Distinguishing $s^\pm$ and $s^{++}$ electron pairing symmetries by neutron spin resonance in superconducting NaFe$_{0.935}$Co$_{0.045}$As

Chenglin Zhang, H.-F. Li, Yu Song, Yixi Su, Guotai Tan, Tucker Netherton, Caleb Redding, Scott V. Carr, Oleg Sobolev, Astrid Schneidewind, Enrico Faulhaber, L. W. Harriger, Shiliang Li, Xingye Lu, Daoxin Yao, Tannoy Das, A. V. Balatsky, Th. Brückel, J. W. Lynn, and Pengcheng Dai.

1 Department of Physics and Astronomy, Rice University, Houston, Texas 77005, USA
2 Department of Physics and Astronomy, The University of Tennessee, Knoxville, Tennessee 37996-1200, USA
3 Jülich Centre for Neutron Science JCNS, Forschungszentrum Jülich GmbH, Outstation at Institut Laue-Langevin, Boîte Postale 156, F-38042 Grenoble Cedex 9, France
4 Institut für Kristallographie der RWTH Aachen, 52056 Aachen, Germany
5 Jülich Centre for Neutron Science JCNS-FRM II, Forschungszentrum Jülich GmbH, Outstation at FRM II, Lichtenbergrasse 1, D-85747 Garching, Germany
6 Department of Physics, Beijing Normal University, Beijing 100875, China
7 Institut für Physikalische Chemie, Georg-August-Universität Göttingen, Tammannstrasse 6, 37077 Göttingen, Germany
8 Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM-II), TU München, D-85747 Garching, Germany
9 Gemeinsame Forschergruppe HZB - TU Dresden, Helmholtz-Zentrum Berlin für Materialien und Energie, D-14109 Berlin, Germany
10 NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, USA
11 Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China
12 State Key Laboratory of Optoelectronic Materials and Technology, Sun Yat-Sen University, Guangzhou 510275, China
13 Jülich Centre for Neutron Science JCNS and Peter Grünberg Institut PGI, JARA-FIT, Forschungszentrum Jülich GmbH, 52425 Jülich, Germany

A determination of the superconducting (SC) electron pairing symmetry forms the basis for establishing a microscopic mechanism for superconductivity. For iron pnictide superconductors, the $s^\pm$-pairing symmetry theory predicts the presence of a sharp neutron spin resonance at an energy below the sum of the hole and electron SC gap energies ($E \leq 2\Delta$) below $T_c$. On the other hand, the $s^{++}$-pairing symmetry expects a broad spin excitation enhancement at an energy above $2\Delta$ below $T_c$. Although the resonance has been observed in iron pnictide superconductors at an energy above $2\Delta$, the mode has also been interpreted as arising from the $s^{++}$-pairing symmetry with $E \geq 2\Delta$ due to its broad energy width and the large uncertainty in determining the SC gaps. Here we use inelastic neutron scattering to reveal a sharp resonance at $E = 7$ meV in SC NaFe$_{0.935}$Co$_{0.045}$As ($T_c = 18$ K). On warming towards $T_c$, the mode energy hardly softens while its energy width increases rapidly. By comparing with calculated spin-excitation spectra within the $s^\pm$ and $s^{++}$-pairing symmetries, we conclude that the ground-state resonance in NaFe$_{0.935}$Co$_{0.045}$As is only consistent with the $s^\pm$-pairing, and is inconsistent with the $s^{++}$-pairing symmetry.

PACS numbers: 74.25.Ha, 74.70.-b, 78.70.Nx

I. INTRODUCTION

A determination of the superconducting (SC) electron pairing symmetry is an important step to establish a microscopic theory for high-transition temperature (high-$T_c$) superconductivity. Since the discovery of iron pnictide superconductors, a peculiar unconventional pairing state, where superconductivity arises from sign-reversed quasiparticle excitations between the isotropic hole and electron Fermi pockets near the $\Gamma$ and $M$ points, respectively, has been proposed. A consequence of this so-called $s^\pm$-pairing state is that the sign-reversed quasiparticle excitations necessitate a sharp resonance in the spin excitations spectra (termed spin resonance) occurring below the sum of the hole and electron SC gap energies ($E \leq 2\Delta = \Delta_h + \Delta_e$) at the antiferromagnetic (AF) wave vector $Q$ connecting the two Fermi surfaces [inset in Fig. 1(a)] below $T_F$. The experimental discovery of the resonance by neutron scattering in hole and electron-doped BaFe$_2$As$_2$ iron pnictide superconductors and iron chalcogenide Fe(Se,Te) family of materials has provided strong evidence for the $s^\pm$-pairing symmetry. However, the neutron scattering experiments on single crystals of Ba$_{1-x}$K$_x$Fe$_2$As$_2$, BaFe$_{2-x}$Co$_x$As$_2$, and Fe(Se,Te) superconductors have also revealed that the resonance is rather broad in energy [Fig. 1(e)]. In addition, the SC gap energies $2\Delta$ determined from the angle resolved photoemission (ARPES) experiments for hole and electron-doped BaFe$_2$As$_2$ superconductors by different groups can differ dramatically for even...
the same material, ranging from below to above the neutron spin resonance energy [Fig. 1(e)], and these values can also be quite different from those estimated by specific heat and penetration depth measurement. Because the superconducting gap values for the multi-band iron pnictide superconductors are different for different bands, the resonance energy is determined by the superconducting gaps in electron and hole bands contributing most to the quasiparticle nesting condition. Therefore, the resonance in some materials may be broad in energy, and this has allowed some workers to argue that superconductivity in iron pnictides arises from orbital fluctuation mediated $s^{++}$-pairing superconductivity, where one expects a broad spin excitation enhancement (neutron spin resonance) at an energy of $E \geq 2\Delta$ below $T_c$. Given the current debates concerning the universality of the electron pairing symmetry in iron-based superconductor, it is important to compare spin excitations in different classes of iron superconductors and determine if the resonance in these materials agrees with predictions of the $s^{\pm}$- and $s^{++}$-pairing symmetries.

