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## Neutron spin resonance as a probe of superconducting gap anisotropy in partially detwinned electron underdoped NaFe<sub>0.985</sub>Co<sub>0.015</sub>As

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We use inelastic neutron scattering (INS) to study the spin excitations in partially detwinned NaFe<sub>0.985</sub>Co<sub>0.015</sub>As which has coexisting static antiferromagnetic (AF) order and superconductivity ( $T_c = 15 \text{ K}, T_N = 30 \text{ K}$ ). In previous INS work on a twinned sample, spin excitations form a dispersive sharp resonance near  $E_{r1} = 3.25 \text{ meV}$  and a broad dispersionless mode at  $E_{r1} = 6 \text{ meV}$  at the AF ordering wave vector  $\mathbf{Q}_{AF} = \mathbf{Q}_1 = (1, 0)$  and its twinned domain  $\mathbf{Q}_2 = (0, 1)$ . For partially detwinned NaFe<sub>0.985</sub>Co<sub>0.015</sub>As with the static AF order mostly occurring at  $\mathbf{Q}_{AF} = (1, 0)$ , we still find a double resonance at both wave vectors with similar intensity. Since spin rotational symmetry is explicitly broken at  $\mathbf{Q}_1 = (1, 0)$ , these results indicate that the double resonance cannot be due to the static and fluctuating AF orders, but originate from the superconducting gap anisotropy.

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The neutron spin resonance is a collective magnetic excitation observed by inelastic neutron scattering (INS) at the antiferromagnetic (AF) ordering wave vector of unconventional superconductors below  $T_c^{1-4}$ . First discovered in the optimally hole-doped  $YBa_2Cu_3O_{6+x}$  family of copper oxide superconductors<sup>1</sup>, the mode was also found in iron pnictide superconductors at the AF wave vector  $\mathbf{Q}_{AF} = (1,0)$  in reciprocal space [Figs. 1(a) and 1(b)<sup>5-16</sup>, and is considered one of the hall marks of unconventional superconductivity<sup>17</sup>. Experimentally, the neutron spin resonance appears as an enhancement of the magnetic spectral weight at an energy  $E_r$  in the superconducting state at the expense of normal state spin excitations for energies below it. For iron pnictide superconductors with hole and electron Fermi surfaces near the  $\Gamma$  and M points, respectively [Fig. 1(c)]<sup>18,19</sup>, the mode is generally believed to arise from sign reversed quasiparticle excitations between the hole and electron Fermi surfaces and occur at an energy below the sum of their superconducting gap energies  $(E_r \leq \Delta_h + \Delta_e)^{20,21}$ .

If the energy of the resonance is associated with the superconducting gap energies at the hole and electron Fermi surfaces, it should be sensitive to their anisotropy on the respective Fermi surfaces<sup>22,23</sup>. Indeed, recent INS experiments on the NaFe<sub>1-x</sub>Co<sub>x</sub>As family of iron pnictide superconductors reveal the presence of a dispersive sharp resonance near  $E_{r1} = 3.25$ meV and a broad dispersionless mode at  $E_{r2} = 6$ meV at  $\mathbf{Q}_{AF} = (1,0)$  in electron underdoped superconducting NaFe<sub>0.985</sub>Co<sub>0.015</sub>As with static AF order ( $T_c = 15$  K and  $T_N = 30$  K)<sup>15,16</sup>. From the electronic phase diagram of NaFe<sub>1-x</sub>Co<sub>x</sub>As determined from specific heat<sup>24</sup>, scanning tunneling microscopy<sup>25</sup>, and nuclear magnetic resonance  $(NMR)^{26,27}$  experiments, we know that  $NaFe_{0.985}Co_{0.015}As$  is a bulk superconductor with microscopically coexisting static AF ordered and superconducting phases. For Co-doping near optimal superconductivity around x = 0.0175,  $NaFe_{1-x}Co_xAs$  becomes mesoscopically phase separated with the static AF ordered and paramagnetic super-conducting  $\rm phases^{27}, \ similar$  to the co-existing cluster spin glass and superconducting phases in optimally electron-doped BaFe<sub>2</sub>As<sub>2</sub><sup>28,29</sup>. Since angle resolved photoemission spectroscopy (ARPES) experiments on  $NaFe_{0.985}Co_{0.015}As$  found a large superconducting gap anisotropy in the electron Fermi pockets<sup>30</sup>, the double resonance may result from orbital-selective pairing induced superconducting gap anisotropy along the electron Fermi surfaces $^{31}$ . Upon increasing electron doping to x = 0.045 to form superconducting NaFe<sub>0.955</sub>Co<sub>0.045</sub>As  $(T_c = 20 \text{ K})$ , the superconducting gap anisotropy disappears<sup>32,33</sup> and INS reveals only a single sharp resonance coupled with superconductivity<sup>34</sup>.

