Se$_x$ samples ($x \leqslant 0.3$), spin excitations coexist at both the $(1/2, 0, 1/2)$ AFM wave vector, and the $(1/2, 1/2, L)$ wave vector associated with nesting of electron and hole pockets on the Fermi surface.[35,36,38–40] Upon reaching optimal doping, spin excitations at the AFM wave vector are suppressed, however, they remain strong near the nesting vector and consist of a commensurate resonance mode (in the superconducting state) sitting on top of an incommensurate magnetic signal that follows an hourglass dispersion at low energies.[34] We chose the $[H,H,L]$ scattering plane for our experiments since this zone gives us full freedom to probe the L dependence of the resonance. In general, the excitations in this system are extremely diffuse and, as a result, much broader than the instrumental resolution. To quantify this, we have calculated the resolution along the $(H, 1 - H)$ direction at the $(0.5, 0.5)$ position as a function of energy. The resulting instrumental resolution width in FWHM is roughly 20 times smaller than the incommensurate peak separation. Thus our data collection is a good measure of signal centered directly at the $(0.5, 0.5)$ position.

III. RESULTS

In previous work on electron-doped BaFe$_{2-x}$(Co,Ni)$_x$As$_2$ superconductors, the neutron spin resonance has been found to be dispersive along the c axis, occurring at slightly different energies for $L = 0$ and $L = 1$.[21–24] Although previous measurements suggest that the resonance in FeTe$_{1-x}$Se$_x$ is two dimensional,[29,30] there have been no explicit measurements of the resonance at different L values. With this in mind, we have carried out detailed energy scans of bulk superconducting FeTe$_{0.6}$Se$_{0.4}$ at the resonance wave vector $(1/2, 1/2, L)$ as a function of temperature and L. Figures 1(c)–1(e) show constant-Q scans at the signal $Q = (0.5, 0.5, 0)$, $(0.5, 0.5, 0.5)$, $(0.5, 0.5, 1)$ and background $Q = (0.65, 0.65, 0)$ positions above and below T_c. Consistent with earlier results,[29,30] we see a clear enhancement of scattering around $E \approx 7$ meV below T_c at the signal wave vectors for all the L values probed. Figure 1(f) overplots the temperature differences between 2 and 25 K data for three L values. It is clear that for all L values the resonance energy is the same within the errors of our measurements ($E = 6.95 \pm 0.5$ meV). Therefore the mode is indeed two dimensional and has no dispersion along the c axis.[29,30]