In this article, we present inelastic neutron scattering results on single crystals of SC NaFe$_{0.935}$Co$_{0.045}$As with $T_c = 18$ K [Figs. 1(a)-1(d)]. In the normal state, the imaginary part of the dynamic susceptibility, $\chi''(Q, E)$, at the AF wave vector increases linearly with increasing energy $E$. Upon entering into the low-temperature SC state, a spin gap opens below 5.5 meV and a sharp neutron spin resonance appears at $E = 7$ meV with an energy width of $1.5 \pm 0.1$ meV [Figs. 2 (d) and 2(f)]. On warming to $T_c$, the resonance broadens in energy width but its central position does not follow the decreasing SC gap energy, different from the earlier work on electron-doped BaFe$_2$As$_2$. By comparing the neutron scattering results with a random-phase approximation (RPA) spin-susceptibility calculation within the $s^{\pm}$- and $s^{++}$-pairings, we find that our data are consistent with the $s^{\pm}$-symmetry in the low-temperature SC state and cannot be explained by the $s^{++}$ pairing symmetry.

II. RESULTS

We carried out inelastic neutron scattering experiments on the thermal (PUMA) and cold (PANDA) triple-axis spectrometers at the FRM-II, TU Munich, Germany and also on the SPINS cold triple-axis spectrometer at NIST Center for Neutron Research, Gaithersburg, Maryland. For the experiments, we coalesged 5 pieces of self-flux grown NaFe$_{1-x}$Co$_x$As single crystals with a total mass of 5.5 g (mosaic about 3°) for PUMA and PANDA experiment. Samples are loaded inside a 4 K displux controlled by a calibrated temperature sensor.

For SPINS measurements, we used ~10 g of single crystals with mosaic less than 3°. The chemical compositions of the samples are determined as Na$_{1.06}$Fe$_{0.935}$Co$_{0.045}$As by inductively coupled plasma atomic-emission spectroscopy, which has an accuracy of about 2%. Samples from different batches show almost identical chemical composition, which we denote as NaFe$_{0.935}$Co$_{0.045}$As. The wave vector $Q$ at $q_x = q_y = q_z$ is in Å$^{-1}$ is defined as $(H,K,L) = (q_x/2\pi, q_y/2\pi, q_z/2\pi)$ reciprocal lattice unit (r.l.u) using the tetragonal unit cell (space group P4/nmm, $a = 3.921$ Å, $c = 6.911$ Å at 5 K). We used focusing pyrolytic graphite (PG) monochromator and analyzer with fixed final energies of $E_f = 14.7$ meV and $E_f = 5$ meV at PUMA and PANDA, respectively. For SPINS measurements, we used $E_f = 5$ meV with flat monochromator and analyzer. Both the $[H, H, L]$ and $[H, K, 0]$ scattering zones have been used in the experi-

FIG. 1: (Color online) Phase diagram, Fermi surfaces, reciprocal space, transport, and susceptibility measurements. (a) The electronic phase diagram of NaFe$_{1-x}$Co$_x$As, where the arrow indicates the Co-doping level of our samples. Inset shows the hole and electron Fermi pockets near $\Gamma$ and $M$ positions, respectively (Ref. [34]). (b) Reciprocal space probed in the present experiment. The scan directions are shown as arrows. (c) Temperature dependence of the in-plane resistivity $\rho_{ab}$ in NaFe$_{0.935}$Co$_{0.045}$As. The inset displays the low-T resistivity measured in zero-field and 9 T. (d) The temperature dependence of the bulk susceptibility measured by DC magnetic susceptibility. (e) The schematics of neutron spin resonance for optimally hole (Ref. [34], red solid line) and electron (Ref. [34], blue solid line) iron pnictide superconductors. The green solid line shows results for FeTe$_{0.8}$Se$_{0.2}$ superconductors and black solid lines show results from the present work on NaFe$_{0.935}$Co$_{0.045}$As. The red, blue, and black dashed regions show the range of $2\Delta$ as determined from different ARPES and other experiments. In all cases, the resonance can be either below or above the $2\Delta$. 
ments and the scan directions are marked in Fig. 1(b). To characterize the samples, we have carried out resistivity and DC magnetic susceptibility measurements using commercial physical property measurement system and SQUID magnetometer. Based on the early neutron diffraction measurements, AF Bragg peaks and low-energy spin excitations are expected to occur around the (0, 5, 0, 5, L) positions with L = 0.5, 1.5, ∙∙∙ [Fig. 1(b)].