Although the superconducting gap anisotropy provided a possible interpretation<sup>31</sup>, the double resonance in underdoped NaFe<sub>0.985</sub>Co<sub>0.015</sub>As may also be due to the coexisting static AF order with superconductivity<sup>35,36</sup>. Since  $\mathbf{Q}_{AF} = \mathbf{Q}_1 = (1,0)$  characterizes the explicit breaking of the spin rotational symmetry in the AF ordered state of a completely detwinned sample [Fig. 1(b) and 1(c)]<sup>37</sup>, one should expect magnetic susceptibility anisotropy at  $\mathbf{Q}_{AF} = \mathbf{Q}_1 = (1,0)$  and  $\mathbf{Q}_2 = (0,1)$ . At the AF ordering wave vector  $\mathbf{Q}_{AF} = \mathbf{Q}_1 = (1,0)$ , the resonance appears in the longitudinal susceptibility, whereas the transverse component displays a spin-wave Goldstone mode. At the other momentum  $\mathbf{Q}_2 = (0,1)$ , the reso-

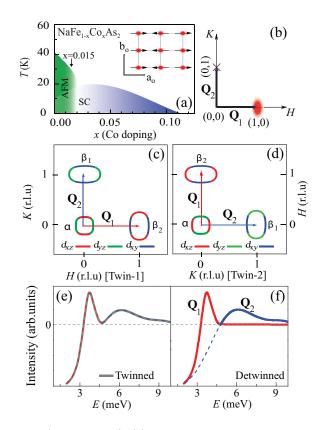


FIG. 1: (Color online) (a) The electronic phase diagram of NaFe<sub>1-x</sub>Co<sub>x</sub>As, where the arrow indicates the Co-doping level of our sample. The inset shows the magnetic structure in orthorhombic notation. (b) In a 100% detwinned sample, one should observe magnetic order at  $\mathbf{Q}_{AF} = \mathbf{Q}_1 = (1, 0)$  but not at  $\mathbf{Q}_2 = (0, 1)$ . (c) The schematic drawings of the Fermi surfaces in a detwinned sample. (d) Its twin domain rotated 90° away. The arrows mark Fermi surface nesting wave vectors. (e) In a twinned sample, one cannot distinguish  $\mathbf{Q}_1 = (1, 0)$ and  $\mathbf{Q}_2 = (0, 1)$ , and therefore there should be two resonances at both wave vectors. (f) In the AF order and superconductivity coexisting theory,  $E_{r1}$  should appear at  $\mathbf{Q}_1$  and  $E_{r2}$  at  $\mathbf{Q}_2$  in a completely detwinned superconducting sample with static AF order at  $\mathbf{Q}_1$ .

nance has both longitudinal and transverse components and is isotropic in space. If the resonance shows distinct energy scales at  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$ , one would expect to find a double resonance in a twinned sample as shown in Fig.  $1(e)^{35,36}$ . However, one would then expect a single resonance of energy  $E_{r1}$  at  $\mathbf{Q}_1$  and that of energy  $E_{r2}$  at  $\mathbf{Q}_2$ in a completely detwinned superconducting sample with static AF order [Fig. 1(f)].

To test if this is indeed the case, we have carried out INS experiments on uniaxial strain partially detwinned NaFe<sub>0.985</sub>Co<sub>0.015</sub>As to study the neutron spin resonance at  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$ . Instead of  $E_{r1}$  at  $\mathbf{Q}_1$  and  $E_{r2}$  at  $\mathbf{Q}_2$  as expected from the theory of coexisting static AF order with superconductivity<sup>35,36</sup>, we find that both  $E_{r1}$  and  $E_{r2}$  are present at  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$  as in the twinned case. Therefore, the presence of the double resonance is not directly associated with the breaking of the spin rotational symmetry