In previous neutron-scattering experiments on optimally electron-doped BaFe$_{2-x}$Co$_x$As$_2$, careful temperature dependence measurements revealed that the energy of the resonance with increasing temperature tracks the temperature dependence of the superconducting gap energy.[27] These results, as well as the magnetic-field effect of the resonance,[28] provided compelling evidence that the resonance energy is intimately associated with the superconducting electronic gap energies.
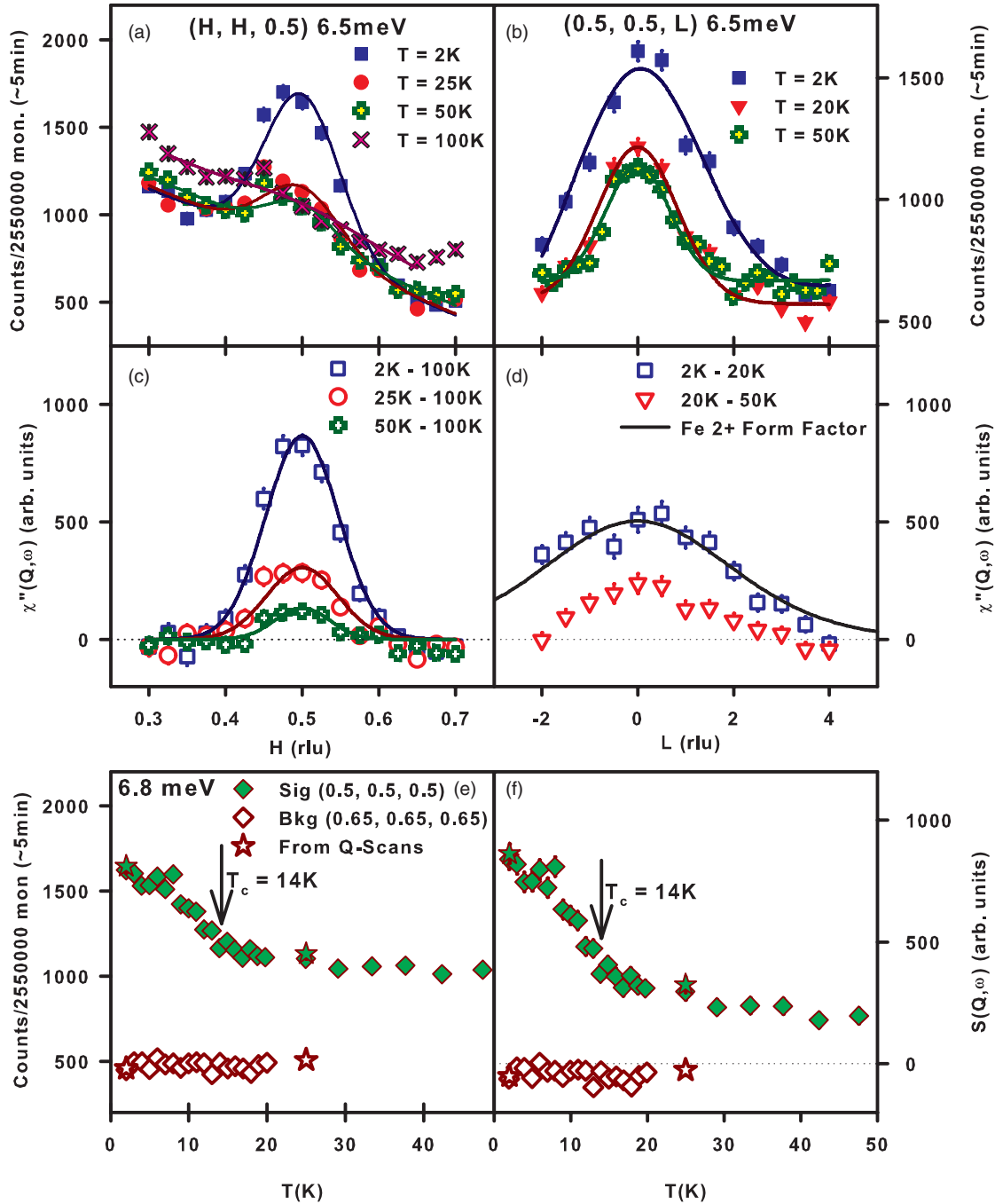
FIG. 3. (Color online) (a),(b) Raw Q-scan data along $[H,H]$ and L, respectively, at $E_R = 6.5$ meV at several temperatures. (c),(d) $\chi''(Q,\omega)$ is determined by subtraction of the background and correcting for the Bose factor. In (c) the 100 K data were used as a final background subtraction in order to remove a spurion at (0.45, 0.45, 0.5) and a phonon tail for points near (0.7, 0.7, 0.5). (d) The intensity gain due to the resonance is determined by subtraction of the 2 and 20 K data. The resulting signal is very broad and fits well to the Fe^{2+} form factor; a testament to the two-dimensional (2D) nature of the resonant mode. (e),(f) Temperature dependence of the resonance for $Q = (1/2, 1/2, 1/2)$ and $E = 6.8$ meV. The resonance suppresses as an order parameter as T_c is approached.