Figure 1(c) plots the in-plane resistivity $\rho_{ab}$ measurement at zero field which gives $T_c = 18$ K. The inset shows the magnetic field dependence of $\rho_{ab}$ at 0 and 9-T, indicating a field-induced $T_c$ suppression of ∼2 K. Figure 1(d) shows the magnetic susceptibility measurements on the sample again showing a $T_c = 18$ K. Given the known electronic phase diagrams of NaFe$_{1-x}$Co$_x$As$_2$, it is clear that our NaFe$_{0.935}$Co$_{0.05}$As samples are in the slightly overdoped regime and do not have static AF order co-existing with superconductivity [Figs. 1(a)]. Our elastic neutron diffraction scans through the AF Bragg peak positions are featureless and thus confirm this conclusion (see Fig. 7 in the appendix for details). Figure 1(e) shows schematics of the resonance for optimally hole doped iron pnictides, as well as iron chalcogenide Fe(Se,Te) family of materials as determined from ARPES and other techniques. For comparison, we also show the SC gaps determined from ARPES for NaFe$_{0.95}$Co$_{0.05}$As. As we can see, the ARPES measurements from two groups on NaFe$_{0.95}$Co$_{0.05}$As have yielded SC gaps different by a factor of two.

In previous neutron scattering work on electron doped BaFe$_{2-x}$Ni$_x$As$_2$ pnictides superconductors, the neutron spin resonance was found to be dispersive, occurring at slightly different energies for different c-axis wave vector transfers. To see if this is also the case for spin excitations in NaFe$_{0.935}$Co$_{0.05}$As, we carried out constant-Q scans at wave vectors $Q = (0.5, 0.5, 1)$ and (0.5, 0.5, 1.5) below and above $T_c$ on PUMA. While the background scattering (BKG) taken at $Q = (0.65, 0.65, 1)$ and (0.65, 0.65, 1.5) showed no change below and above $T_c$ [Figs. 2(a) and 2(c)], the scattering at the in-plane AF wave vector revealed dramatic changes across $T_c$. In the normal state ($T = 25$ K), the scattering above BKG is featureless and increases with increasing energy. Upon entering into the SC state ($T = 5$ K), a spin gap forms below ~5.5 meV and a sharp resonance appears at $E = 7$ meV [Figs. 2(a) and 2(c)]. The corresponding $\chi''(Q, E)$, obtained by subtracting the BKG and correcting for the Bose population factors using $\chi''(Q, E) = [1 - \exp(-E/k_B T)]S(Q, E)$, are shown in Figs. 2(b) and 2(d). Inspection of Figs. 2(a-2(d) reveals that the resonance exhibits no c-axis dispersion and has an energy width of ~3 meV. Figures 2(e) and 2(f) show similar scans on SPINS, which reveal a 5 meV spin gap and a sharp resonance at $E = 6.8 \pm 0.1$ meV with an energy width of ~1.5 meV in the SC state at 3 K. This is much narrower than the energy widths of the resonances in the hole and electron-doped BaFe$_2$As$_2$ [Fig. 1(e)]

On warming to 10, 15, and 17 K, the resonance energy widths become broader, but its peak position remains almost unchanged up to $T = 0.94T_c = 17$ K [Fig. 2(f)]. This is different from earlier work on the temperature dependence of the resonance energy in BaFe$_{1.85}$Co$_{0.15}$As$_2$ ($T_c = 25$ K) but somewhat similar to the temperature dependence of the resonance in Fe(Se,Te) family of materials.

To confirm the SC spin gap and determine the wave vector dependence of the resonance, we carried out constant-energy scans at $E = 4$, 7, and 15 meV below and above $T_c$. Figures 3(a-c) and 3(d-f) show $\chi''(Q, E)$ along the $[H, H, 1]$ and $[H, H, 1.5]$ directions, respectively. In the SC state at $T = 5$ K, $\chi''(Q, E)$ is featureless at $E = 4$ meV and thus confirms the presence of a spin gap. For other excitation energies, the scattering profiles can be fitted by Gaussians on linear BKG. Taking the Fourier transforms of the fitted Gaussian peaks along the $[H, H, 1]$ direction, we find that the in-plane spin-spin correlation lengths at the energy of the resonance are
La = 24 ± 1 Å at 25 K and ξ = 30 ± 1 Å at 5 K. Along the [H, H, 1.5] direction, ξ = 32 ± 3 Å are unchanged from 25 K to 5 K. Increasing the excitation energy to E = 15 meV (> 2Δ), there are no observable differences in scattering intensity and spin correlation lengths (ξ = 20 ± 1 Å) below and above Tc.