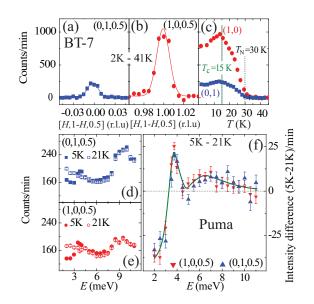


FIG. 2: (Color online) Temperature differences of the elastic scattering below (2 K) and above (41 K) the Néel temperature  $(T_N = 33 \text{ K})$  at reciprocal positions (a) (0,1,0.5)and (b) (1,0,0.5) in a uniaxial strained NaFe<sub>0.985</sub>Co<sub>0.015</sub>As single crystal. The uniaxial strain along the b axis is  $\sim$ 10 MPa and data was collected on BT-7. By comparing the area ratio of the two peaks, we estimate that  $\sim$  $58\% \approx (I(1,0,0.5) - I(0,1,0.5))/(I(1,0,0.5) + I(0,1,0.5))]$  of the crystal is detwinned. The solid lines are Gaussian fits to the data. Similar detwinning ratio is also obtained at PUMA. After releasing the uniaxial strain, we find that the sample returns to the twinned state. (c) Temperature dependence of the magnetic order parameters at  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$  in unixially strained detwinned NaFe<sub>0.985</sub>Co<sub>0.015</sub>As. (d) and (e) Constant  $\mathbf{Q}$ -scans at  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$  below and above  $T_c$ , respectively, in the partially detwinned sample. The peaks around  ${\sim}9~{\rm mV}$ in the raw data are mostly temperature independent background scattering. (f) Temperature differences between 5 K and 21 K, revealing the superconductivity-induced resonance at  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$ . The nearly identical resonances at these two vectors suggests that the coupling between spin waves and superconductivity is weak in Co-doped NaFeAs. The solid line is a guide to the eye.

in detwinned NaFe<sub>0.985</sub>Co<sub>0.015</sub>As. Instead, our results are consistent with the notion that the splitting of the resonance is due to superconducting gap anisotropy in the underdoped NaFe<sub>0.985</sub>Co<sub>0.015</sub>As, suggesting weak direct coupling between spin waves and superconductivity. These results are also consistent with polarized neutron scattering data, where the longitudinal spin excitations of  $E_{r1}$  reveals a clear order-parameter-like increase below  $T_c$  reminiscent of the resonance, while the transverse spin excitations of the  $E_{r1}$  from the spin-wave Goldstone mode have no anomaly across  $T_c^{-16}$ .

We prepared single crystals of NaFe<sub>0.985</sub>Co<sub>0.015</sub>As by the self-flux method<sup>15</sup> and cut a large crystal into the rectangular shape along the [1,0,0] and [0,1,0] directions (16.11 × 8.41 × 1.31 mm<sup>3</sup>, ~0.79 g). From NMR measurements<sup>27</sup>, we know that the tetragonal-

to-orthorhombic structural transition happens around  $T_s \approx 40$  K, above  $T_N$  and  $T_c$ . Our neutron scattering experiments were carried out on the PUMA and BT-7 thermal triple-axis spectrometers at the MLZ, TU Müchen, Germany<sup>11</sup>, and NIST center for neutron research (NCNR), Gaithersburg, Maryland<sup>38</sup>, respectively. In both cases, we used vertically and horizontally focused pyrolytic monochromator and analyzer with a fixed final neutron energy of  $E_f = 14.7$  meV. The wave vector **Q** at  $(q_x,q_y,q_z)$  in Å<sup>-1</sup> is defined as (H,K,L) = $(q_x a/2\pi, q_y a/2\pi, q_z c/2\pi)$  reciprocal lattice unit (r.l.u.) using the orthorhombic unit cell ( $a \approx b \approx 5.589$  Å and c = 6.980 Å at 3 K). In this notation, the AF Bragg peaks occur at the (1, 0, L) positions with  $L = 0.5, 1.5, \cdots$  and there are no magnetic peaks at (0, 1, L) [Figs. 1(a) and 1(b)<sup>39</sup>. We have used a detwinning device similar to that of the previous INS work on  $BaFe_{2-x}Ni_xAs_2^{40}$ . The samples are aligned in the  $[1,0,0.5] \times [0,1,0.5]$  scattering plane. In this scattering geometry, we can probe the static AF order and spin excitations at both  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$ , thus allowing a conclusive determination of the detwinning ratio and spin excitation anisotropy at these wave vectors. Figure 2(a) and 2(b) shows the temperature differences in transverse elastic scans along the [H, 1-H, 0.5] and [H, 1-H, 0.5] directions, respectively, for NaFe<sub>0.985</sub>Co<sub>0.015</sub>As between 2 K and 41 K. By comparing the scattering intensity at these two wave vectors, we estimate that the sample is about 58% detwinned. Figure 2(c) shows the temperature dependence of the magnetic order parameters. Consistent with previous data on twinned sample<sup>15</sup>, the uniaxial strain used to detwinn the sample does not seem to alter  $T_N \approx 30$  K and  $T_c = 15$  K.

