Effect of the in-plane magnetic field on the neutron spin resonance in optimally doped $FeSe_{0.4}Te_{0.6}$ and $BaFe_{1.9}Ni_{0.1}As_2$ superconductors

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We use inelastic neutron scattering to study the effect of an in-plane magnetic field on the magnetic resonance in optimally doped superconductors $FeSe_{0.4}Te_{0.6}$ ($T_c = 14$ K) and $BaFe_{1.9}Ni_{0.1}As_2$ ($T_c = 20$ K). While the magnetic field up to 14.5 T does not change the energy of the resonance, it partially suppresses T_c and the corresponding superconductivity-induced intensity gain of the mode. However, we find no direct evidence for the field-induced spin-1 Zeeman splitting of the resonance. Therefore it is still unclear if the resonance is the long-sought singlet-triplet excitation directly coupled to the superconducting electron Cooper pairs.

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I. INTRODUCTION

The neutron spin resonance is arguably the most important collective magnetic excitations in unconventional superconductors that near an antiferromagnetic (AF) instability.[1–8] Experimentally, the resonance can be broadly defined as a superconductivity-induced gain in the magnetic scattering and the corresponding imaginary part of the dynamical susceptibility, $\chi''(\omega, Q)$, at the AF wave vector. The hallmark of the resonance is the increase of its magnetic intensity below T_c like a superconducting order parameter.[1–8] Although the microscopic origin of the resonance is still unclear, the mode is generally believed to arise from the spin-1 singlet-triplet excitations of the electron Cooper pairs.[9] Here, the process of the singlet-to-triplet excitation of an electron Cooper pair can be denoted as $|0\rangle \Rightarrow |1\rangle$ [Fig. 1(a)], where $|0\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)$ and $|1\rangle = \{|\uparrow\uparrow\rangle, \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle), |\downarrow\downarrow\rangle\}$ are singlet and triplet states, respectively. In some cases,[9] the resonance maybe a singlet-to-doublet excitation that only involves the $|\uparrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$ states [11], as shown in Fig. 1(c).

If the resonance is indeed a spin-1 singlet-triplet excitation,[10] it should Zeeman split into three peaks under the influence of a magnetic field. The degenerate triplet state $|1\rangle$ will split into three energy levels following the Zeeman energy $\Delta E = \pm g\mu_B B$ [Fig. 1(b)],[11] where $g = 2$ is the Lande factor and B is the magnitude of the field. If the resonance is the singlet-doublet excitation [Fig. 1(c)], the splitting of the mode under the field can be observed, as shown in Fig. 1(d). For high-transition-temperature (high-T_c) copper oxide superconductors, application of a 14-T magnetic field only suppresses the intensity of the resonance with no evidence for the expected Zeeman splitting.[12–14] In the case of Fe-based superconductors, where the neutron spin resonance is believed to arise from the electron-hole pocket excitations[15] and has been found in hole-/electron-doped $BaFe_2As_2$ and Fe(Te,Se) superconductors,[3–6,8,16] there are several neutron-scattering experiments probing the effect of a magnetic field on the resonance. In the first neutron measurement, application of a 14.5-T c axis aligned magnetic field on the optimally electron-doped $BaFe_{1.9}Ni_{0.1}As_2$ ($T_c = 20$ K) was found to reduce the intensity and shift down the energy of the mode with no evidence of the Zeeman splitting.[17] In separate experiments on the optimally superconducting Fe(Te,Se),[18–23] a 7-T in-plane magnetic field can suppress the intensity of the resonance without shifting its energy or changing its width.[24] For underdoped $BaFe_{1.92}Ni_{0.08}As_2$ ($T_c = 17$ K), where static AF order co-exists with superconductivity at zero field, application of a 10-T in-plane field enhances the AF order at the expense of the resonance, again with no evidence for the Zeeman splitting.[25] Although a recent neutron-scattering experiment on $FeSe_{0.4}Te_{0.6}$ using a c-axis aligned field suggests the presence of a field-induced Zeeman splitting of the resonance,[26] it is still not clear whether the resonance is a spin-1 mode given the statistics of the data. Since a c-axis aligned field can suppress superconductivity much more efficiently,[27] the best geometry to observe the Zeeman splitting is to align the magnetic field within the Fe plane, where the field-induced suppression of superconductivity is much less.