To see if the resonance in $FeTe_{0.6}Se_{0.4}$ behaves similarly, we carried out a series of energy scans from base temperature (2 K) to just above T_c (20 K) at $Q = (0.5,0.5,0.5)$ [Fig. 2(a)]. As the temperature is increased, we see that the resonance drops monotonically in intensity. To accurately determine the temperature dependence of the mode, the energy scans in the superconducting state were subtracted from the energy scan at 20 K in the normal state. The resulting plots of the resonance intensity gain were then fit to a Gaussian on a linear background with the center left as a free parameter [Fig 2(b)]. By plotting the fitted values of the resonance energy as a function of temperature [Fig. 2(c)], we see that the resonance energy is essentially temperature independent until it abruptly disappears above T_c. This is clearly different from the
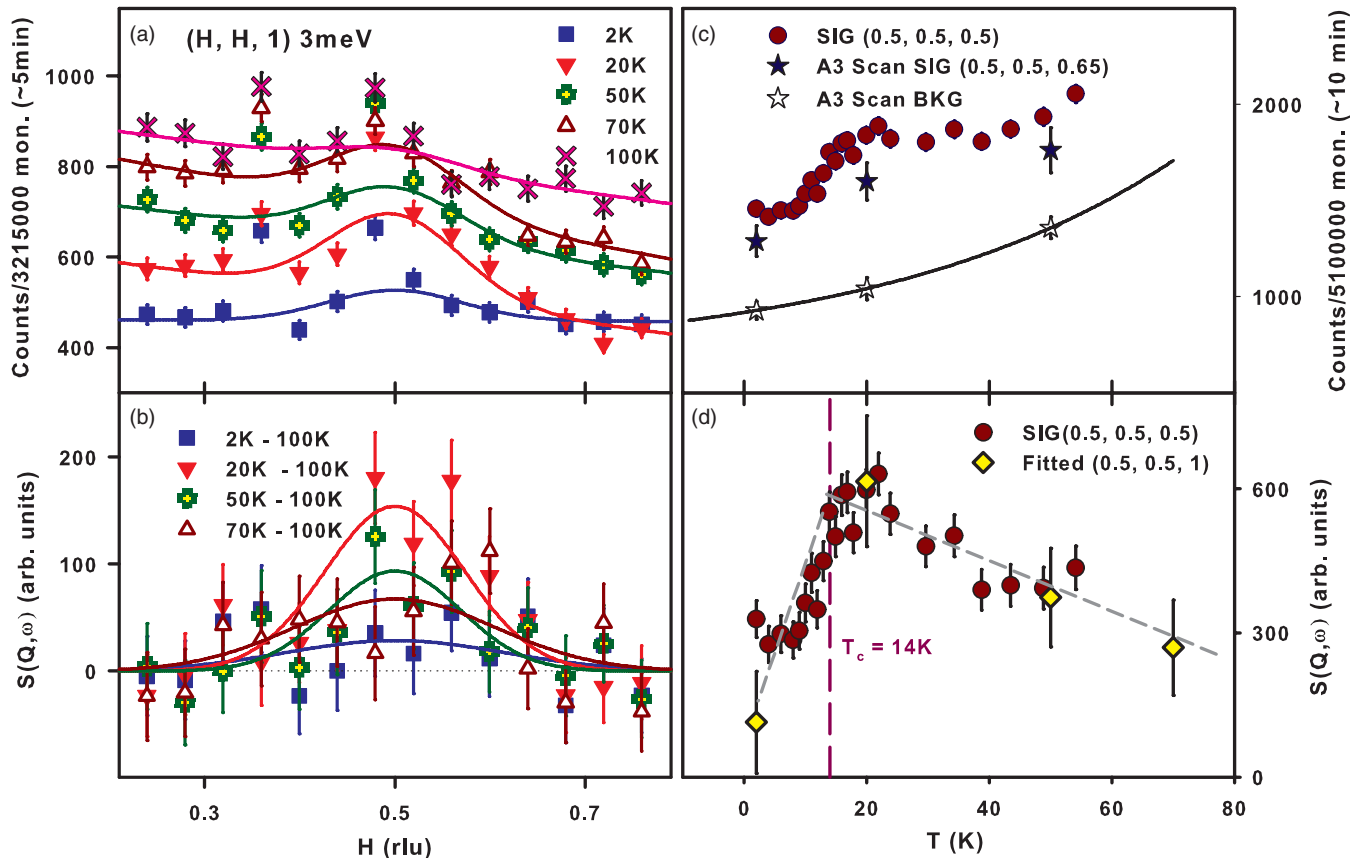
FIG. 4. (Color online) (a),(b) Q-scan data along the $[H,H]$ direction for $L = 1$ and $E = 3$ meV. The scattering becomes stronger as T_c is approached from higher temperatures; upon entering the superconducting state the intensity drops significantly by 2 K but does not fully gap. (c) Temperature dependence at 3 meV inside of the pseudo-spin-gap region reveals that near T_c a gap begins to form but never fully forms by base temperature. (d) $S(Q,\omega)$ of the temperature scan as determined by interpolating and subtracting the background collected using A3 rocking curves. Yellow diamonds correspond to cross checks with fitted Q scans from panels (a) and (b). Since the Q scans and temperature scan were collected on different experiments, the data sets were not normalized to one another by monitor count but rather shifted to coincide at 20 K.