In previous INS work on twinned NaFe<sub>0.985</sub>Co<sub>0.015</sub>As, superconductivity induces a dispersive sharp resonance near  $E_{r1} = 3.25$  meV and a broad dispersionless mode at  $E_{r2} = 6 \text{ meV}$  at  $\mathbf{Q}_1 = (1, 0, 0.5)$  and  $\mathbf{Q}_2 = (0, 1, 0.5)^{15}$ . To explore what happens in the uniaxial strain detwinned  $NaFe_{0.985}Co_{0.015}As$ , we carried out constant-Q scans at wave vectors  $\mathbf{Q}_2$  [Fig. 2(d)] and  $\mathbf{Q}_1$  [Fig. 2(e)] below and above  $T_c$ . While it is difficult to see the resonance in the raw data, the temperature differences between 5 K and 21 K plotted in Fig. 2(f) reveal a sharp peak at  $E_{r1} = 3.75$ meV and a broad peak at  $E_{r2} = 6$  meV, in addition to the negative scattering below 3 meV due to a spin gap. These data indicate the presence of a superconductivityinduced sharp resonance and a broad resonance above a spin gap, similar to the results on twinned samples<sup>15</sup>. Surprising, there are no observable differences for the resonance at  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$ , suggesting that the double resonance is not directly associated with the twinning state of the sample.

To confirm the conclusion of Fig. 2, we carried out constant-energy scans near  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$  at  $E_{r1}$  and  $E_{r2}$  above and below  $T_c$ . Figure 3(a) and 3(b) shows transverse rocking curve scans through  $\mathbf{Q}_2 = (0, 1, 0.5)$  and  $\mathbf{Q}_1 = (1, 0, 0.5)$ , respectively, at the sharp resonance energy  $E_{r1} = 3.75$  meV above and below  $T_c$ . While there

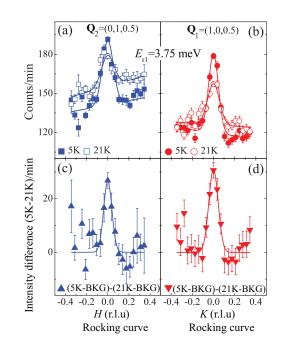


FIG. 3: (Color online) Constant-energy scans at the low-energy resonance  $E_{r1}=3.75$  meV for detwinned NaFe<sub>0.985</sub>Co<sub>0.015</sub>As. (a) and (b) The rocking scans across  $\mathbf{Q}_2 = (0, 1, 0.5)$  and  $\mathbf{Q}_1 = (1, 0, 0.5)$  positions above (21 K) and below (5 K)  $T_c$ , respectively. The wave vector independent background scattering increases slightly on warming. (c) and (d) Corresponding difference between the two temperatures after the background subtraction. The solid lines are fits to Gaussians on linear backgrounds set to zero.

is slightly more magnetic scattering at the AF ordering wave vector  $\mathbf{Q}_1 = (1, 0, 0.5)$  compared with that at  $\mathbf{Q}_2 = (0, 1, 0.5)$  in both the normal and superconducting states<sup>40</sup>, the superconductivity induced intensity changes shown in Fig. 3(c) and 3(d), defined as the resonance<sup>2</sup>, at these two wave vectors are indistinguishable within the statistics of our measurement.

Figures 4(a-d) summarize wave vector scans at the energy of the broad resonance  $E_{r2} = 6$  meV around  $\mathbf{Q}_2 = (0, 1, 0.5)$  and  $\mathbf{Q}_1 = (1, 0, 0.5)$ . Similar to data at  $E_{r1} = 3.75$  meV, we find that the superconductivity-induced intensity gain of the broad resonance is almost indistinguishable at these wave vectors, again confirming the notion that the resonance is not sensitive to the twinning state of the system.