II. EXPERIMENTS

In this paper, we report inelastic neutron-scattering experiments measuring the effect of the in-plane magnetic field on the resonance in $FeSe_{0.4}Te_{0.6}$ ($T_c = 14$ K, ∼4 g, mosaic ∼ 2.5°) and $BaFe_{1.9}Ni_{0.1}As_2$ ($T_c = 20$ K, ∼6 g, mosaic ∼ 2°). Our experiments on $FeSe_{0.4}Te_{0.6}$ were carried out on PANDA cold neutron triple-axis spectrometer at Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II), TU München, Germany and on SPINS cold neutron triple-axis spectrometer at the NIST Center for Neutron Research (NCNR), USA. We have also

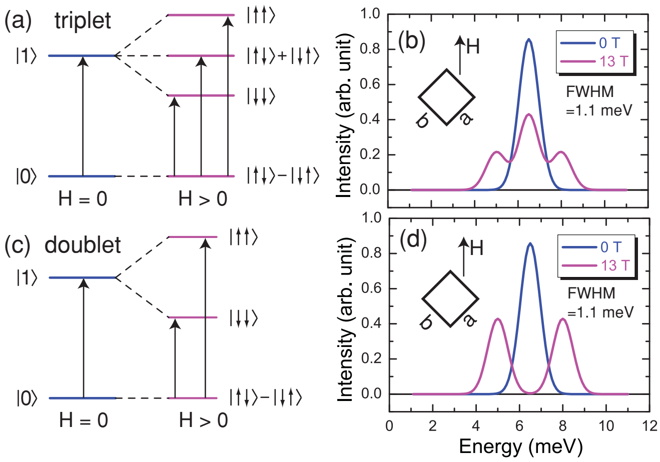

FIG. 1. (Color online) (a) Schematic diagram of the Zeeman splitting of the exciton from singlet $|0\rangle$ to triplet $|1\rangle$ excited states. (b) Schematic diagram of the Zeeman splitting of a spin-1 resonance under 13-T field. The full width at half maximum (FWHM) of the resonance is assumed to be close to the resolutions of our instruments estimated from phonon measurements. The intensity of the central peak is set to be equal to the sum of the other two peaks, which is a requirement of isotropic spin fluctuations. The Landé factor is taken as $g = 2$. (c),(d) Similar schematic diagrams as those in (a) and (b) in the case of a singlet-to-doublet excitation.