temperature dependence of the resonance for electron-doped $BaFe_{2-x}Co_xAs_2$ (Ref. 27) and the temperature dependence of the superconducting gap for $FeTe_{0.6}Se_{0.4}$ as determined from the Andreev reflection measurements [Fig. 2(c)].[42]

To further characterize the resonance, a series of Q scans were carried out at $E = 6.5$ meV. Scans along the $[H,H]$ direction for $L = 0.5$ confirm that the resonance peaks at the (0.5,0.5) position with a strong gain in intensity in the superconducting state [Figs. 3(a) and 3(c)]. For temperatures above 20 K, the drop in intensity is much more gradual with the peak at (0.5,0.5) fully suppressed by 100 K. Similar scans along the $[0.5,0.5,L]$ direction [Figs. 3(d)–3(f)] reveal that the scattering is much broader. The intensity gain of the resonance is extracted by subtraction of the 20 K data from the 2 K data. The L dependence of the signal fits well to the Fe^{2+} form factor, a further indication that the resonance is purely two dimensional in nature. A temperature scan at (0.5,0.5,0.5) for $E = 6.8$ meV confirmed that the resonance is strongest at base temperatures and then reduces like an order parameter to T_c in good agreement with earlier measurements of the system.[29,30,34,38]

Interestingly, the 15 K energy scan in Fig. 2(b) and the temperature dependence of the resonance in Figs. 3(e) and 3(f)

suggest that the resonance mode first forms at a temperature slightly above T_c. This behavior was also observed by Qiu *et al.*[30] in their temperature and energy scans of the resonance in $FeSe_{0.4}Te_{0.6}$. A similar analysis on optimally electron-doped $BaFe_{1.85}Co_{0.15}As_2$ (Ref. 27) and $BaFe_{1.9}Co_{0.1}As_2$ (Ref. 28) does not display such behavior. Although the origin of this effect is unclear, it is consistent with the idea of preformed Cooper pairs developing in the normal state just above T_c. For comparison, we note that the neutron spin resonance in underdoped $YBa_2Cu_3O_{6+x}$ can extend more than 50 K above T_c.[43]