In electron doped NaFe<sub>1-x</sub>Co<sub>x</sub>As, the dominant orbital character of the electron pockets would be  $d_{xy/xz}$ at (1,0) and  $d_{yz/xy}$  at (0,1) in the Brillouin zone [Fig. 1(c)]<sup>30,32,33</sup>. The orbital character of the hole pocket is  $d_{xz/yz}$ . If the superconducting pairing amplitudes are highly orbital dependent, *i.e.*,  $\Delta_{xy} \neq \Delta_{xz/yz}$ , the superconducting gap can be anisotropic along the electron pocket and this gap anisotropy gives rise to a splitting of the resonance peak<sup>31</sup>. Such an orbital-selective pairing scenario is consistent with both ARPES measurements<sup>30</sup>

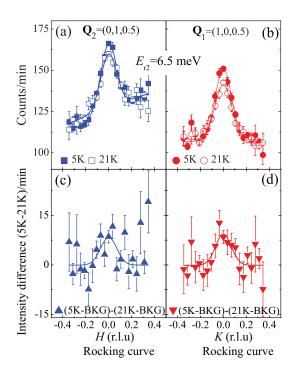


FIG. 4: (Color online) Constant-energy scans through the high-energy resonance  $E_{r2}=6.5$  meV for detwinned NaFe<sub>0.985</sub>Co<sub>0.015</sub>As. (a) and (b) The rocking scans across the  $\mathbf{Q}_2 = (0, 1, 0.5)$  and  $\mathbf{Q}_1 = (1, 0, 0.5)$  positions above (21 K) and below (5 K)  $T_c$ , respectively. (c) and (d) The corresponding difference between the two temperatures. The solid lines are fits to Gaussians on zero backgrounds.

and INS results in twinned samples<sup>15</sup>.

In the uniaxial strain detwinned sample, the degeneracy of the  $d_{xz}$  and  $d_{yz}$  orbitals is lifted and, correspondingly, the Fermi surface is distorted<sup>30</sup>. To investigate whether the double resonances in the orbital-selective pairing scenario still exist in the presence of a splitting between the  $d_{xz}$  and  $d_{yz}$  orbitals, we calculated the imaginary part of the spin susceptibility  $\chi''$  in the superconducting state from a multiorbital  $t - J_1 - J_2$  model with an orbital splitting term  $\epsilon = n_{xz} - n_{yz}^{-31}$ . Our result for a strong orbital selectivity is presented in Fig. 5. We find two resonance peaks at each of the wave vectors  $\mathbf{Q}_1$ and  $\mathbf{Q}_2$  for a nonzero splitting  $\epsilon$ . The intensities of the counterpart peaks at  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$  are comparable. At each resonance peak, there is a relative shift of the resonance energy between the  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$  resonances. This shift is proportional to the splitting  $\epsilon$ . The calculated double-resonances feature at both  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$  is qualitatively consistent with the experimental observation in the detwinned sample. The experiment can not resolve a relative shift of the resonance energy. This could be because either the splitting  $\epsilon$  is small in the detwinned underdoped compound, or the coupling between the superconductivity and the splitting  $\epsilon$  is rather weak. Further comparison between theory and experiments is needed to fully settle the issue.

In conclusion, our INS experiments on partially de-

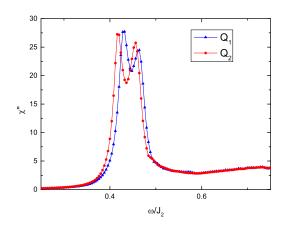


FIG. 5: Imaginary part of the dynamic spin susceptibility  $\chi''$ in the superconducting state of a multiorbital  $t-J_1-J_2$  model with a nonzero splitting between the  $d_{xz}$  and  $d_{yz}$  orbitals,  $\epsilon = 0.02$  eV. Two resonance peaks are present in each of the  $\mathbf{Q}_1$  and  $\mathbf{Q}_2$  wave vectors. In the calculation,  $J_1/J_2 = 0.1$ is taken such that the pairing amplitudes show strong orbital selectivity. See Ref.<sup>31</sup> for details of the model and the method.

twinned NaFe<sub>0.985</sub>Co<sub>0.015</sub>As reveal the presence of two resonances at each of the wave vectors  $\mathbf{Q}_{AF} = \mathbf{Q}_1 = (1,0)$ and  $\mathbf{Q}_2 = (0,1)$ . This is different from the scenario where the two resonances are due to the coexisting AF order with superconductivity<sup>35,36</sup>. Instead, the data are qualitatively consistent with the proposal that the double resonances originate from an orbital dependence of the superconducting pairing. Our results provide further evidence that orbital selectivity plays an important role in understanding not only the normal state but also the superconducting pairing of the multiorbital electrons in the iron pnictides.

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