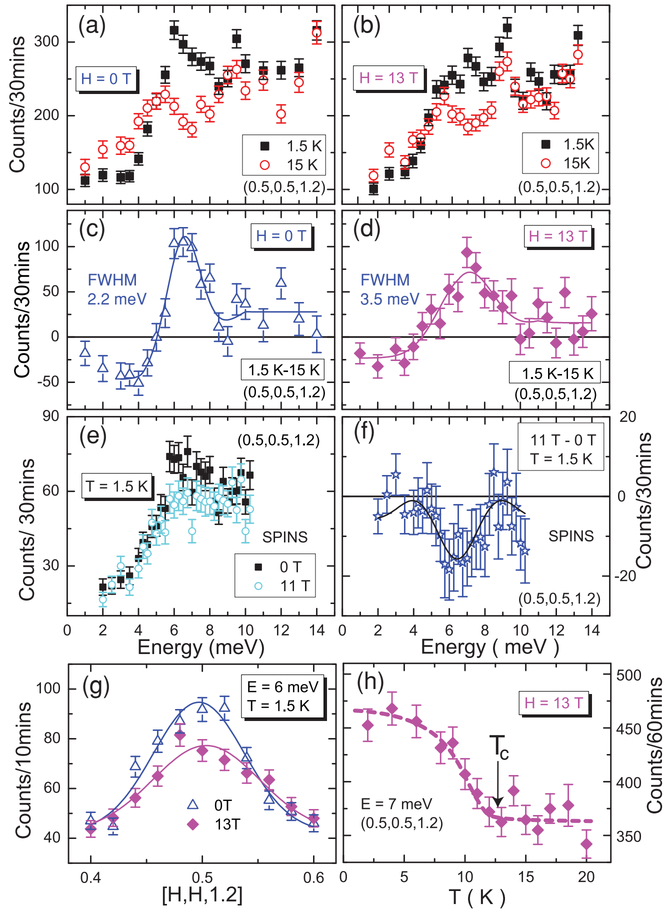

FIG. 2. (Color online) Constant-Q scans at $Q = (0.5,0.5,1.2)$ at 1.5 and 15 K under (a) 0 and (b) 13-T field. (c),(d) Magnetic resonance at 0 and 13 T, respectively, as determined by the difference of data between 1.5 and 15 K. The solid lines are fitted results as described in the text. (e) Constant-Q scans at (0.5,0.5,1.2) at 1.5 K under 0 and 11 T measured at SPINS. (f) Difference of data in (e) between 11 and 0 T. The solid line is fitted by the difference between one and three Gaussian functions as illustrated in Fig. 1(b). (g) Constant-E scans at 6 meV. (h) Temperature dependence of the resonance intensity at peak position ($E = 7$ meV) under 13-T field.

carried out in-plane field measurements on $BaFe_{1.9}Ni_{0.1}As_2$ using the IN22 thermal triple-axis spectrometer at the Institut Laue-Langevin, Grenoble, France.[17] For cold triple-axis measurements, we chose a fixed final neutron energy of $E_f = 5$ meV and put a cooled Be filter before the analyzer. The energy resolution at the elastic line is less than 0.25 meV. Pyrolytic graphite (PG) was used as the monochromator and analyzer. We define the momentum transfer 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), where the lattice parameters of the tetragonal unit cell ($P4/nmm$ space group) are $a = b = 3.786$ Å and $c = 6.061$ Å for $FeSe_{0.4}Te_{0.6}$ and $a = b = 3.963$ Å and $c = 12.77$ Å for $BaFe_{1.9}Ni_{0.1}As_2$. The samples were oriented in the $[H,H,L]$ scattering plane so that the applied field direction is along the $(1,-1,0)$ direction as shown in the inset of Fig. 1(b). The setup for IN22 thermal triple-axis measurements was described before.[17]

III. RESULTS

We first describe the results on $FeSe_{0.4}Te_{0.6}$. Figure 2(a) shows the zero-field constant-Q scans at $Q = (0.5,0.5,1.2)$ at 1.5 and 15 K. While the raw data show some features of unknown origin, the different spectra give rise to the effect induced by the superconductivity. The temperature difference spectrum in Fig. 2(c) shows a clear peak at $E = 6.5 \pm 0.2$ meV consistent with the previous results.[6,8,23] By fitting the peak with a Gaussian on a linear background as shown in Fig. 2(c), we find that the full width half maximum (FHWM) of the resonance is $\Delta E = 2.2 \pm 0.5$ meV. Figure 2(b) shows identical constant-Q scans at $Q = (0.5,0.5,1.2)$ taken under a 13-T in-plane magnetic field. The temperature difference plot in Fig. 2(d) again shows a resonance that becomes broader in energy. A Gaussian fit to the data gives the peak position of $E = 6.9 \pm 0.4$ meV and the FWHM of $\Delta E = 3.5 \pm 0.6$ meV [Fig. 2(d)]. Therefore while a 13-T in-plane magnetic field broadens the resonance, there is no direct evidence for singlet-triplet splitting as suggested in Ref. 26. It is not clear whether such a discrepancy comes from the different field directions applied in these two experiments. In principle, an in-plane magnetic field should offer a better opportunity to observe the field-induced Zeeman effect. We note, however, that the FWHM of the resonance in Ref. 26 (about 5 meV estimated from their data) is much larger than that in our experiment, which may suggest that our samples have a better superconducting quality. A comparison of constant-E scans at $E = 6$ meV in a 0 and 13-T field demonstrates a clear suppression of the resonance intensity under the 13-T field [Fig. 2(g)]. Figure 2(h) shows the temperature dependence of the scattering at $Q = (0.5,0.5,1.2)$ and 13 T, which increases below 12 K instead of 14 K in the zero field, consistent