From Figs. 1 and 2, we see that the intensity gain of the resonance in the superconducting state is accompanied by a loss in signal for energies below 4 meV, suggesting that conservation of spectral weight is satisfied by a reduction of scattering below the resonance energy. However, earlier measurements[34] suggest that the spin gap in $FeTe_{0.6}Se_{0.4}$ is unclean and does not fully open until ~1 meV. Thus it is interesting to investigate the temperature dependence of the spin excitations for energies above the spin gap and below the resonance. Figure 4(a) shows Q scans along the $[H,H,1]$ direction at different temperatures. With increasing temperature from 2 K, a peak at (0.5,0.5,1) above background initially increases at $T = 20$ K,
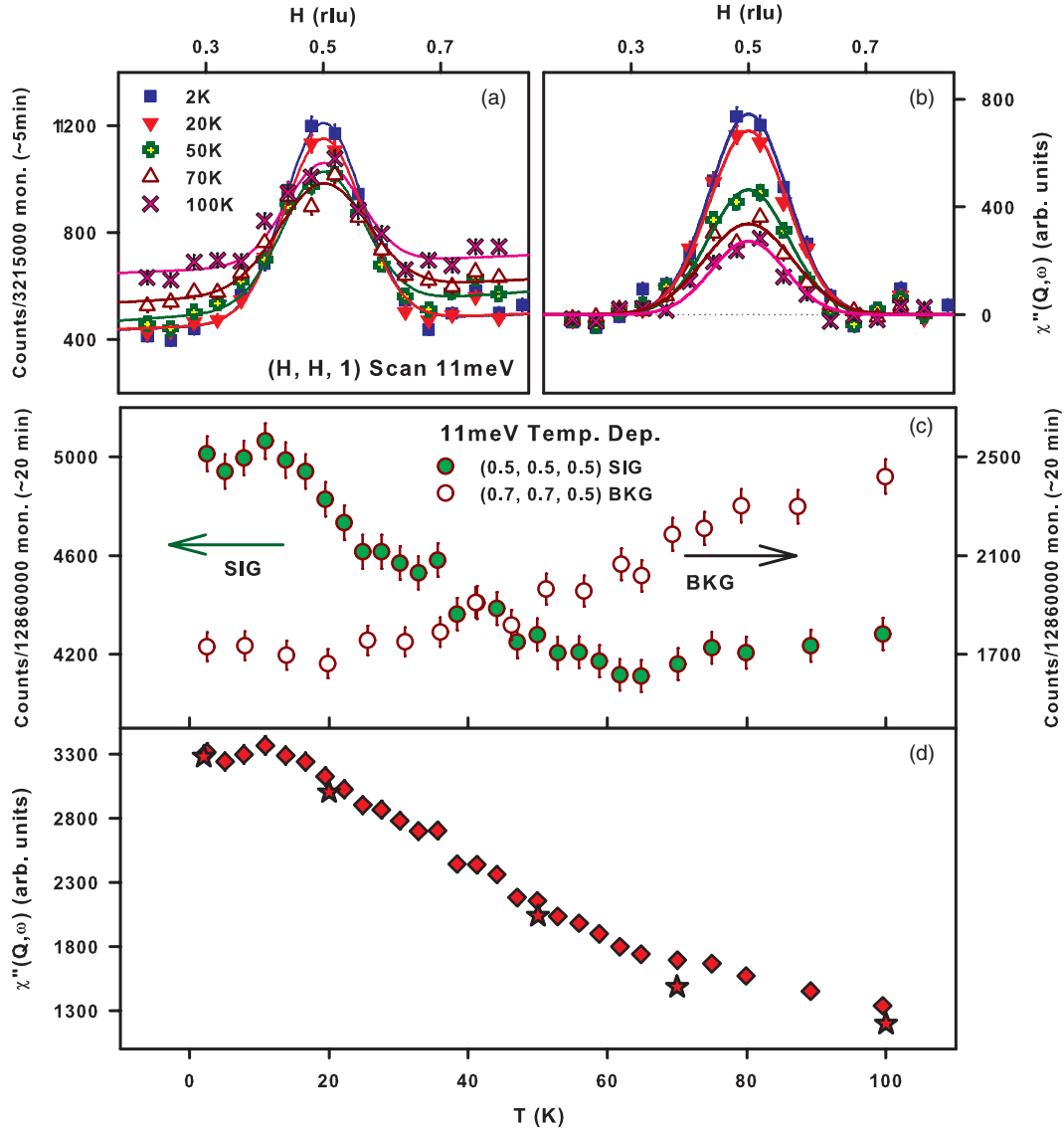
FIG. 5. (Color online) (a) Raw Q-scan data along the $[H,H]$ direction for $L=1$ and $E=11$ meV. (b) $\chi''(Q,\omega)$ determined by background subtraction and correcting for the Bose factor. The resonance is no longer visible; instead the scattering at 2 K is nearly identical to 20 K. Upon entering the normal state, the intensity begins dropping monotonically with increasing temperature but remains robust up to 100 K. (c),(d) Temperature scan at $(1/2, 1/2, 1/2)$ for $E=11$ meV. Red stars correspond to cross checks with fitted peak intensities from Q scans in panel (a) that have been form factor corrected and normalized by monitor count.