Figure 4(a) shows the temperature dependence of the scattering at the AF wave vector Q = (0.5, 0.5, 1.5) for the resonance (E = 7 meV) and spin gap (E = 4 meV) energies. While the intensity increases dramatically below Tc at the resonance energy, it decreases at E = 4 meV signaling the opening of a SC spin gap. To test if spin excitations in NaFe0.935Co0.045As are indeed two-dimensional in reciprocal space like in the case of CdO-doped BaFe2As2, we show in Fig. 4(b) BKG subtracted constant-energy scans along the [0.5, 0.5, L] direction at the resonance energy (E = 7 meV) below and above Tc. The monotonous decrease of the scattering with increasing L is consistent with the square of the Fe2+ magnetic form factor, thus confirming the two-dimensional and magnetic nature of the resonance.

In the optimally hole-doped BaFe2As2 and electron-doped BaFe2As2, the resonances form longitudinally and transversely elongated ellipses, respectively. Since NaFe0.935Co0.045As belongs to the electron-doped iron pnictide superconductor, we mapped out spin excitations in the [H, K, 0] scattering plane on PUMA. Figures 4(c) and 4(d) show constant-energy scans at E = 7 meV along the transverse [1/2 + H, 1/2 − H, 0] and longitudinal [H, H, 0] directions [perpendicular and parallel to the in-plane AF ordering wave vector Q = (0.5, 0.5, 0)] respectively, below and above Tc. Although there is no evidence for transverse incommensurate magnetic scattering as in the case of LiFeAs, the spin resonance in NaFe0.935Co0.045As is considerably broader along the transverse direction than that of the longitudinal direction. Figure 4(g) shows the two-dimensional image of the resonance in the SC state, further confirming the results of Fig. 4(c) and 4(d).

Figure 4(e) and 4(f) summarizes the full-width-half-maximum (FWHM) of the low-energy spin excitations along the longitudinal and transverse directions below and above Tc, respectively. In the normal state, spin excitations in NaFe0.935Co0.045As are gapless, comparing with the ~10 meV anisotropy gap in spin waves of the undoped NaFeAs. The data points are the FWHM of spin excitations along the two high symmetry directions. On cooling to below Tc, the effect of superconductivity is
between band indices and appendix. Results for the five-orbital tight-binding model taken from Ref. 48 are essentially unaffected by superconductivity.

III. DISCUSSION AND CONCLUSIONS

To compare with the experiment, we have performed RPA spin-susceptibility calculation in the SC state, using the five-orbital tight-binding model taken from Ref. 48. The details of the calculations can be found in Refs. 49 and appendix. Results for the \( s^\pm \) and \( s^{++} \) pairing symmetries are given in Figs. 5(a)-5(c). For \( s^\pm \)-pairing, a spin-resonance appears due to the inelastic broadening of 1 meV, SC gap of \( \Delta_0 = 6 \text{ meV} \), and Coulomb interaction \( U = 1.6 \text{ eV} \).

Contrasting \( s^\pm \)-pairing due to self-consistently evaluated change in the Fermi level (shown in the inset) at each temperature, while that for the \( s^{++} \) pairing follows the gap function at each temperature. (c) Spin-resonance peak and intensity as a function of energy at the commensurate wave vector at different temperatures (see the colorscale) for \( s^\pm \) symmetry. (d) Same as (c) but for the \( s^{++} \) pairing symmetry. Contrasting evolution of the resonance energy and intensity can be marked between \( s^\pm \) and \( s^{++} \) pairing, and our experimental data are consistent with the former pairing symmetry results.

Fig. 2 are only consistent with \( s^\pm \)-pairing symmetry regardless of the actual values of the SC gaps.

On the other hand, assuming that temperature dependence of the SC gaps in NaFe\(_{0.935}\)Co\(_{0.045}\) follows the BCS form, as demonstrated in many pnictide superconductors, we observe that the resonance energy remains very much temperature-independent except near \( T_c \) due to self-consistently evaluated change in the Fermi level. To explain this behavior, we calculate the temperature evolution of the spin-susceptibility and the results are shown in Fig. 6(b-d). For each temperature, we evaluate the Fermi energy \( E_F \) constrained by the fixed number of electrons. As shown in inset to Fig. 6(b), \( E_F \) increases as gap decreases with temperature. The corresponding change in the Fermi surface topology yields a change in the effective gap value for \( s^\pm \) pairing due to the anisotropic gap structure factor \( g(k) \) defined before.
pairing state, and for the temperature independent interaction $U$, the resonance condition shifts to a higher energy, as illustrated in the appendix. Due to the interplay between these two changes as a function of temperature, the resonance energy shifts to higher energy compared to its corresponding $2\Delta(T)$ value, but remains very much temperature-independent with respects to its $2\Delta(0)$ energy [Fig. 6(a)]. This analysis does not apply to the $s^{\pm}$ pairing symmetry since it is insensitive to the change in the underlying Fermi surface. Therefore, the resonance energy in this pairing symmetry, which lies above $2\Delta(T)$, very much follows the gap function.