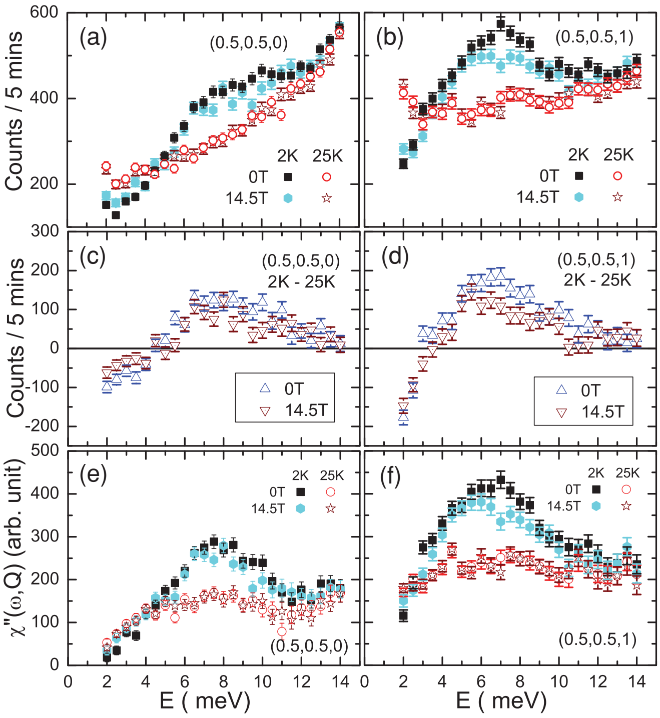

FIG. 3. (Color online) Constant-Q scans at (a) $Q = (0.5,0.5,0)$ and (b) $Q = (0.5,0.5,1)$. The corresponding temperature differences are shown in (c) and (d), while the $\chi''(Q,\omega)$ are shown in (e) and (f).

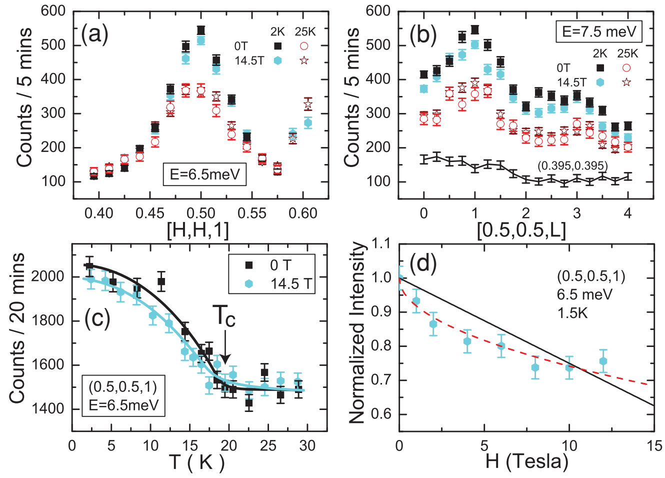

FIG. 4. (Color online) (a) Constant-E scans at 6.5 meV and $L = 1$. (b) L scan at 7.5 meV. (c) Temperature dependence of the resonance peak ($E = 6.5$ meV) intensity at (0.5,0.5,1). The lines are guided by eye. (d) Field dependence of the resonance peak intensity at (0.5,0.5,1). The solid and dashed lines are fitted by the linear and square-root functions, respectively.

with the fact that the in-plane magnetic field also suppresses superconductivity. To further demonstrate that the application of an in-plane magnetic field does not split the resonance, we have also carried out similar measurements on SPINS. Figure 2(e) shows the raw data taken at 0 and 11-T field. The field-on minus field-off data are shown in Fig. 2(f). Inspection of these data again reveal no evidence for Zeeman splitting. However, we note that the broadening of resonance in Fig. 2(d) is consistent with overlapping of three peaks with intrinsic FHWMs much larger than the instrumental resolution. No requirement of a large intrinsic anisotropic field is needed, contrary to the suggestion in Ref. 26.