then decreases upon further warming until disappearing at 100 K. Assuming that there are only background scattering at 100 K, the temperature difference plots in Fig. 4(b) confirm that the magnetic scattering increases on warming to T_c and then decreases with further increasing temperature. Figure 4(c) shows the detailed temperature dependence data at the signal $Q=(0.5,0.5,0.5)$ and background (sample rotated away from the signal position by 30°) position. As we can see, the scattering shows a clear kink at T_c and decreases monotonically above T_c with warming. Figure 4 shows the background corrected temperature dependence of the magnetic scattering assuming that the temperature dependence of the background follows the solid line in Fig. 4(c). The effect of superconductivity is to open a pseudogap in spin excitations spectrum below T_c.

For optimally electron-doped $BaFe_2As_2$, the enhancement of the resonance occurs at the expense of a full spin gap opening below the resonance.[20,21] However, the situation for Fe(Se,Te) was not completely clear since there are no clean spin gaps for Fe(Se,Te). Furthermore, it was not even clear whether the reduction in magnetic intensity at energies below the resonance occurs around T_c, when the resonance appears. From our data, we see that this is indeed the case. It is worth noting that in lightly doped, nonsuperconducting FeTe, measurements at (0.5,0.5) also reveal a loss in scattering at 3 meV. However, for this underdoped system no resonance is present to suck away spectral weight. Rather, the signal loss is due to the fact that at lower dopings there exists inelastic magnetic scattering at both (0.5,0) and (0.5,0.5) with a strong crossover of spectral weight between these wave vectors occurring around 3 meV.[40]
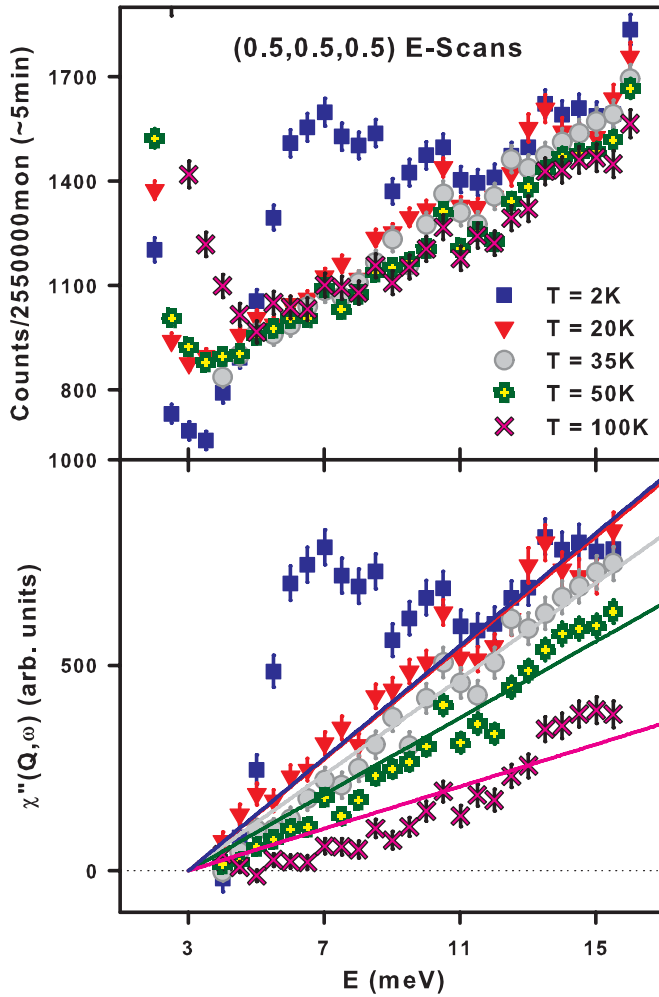
FIG. 6. (Color online) (a) Energy scans focusing on temperatures above T_c. (b) The background subtraction of $\chi''(Q,\omega)$ is determined from Q scans. Aside from the resonance in the 2 K data, all other energy scans follow a similar linear trend; fanning out as a function of temperature.