The corresponding lineshape of the resonance is shown in Figs. 6(c) and 6(d) for $s^\pm$ and $s^{++}$, respectively. Contrasting temperature evolution of $\chi''$ at the commensurate wave vector can be marked between the two pairing symmetries. For $s^\pm$ pairing, the resonance intensity decreases according to the BCS prediction, but its position does not shift in energy until the temperature reaches the vicinity of $T_c$, where the resonance shifts to lower energy below its corresponding $2\Delta(T)$. On the contrary, for $s^{++}$ pairing the resonance intensity increases and gradually shifts to lower energy. This contrasting behavior can be understood from the physics of resonance described in the appendix. As shown in Fig. 8(b), the weak intensity hump above the $2\Delta$ in the $s^{++}$ arise from the discontinuous jump from the particle-hole channel to the SC region. As gap decreases, the energy scale of the jump also decreases, and thus the resonance energy follows gap evolution, and remains almost insensitive to the modification in the underlying electronic structure.

In summary, we have discovered a sharp resonance in electron-overdoped NaFe$_{0.935}$Co$_{0.045}$As in the low-temperature SC state. Our experimental data is consistent with the $s^\pm$ pairing state both in terms of intensity and energy scales as a function of energy, and thus provide further evidence to the presence of $s^\pm$ pairing symmetry in pnictide superconductors.

IV. ACKNOWLEDGEMENTS

The single crystal growth efforts and neutron scattering work at UT/Rice are supported by the US DOE, BES, through contract DE-FG02-05ER46202. Work at IOP is supported by MOST (973 Project: 2012CB82400). The work at JCNS and RWTH Aachen University was partially funded by the BMBF under contract No. 05K10PA3. Work at LANL was supported by the NNSA of the US DOE under contract DE-AC52-06NA25396.

V. APPENDIX

In this appendix, we first present elastic neutron diffraction data on the sample and then give the details of our calculations presented in the main text. The elastic neutron scattering scans across the AF Bragg peak positions are featureless and thus confirm this conclusion that the material does not have static AF order.

We evaluate the spin-resonance susceptibility in the superconducting (SC) state within the BCS-RPA formalism, which is given by

$$\chi_{BCS}^{rstu}(q, \omega) = \frac{1}{N} \sum M^{nu}_{rstu}(k, q) \left\{ \frac{1}{2} \left[ 1 + \frac{\xi_k^\nu \xi'_k + \Delta_k^\nu \Delta'_k}{S_k^\nu S_k^{\nu'}} f(S_k^\nu f(S'_k) - f(S'_k) f(S_k^\nu) S_k^\nu S_k^{\nu'} + 4i\delta \right) \right\}$$

Here $S_k^\nu = [(\xi_k^\nu)^2 + (\Delta_k^\nu)^2]^{1/2}$ is the SC quasiparticle state for the eigenstate $\xi_k^\nu$ ($\nu$ is band index) and the SC gap $\Delta_k^\nu$. For $s^\pm$-pairing symmetry we have $\Delta_k^\nu = \Delta_k^\nu = (\cos k_x + \cos k_y)$ and for $s^{++}$-wave pairing symmetry we take $\Delta_k^\nu = \Delta_k^\nu$ to be same for all bands. $M$ is the matrix element consists of the eigenstates of the initial and final scattered quasiparticle state as given by $M^{nu}_{rstu}(k, q) = \phi_{r}^{-}(k + q)\phi_{s}^{-}(k)\phi_{t}^{+}(k)\phi_{u}^{+}(k + q)$, where $\phi_{r}^{-}(k)$ is the eigenfunction of the $\nu^{th}$ band derived from $r^{th}$-orbital.