Having shown that there is no conclusive evidence for the Zeeman splitting of the resonance in $FeSe_{0.4}Te_{0.6}$, we now turn to our in-plane field measurements on $BaFe_{1.9}Ni_{0.1}As_2$. Figures 3(a) and 3(b) show constant-Q scans at $Q = (0.5,0.5,0)$ and $Q = (0.5,0.5,1)$ below and above T_c at 0 and 14.5-T field.[28] In the normal state, a 14.5-T in-plane field has no observable effect on spin excitations at both wave vectors, similar to that of a c-axis field.[17] When cooling the system down to 1.5 K, the field again only has a small effect on the spin excitations. Figures 4(c) and 4(d) show the temperature difference plots in 0 and 14.5-T field. Within the error of our measurements, a 14.5-T field has no observable effect on the resonance at $Q = (0.5,0.5,0)$ [Fig. 3(c)]. At wave vector $Q = (0.5,0.5,1)$, the field slightly suppresses the intensity of the resonance, again with no evidence for the expected Zeeman splitting. Figures 3(e) and 3(f) show our estimated imaginary part of the dynamic susceptibility, $\chi''(Q,\omega)$, at $Q = (0.5,0.5,0)$ and $Q = (0.5,0.5,1)$ below and above T_c in 0 and 14.5-T field, respectively. These results are obtained by subtracting the background and correcting for the Bose population factor. They again suggest no observable field-induced effect at $Q = (0.5,0.5,0)$ and a small suppression of the resonance at $Q = (0.5,0.5,1)$.

Figure 4(a) gives constant-energy scans below and above T_c in 0 and 14.5-T fields along the $[H,H,1]$ direction, the applied field only has limited effect on spin excitations in the superconducting state and no effect in the normal state. Figure 4(b) shows scans long the $[0.5,0.5,L]$ direction which again reveal a weak magnetic-field effect at 2 K. Figure 4(c) plots the temperature dependence of the scattering at the resonance energy. In contrast to the earlier measurements for a c-axis aligned field,[17] the in-plane field has virtually no effect on T_c and only suppresses the intensity of the resonance moderately. Figure 4(d) shows the magnetic-field dependence of the scattering at the resonance energy and $Q = (0.5,0.5,1)$. In Ginzburg-Landau theory, the magnetic-field dependence of the superconducting gap $\Delta(B)$ is related to the zero-field gap $\Delta(0)$ via $\Delta(B)/\Delta(0) = \sqrt{1 - B/B_{c2}}$, where B_{c2} is the upper critical field.[17] Assuming that the intensity of the resonance is associated with the superconducting volume fraction or superfluid density, one would expect the intensity of the mode to decrease linearly with increasing field,[12] or $I/I_0 = 1 - B/B_{c2}$. We have used both linear (solid line) and square root (dash line), where $I/I_0 = 1 - (B/B_{c2})^{1/2}$, relations to fit the field dependence data. The outcome gives the B_{c2} as 40 and 150 T, respectively. While these results suggest that the square-root relationship fits the data better, its physical significance is unclear.

IV. CONCLUSION

In summary, we have studied the effect of an in-plane magnetic field on the neutron spin resonance of $FeSe_{0.4}Te_{0.6}$ and $BaFe_{1.9}Ni_{0.1}As_2$ superconductors. While our initial purpose is to study the Zeeman splitting of the spin-1 triplet of the resonance, we are unable to conclusively establish that the mode is indeed a singlet-triplet excitation. From recent polarized neutron-scattering measurements on $BaFe_{1.9}Ni_{0.1}As_2$,[10] we know that the resonance is inconsistent with a simple isotropic singlet-triplet excitation, and appears only for spin moment parallel to the Fe plane, which may result in a singlet-doublet excitation [Fig. 1(c)]. This is consistent with the present magnetic-field effect, where no field-induced broadening of the mode was observed since the picture in Fig. 1(d) is only applied to the isotropic case. On the other hand, polarized neutron-scattering experiments have found quasi-isotropic spin resonance in $FeSe_{0.5}Te_{0.5}$;[29] this may explain why an in-plane magnetic field of 14.5 T can clearly broaden the resonance (Fig. 2). Although these results may be consistent with the mode being a singlet-triplet excitation in $FeSe_{0.5}Te_{0.5}$, we cannot conclusively establish this based on the present data. Regardless of whether the resonance is a spin-1 singlet-to-triplet excitation, it is directly associated with superfluid density and superconducting volume fraction. Therefore understanding its microscopic origin is still important for determining the role of spin excitations for high-T_c superconductivity.

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