To determine whether spin excitations at energies above the resonance also respond to superconductivity, we carried out a series of constant-energy $E = 11$ meV scans along the $[H,H,1]$ direction. The outcome shown in Figs. 5(a) and 5(b) reveals that magnetic scattering gradually increases in intensity on cooling. However, upon entering the superconducting state, the scattering appears to level off with the 2 and 20 K Q scans nearly identical in intensity. Temperature scans at $E = 11$ meV at the signal $[Q = (0.5,0.5,0.5)]$ and background $[Q = (0.7,0.7,0.5)]$ positions are shown in Fig. 5(c). The background and Bose factor corrected temperature-dependent imaginary part of the dynamic susceptibility, $\chi''(Q,E)$, at $Q = (0.5,0.5,0.5)$ and $E = 11$ meV is shown in Fig. 5(d). It is clear that the magnetic scattering grows with decreasing temperature but essentially saturates at temperatures below ~ 15 K.

Finally, Fig. 6(a) shows the temperature evolution of the constant-Q $[Q = (0.5,0.5,0.5)]$ scans from 2 to 100 K. After correcting for the temperature dependence of the background scattering and Bose population factor, we obtain the temperature dependence of $\chi''(Q,E)$ at $Q = (0.5,0.5,0.5)$ [Fig. 6(b)]. The $\chi''(Q,E)$ increases linearly with increasing

energy, and the resonance appears below T_c together with the opening of a spin gap at lower energies. These results are consistent with earlier work.[29,30]

IV. DISCUSSIONS AND CONCLUSIONS

The presence of a neutron spin resonance in various high-T_c copper oxide and Fe-based superconductors has been suggested as the result of a spin-fluctuation mediated electron pairing mechanism.[41,44] In an earlier work mostly on copper oxide superconductors,[44] it was proposed that the resonance energy is universally associated with the superconducting electronic gap Δ via $\hbar\omega_{res}/2\Delta = 0.64$ instead of being proportional to the superconducting transition temperatures T_c.[45] In a more recent summary of neutron-scattering data on iron-based superconductors,[41] it was found that the energies of the resonance for underdoped $BaFe_{2-x}(Co,Ni)_xAs_2$ deviate from this relationship, particularly for the resonance energy at $L = 0$. For $FeTe_{0.6}Se_{0.4}$, angle-resolved photoemission spectroscopy experiments[46] reveal a 4.2 meV gap on the electron Fermi surface and a 2.5 meV gap on the hole Fermi surface. Since the addition of the electron and hole superconducting electronic gap energies is consistent with the energy of the resonance at low temperature, the result has been interpreted as evidence that the resonance in $FeTe_{0.6}Se_{0.4}$ also arises from electron-hole pocket excitations.[46] However, given that the superconducting gap energy gradually decreases for temperatures approaching T_c, the resonance energy will exceed that of the superconducting gap energy, contrary to the expectation for a spin exciton in the sign revised s-wave electron pairing scenario.[15–17]

If superconductivity in iron-based materials is mediated by orbital fluctuations associated with a fully gapped s-wave state without sign reversal (s^{++}-wave state), one would expect a neutron spin resonance at an energy above the addition of the electron and hole superconducting electronic gap energies.[47] Since the superconducting gaps decrease with increasing temperature, one would expect a reduction in the resonance energy with increasing temperature even in this scenario, contrary to the observation. In the SO(5) theory for high-T_c superconductivity,[48] the neutron spin resonance is a product of particle-particle excitations and is fixed in energy in the superconducting state. Although this is consistent with present work, it remains unclear how the SO(5) theory originally designed for high-T_c copper oxide superconductors would apply in the case of iron-based superconductors. In any case, our results suggest that the resonance itself may not be directly associated with the superconducting electronic gap in the $FeTe_{0.6}Se_{0.4}$ system.

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