In Eq. (1) in the appendix, the first term is called particle-hole scattering term which vanishes in the SC state due to particle-hole symmetry regardless of any pairing symmetry. The second and third term are called particle-particle and hole-hole scatterings, respectively which become active in the SC state. Focusing on the third term (the same analysis applies to the second term), we find that this term contributes a non-zero value only when $\text{sign}(|\Delta_k^\nu|) \neq \text{sign}(|\Delta_{k+q}^\nu|)$. Since $\xi_k^\nu = 0$ on the Fermi surface. A pole thus obtained in the imaginary part of $\chi_{BCS}$ as

$$E = |\Delta_k^\nu| + |\Delta_{k+q}^\nu|.$$
inside the SC region, which virtually washes out the
considerably broad and is obtained from the dynamical nesting between different bands as $E_q = \xi_k' - \xi_k''$. For $s^\pm$-pairing, $\chi_{BCS}$ gives a very sharp resonance feature inside the SC region, which virtually washes out the in-
formation of the p-h continuum above this energy scale. On the other hand, for $s^{++}$-pairing, $\chi_{BCS} = 0$ and the sharp crossover to $\chi_0$ at $E = 2\Delta$ acts as an abrupt change in the real part of $\chi_0$. In both cases, the Fermi surface nesting between electron-pockets sitting at M point and the hole-pockets at Γ-point is most dominant, giving features at the commensurate wave vector $Q = (\pi, \pi)$. The characteristic differences in the bare susceptibilities for the two pairing channels are given in the supplementary Fig. 2.

We include the many-body correction to the spin-excitation spectrum via standard RPA formalism as $\tilde{\chi}_{RPA} = \chi_{BCS}/(1 - \tilde{U}\chi_{BCS})$, where the symbol tilde over a quantity represents that it is a matrix in the orbital basis. The interaction vertex consists of intra-orbital Coulomb interaction $U$, inter-orbital term $V = 2U' - J$, where $U' = U - 5/4J$ and $J = U/8$ is the Hund’s coupling and $J' = J/2$ is the pair hopping term. For a single band system, a spin-resonance occurs when the denominator of RPS susceptibility vanishes at $\chi_{BCS} = 1/U$, and the peak is broadened by $\chi_{BCS}''$. The situation is equivalent, yet complicated in the multi-orbital case. We obtain a sharp resonance peak for the $s^\pm$-pairing case below $2\Delta$ where the broadening associated with $\chi_{BCS}''$ is much reduced, as shown in supplementary Fig. 1(a). On the other hand, for $s^{++}$ the resonance in the p-h continuum is broadened by large value of $\chi_0$ [see green dashed line in supplementary Fig. 1(b)].

* Electronic address: pdai@rice.edu

1 C. C. Tsuei, J. R. Kirtley, Rev. Mod. Phys. 72, 969 (2000).
2 Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, J. Am. Chem. Soc. 130, 3296 (2008).
3 M. Rotter, M. Tegel, and D. Johrendt, Phys. Rev. Lett. 101, 107006 (2008).
4 C. W. Chu, F. Chen, M. Gooch, A.M. Guloy, B. Lorenz, B. Liv, K. Sasmal, Z. J. Tang, J. H. Tapp, Y. Y. Xue, Physica C 469, 326 (2009).
5 I. I. Mazin, D. J. Singh, M. D. Johannes, and M. H. Du, Phys. Rev. Lett. 101, 057003 (2008).
6 K. Kuroki, S. Onari, R. Arita, H. Usui, Y. Tanaka, H. Kontani, and H. Aoki, Phys. Rev. Lett. 101, 087004 (2008).
7 K. Seo, B. A. Bernevig, J. P. Hu, Phys. Rev. Lett. 101, 206404 (2008).
8 M. M. Korshunov and I. Eremin, Phys. Rev. B 78, 140509(R) (2008).
9 T. A. Maier and D. J. Scalapino, Phys. Rev. B 78, 020514(R) (2008).
10 A. D. Christianson, E. A. Goremychkin, R. Osborn, S. Rosenkranz, M. D. Lumsden, C. D. Malliakas, I. S. Todorov, H. Claus, D. Y. Chung, M. G. Kanatzidis, R. I. Bewley, and T. Guidi, Nature 456, 930 (2008).
11 Chenglin Zhang, Meng Wang, Huiqian Luo, Miaoyin Wang, Mengshu Liu, Jun Zhao, D. L. Abernathy, T. A. Maier, Karol Marty, M. D. Lumsden, Songxue Chi, Sung Chang, Jose A. Rodriguez-Rivera, J. W. Lynn, Tao Xiang, Jiangping Hu, and Pengcheng Dai, Scientific Reports 1, 115 (2011).
12 J.-P. Castellan, S. Rosenkranz, E. A. Goremychkin, D. Y. Chung, I. S. Todorov, M. G. Kanatzidis, I. Eremin, J. Knolle, A. V. Chubukov, S. Maiti, M. R. Norman, F. Weber, H. Claus, T. Guidi, R. I. Bewley, and R. Osborn, Phys. Rev. Lett. 107, 177003 (2011).
13 M. D. Lumsden1, A. D. Christianson, D. Parshall, M. B. Stone, S. E. Nagler, G. J. MacDougall, H. A. Mook, K. Lokshin, T. Egami, D. L. Abernathy, E. A. Goremychkin, R. Osborn, M. A. McGuire, A. S. Safat, R. Jin, B. C. Sales, and D. Mandrus, Phys. Rev. Lett. 102, 107005 (2009).
14 Songxue Chi, Astrid Schneidewind, Jun Zhao, Leland W. Harriger, Linjun Li, Yongkang Luo, Guanghan Cao, Zhu’an Xu, Micheal Loewenhaupt, Jiangping Hu, and Pengcheng Dai, Phys. Rev. Lett. 102, 107006 (2009).
15 D. S. Inosov, J. T. Park, P. Bourges, D. L. Sun, Y. Sidis, A. Schneidewind, K. Hradil, D. Hradil, D. Haug, C. T. Lin, B. Keimer, and V. Hinkov, Nat. Phys. 6, 178 (2010).
16 C. Lester, Jinn-Haw Chu, J. G. Analytis, T. G. Perring, I. R. Fisher, and S. M. Hayden, Phys. Rev. B 81, 064505 (2010).
17 J. T. Park, D. S. Inosov, A. Yaresko, S. Graser, D. L. Sun, Ph. Bourges, Y. Sidis, Yuan Li, J.-H. Kim, D. Haug, A. Ivanov, K. Hradil, A. Schneidewind, P. Link, E. Faulhaber, I. Glavatskyy, C. T. Lin, B. Keimer, and V. Hinkov, Phys. Rev. B 82, 143503 (2010).
FIG. 8: (a) Various components of the spin-susceptibility are plotted together for $s^\pm$-pairing. The RPA value of the susceptibility (red line) is very sharp (of the order of 100 eV$^{-1}$), and thus we divide it by 30 to fit it into the same figure visualization. For the same reason, the real part of the bare susceptibility $\chi$ or visualization. For the same reason, the real part of the $\chi_{BCS}$ susceptibility is multiplied by 2. (b) Same as (a) but for $s^{++}$-pairing. Here $\chi_{RPA}$ is divided by 20 and $\chi_{BCS}$ is multiplied by 2. The horizontal dashed line gives 1/$U$, which illustrated the occurrence of the resonance mode when it cuts through $\chi_{BCS}$. Finally, the vertical dashed line marks the $2\Delta$ line which separates the SC scattering region to the p-h continuum.

18 Jun Zhao, Louis-Pierre Regnault, Chenglin Zhang, Misoying Wang, Zhengcai Li, Fang Zhou, Zhongxian Zhao, Chen Fang, Jiangping Hu, and Pengcheng Dai, Phys. Rev. B 81, 180505(R) (2010).

19 Mengshu Liu, Leland W. Harriger, Huiqian Luo, Meng Wang, R. A. Ewings, T. Guidi, Hyowon Park, Kristjan Haule, Gabriel Kotliar, S. M. Hayden, and Pengcheng Dai, Nat. Phys. 8, 376-381 (2012).

20 D. S. Inosov, J. T. Park, A. Charnukha, Yuan Li, A. V. Boris, B. Keimer, and V. Hinkov, Phys. Rev. B 83, 214520 (2011).

21 Mengshu Dai, Jiangping Hu, and Elbio Dagotto, Nat. Phys. 8, 709-718 (2012).

22 H. A. Mook, M. D. Lumsden, A. D. Christianson, S. E. Nagler, Brian C. Sales, Rongying Jin, Michael A. McGuire, Athena S. Sefat, Danny S. Inosov, J. T. Park, G. Friemel, T. Loew, V. Hinkov, Yuan Li, B. Keimer, and V. Hinkov, Phys. Rev. Lett. 104, 187002 (2010).

23 Yiming Qiu, Wei Bao, Y. Zhao, Collin Broholm, V. Stanev, Z. Tesanovic, Y. C. Gasparovic, S. Chang, Jin Hu, Bin Qian, Minghu Fang, and Zhiqiang Mao, Phys. Rev. Lett. 103, 067008 (2009).

24 Leland W. Harriger, O. J. Lipscombe, Chenglin Zhang, Huiqian Luo, Meng Wang, Karol Marty, M. D. Lumsden, and Pengcheng Dai, Phys. Rev. B 85, 054511 (2012).

25 P. Richard, T. Sato, K. Nakayama, T. Takahashi, and H. Deng, Report on Progress in Physics 74, 124512 (2011).

26 K. Terashima, Y. Sekiba, J. H. Bowen, K. Nakayama, T. Kawahara, T. Sato, P. Richard, Y.-M. Xu, L. J. Li, G. H. Cao, Z.-A. Xu, H. Ding, and T. Takahashi, PNAS 106, 7330 (2009).

27 D. V. Evtushinsky, D. S. Inosov, V. B. Zabolotnyy, A. Koitzsch, M. Knupfer, B. Büchner, M. S. Viazovska, G. L. Sun, V. Hinkov, A. V. Boris, C. T. Lin, B. Keimer, A. Varykhalov, A. A. Kordyuk, and S. V. Borisenko, Phys. Rev. B 79, 054517 (2009).

28 W. Malaeb, T. Shimojima, Y. Ishida, K. Okazaki, Y. Ota, K. Ohgushi, K. Kihou, T. Saito, C. H. Lee, S. Ishida, M. Nakajima, S. Uchida, H. Fukazawa, Y. Kohori, A. Iyo, H. Eisaki, C.-T. Chen, S. Watanabe, H. Ikeda, and S. Shin, Phys. Rev. B 86, 165117 (2012).

29 F. Hardy, P. Burger, T. Wolf, R. A. Fisher, P. Schweiss, P. Adelmann, R. Heid, R. Fromknecht, R. Eder, D. Ernst, H. v. Löhneysen, and C. Meingast, Europhys. Lett. 91, 47008 (2010).

30 Lan Luan, Thomas M. Lippman, Clifford W. Hicks, Julie A. Bert, Ophir M. Auslaender, Jiu-Hwao Chiu, James G. Analytis, Ian R. Fisher, and Kathryn A. Moler, Phys. Rev. Lett. 106, 067001 (2011).

31 S. Onari, H. Kontani, and M. Sato, Phys. Rev. B 81, 060504(R) (2010).

32 S. Onari and H. Kontani, Phys. Rev. B 84, 144518 (2011).

33 Y. Nagai and K. Kuroki, Phys. Rev. B 83, 220516(R) (2011).

34 Sergey V. Borisenko, Volodymyr B. Zabolotnyy, Alexnader A. Kordyuk, Danil V. Evtushinsky, Timur K. Kim, Igor V. Morozov, Rolf Follath, and Bernd Büchner, Symmetry 4, 251-264 (2012).

35 P. J. Hirschfeld, M. M. Korshunov, and I. I. Mazin, Rep. Prog. Phys. 74, 124508 (2011).

36 A. Chubukov, Annu. Rev. Condens.Matter Phys. 3, 57 (2012).

37 J. P. Hu, Phys. Rev. X 3, 031004 (2013).

38 R. M. Fernandes and A. J. Millis, Phys. Rev. Lett. 110, 117004 (2013).

39 E. Dagotto, Rev. Mod. Phys. 85, 849 (2013).

40 Z.-H. Liu, P. Richard, K. Nakayama, G.-F. Chen, S. Dong, J.-B. He, D.-M. Wang, T.-L. Xia, K. Umezawa, T. Kawahara, S. Souma, T. Sato, T. Takahashi, T. Qian, Yaobo Huang, Nan Xu, Yingbo Shi, H. Ding, and S.-C. Wang, Phys. Rev. B 84, 064519 (2011).

41 S. Thirupathaiah, D. V. Evtushinsky, J. Maletz, V. B. Zabolotnyy, A. A. Kordyuk, T. K. Kim, S. Wurmehl, M. Roslova, I. Morozov, B. Büchner, and S. V. Borisenko, Phys. Rev. B 86, 214508 (2012).

42 Shiliang Li, Clarina de la Cruz, Q. Huang, G. F. Chen, T.-L. Xia, J. L. Luo, N. L. Wang, and Pengcheng Dai, Phys. Rev. B 80, 020504(R) (2009).

43 Dinah R. Parker, Matthew J. P. Smith, Tom Lancaster, Andrew J. Steele, Isabel Franke, Peter J. Baker, Francis L. Pratt, Michael J. Pitcher, Stephen J. Blundell, and Simon J. Clarke, Phys. Rev. Lett. 104, 057007 (2010).

44 A. F. Wang, X. G. Luo, Y. J. Yan, J. J. Ying, Z. J. Xiang, G. J. Ye, P. Cheng, Z. Y. Li, W. J. Hu, and X. H. Chen, Phys. Rev. B 85, 224521 (2012).

45 Guotai Tan, Ping Zheng, Xiancheng Wang, Yanchao Chen, Xiaotian Zhang, Jianlin Luo, Tucker Netherton, Yu Song, Pengcheng Dai, Chenglin Zhang, and Shiliang Li Phys. Rev. B 87, 144512 (2013).

46 N. Qureshi, P. Steffens, Y. Drees, A. C. Komarek, D. Lamago, Y. Sidis, L. Harnagea, H.-J. Grafe, S. Wurmehl, B. Büchner, and M. Braden, Phys. Rev. Lett. 108, 117001 (2012).

47 J. T. Park, G. Friemel, T. Loew, V. Hinkov, Yuan Li, B. H. Min, D. L. Sun, A. Ivanov, A. Piovano, C. T. Lin, B. Keimer, Y. S. Kwon, and D. S. Inosov, Phys. Rev. B 86, 024437 (2012).

48 S Graser, T A Maier, P J Hirschfeld, and D J Scalapino, New J. Phys. 11, 025016 (2009).

49 T. Das and A. V. Balatsky, Phys. Rev. Lett. 106, 157004 (2011).
T. A. Maier, S. Graser, D. J. Scalapino, P. Hirschfeld, Phys. Rev. B 79, 134520 (2009).