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Electron doping evolution of the neutron spin resonance in NaFe$_{1-x}$Co$_x$As

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Neutron spin resonance, a collective magnetic excitation coupled to superconductivity, is one of the most prominent features shared by a broad family of unconventional superconductors including copper oxides, iron pnictides, and heavy fermions. In this work, we study the doping evolution of the resonances in NaFe$_{1-x}$Co$_x$As covering the entire superconducting dome. For the underdoped compositions, two resonance modes coexist. As doping increases, the low-energy resonance gradually loses its spectral weight to the high-energy one but remains at the same energy. By contrast, in the overdoped regime we only find one single resonance, which acquires a broader width in both energy and momentum, but retains approximately the same peak position even when $T_c$ drops by nearly a half compared to optimal doping. These results suggest that the energy of the resonance in electron overdoped NaFe$_{1-x}$Co$_x$As is neither simply proportional to $T_c$ nor the superconducting gap, but is controlled by the multi-orbital character of the system and doped impurity scattering effect.

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I. INTRODUCTION

Although the microscopic origin of superconductivity remains unresolved nearly 30 years after the discovery of high-transition temperature (high-$T_c$) copper oxides¹, it is generally believed that spin fluctuation mediated electron pairing is a common thread for unconventional superconductors including copper oxide, iron-based, and heavy-fermion superconductors²-⁵. Regardless of the dramatic differences in the ground states of their parent compounds and the microscopic origins of magnetism in different families of unconventional superconductors, inelastic neutron scattering experiments have revealed that superconductivity induces a collective magnetic excitation, termed neutron spin resonance, near the antiferromagnetic (AF) ordering wave vector of their parent compounds⁴-⁸. Experimentally, the resonance occurs at an energy $E_r$ and enhances dramatically below $T_c$ like the superconducting order parameter. In the Fermi surface nesting (itinerant electron) picture⁵, the resonance is a spin-exciton mode in the particle-hole channel. If the superconducting order parameter has a sign-change below $T_c$, the dynamic spin susceptibility will develop a pole, namely the resonance, at an energy $E_r$ below the particle-hole continuum $2\Delta$ (where $\Delta$ is the superconducting gap)⁵. In the case of iron pnictide superconductor NaFe$_{1-x}$Co$_x$As with hole and electron Fermi surfaces at $\Gamma$ and $M$ points, respectively [Fig. 1(a),1(e)-1(g)]¹⁰-¹⁶, the resonance arises from quasiparticle excitations between the sign-reversed hole and electron Fermi surfaces and occurs at an energy below the sum of their superconducting gap energies ($E_r \leq \Delta_h + \Delta_e = 2\Delta$, where $\Delta_h$ and $\Delta_e$ are superconducting gaps at hole and electron Fermi surfaces, respectively) [Fig. 1(b)-1(d)]¹⁷,¹⁸. Although $T_c$ differs dramatically for copper oxide, iron-based, and heavy-fermion superconductors, the resonance energy $E_r$ is approximately related to $T_c$ via $E_r/k_BT_c \approx 4-6$ or the superconducting gap energy $\Delta$ via $E_r/2\Delta = 0.64$ ($2\Delta/k_BT_c = A + BT_c$, where $A$ and $B$ are constants)¹⁹-²¹. While these results suggest that the resonance may be a common thread for unconventional superconductors², most of the inelastic neutron scattering measurements on the resonance in iron pnictides are focused on underdoped and optimally doped samples with few experiments on overdoped regime of the phase diagram²⁰.

In this paper, we report systematic inelastic neutron scattering studies of the resonance in iron pnictide superconductors NaFe$_{1-x}$Co$_x$As for Co compositions throughout the entire superconducting dome²²-²⁶. In previous work on electron overdoped NaFe$_{0.985}$Co$_{0.015}$As where static AF order coexists with superconductivity ($T_N = 30$ K, and $T_c = 15$ K), we find a dispersive sharp resonance near $E_r = 3.25$ meV and a broad dispersion-less mode at $E_r = 6$ meV at the AF ordering wave vector $Q_{AF}^{12,27}$. Upon moving to electron overdoped NaFe$_{0.955}$Co$_{0.045}$As without static AF order ($T_c = 20$ K), there is only one sharp resonance at $E_r = 7$ meV²⁸. By carrying out systematic measurements on NaFe$_{1-x}$Co$_x$As with nominal Co-doping of $x = 0.012, 0.0135, 0.0175, 0.025, 0.08$ [Fig. 1(a)], we establish the electron-doping evolution of the resonance throughout the superconducting phase. In the underdoped regime, we confirm the earlier results showing the presence of double resonance peaks at $E_{r1}$ and $E_{r2}$ as shown in Fig. 1(b). As doping increases, $E_{r1}$ stays almost the same value while $E_{r2}$ moves to higher energies. At optimal doping and in slightly overdoped samples, the low-energy resonance disappears and only a single sharp resonance occurs at $E_{r2} = 7$ meV [Fig. 1(c)]. For
heavily overdoped $x = 0.08$, the resonance becomes much broader in energy but retains its peak position [Fig. 1(d)]. These results indicate that the resonance energy in the electron overdoped regime is neither directly associated with $T_c$ via the empirical relation $E_r/k_B T_c = 4 \sim 6$ nor with $\Delta$ via $E_r/\Delta_b + \Delta_e = 0.64^{19-21}$, thus suggesting that the multi-orbital character and the interband nonmagnetic impurity scattering due to Co-doping in NaFe$_{1-x}$Co$_x$As play an important role in determining the properties of the resonance.

II. EXPERIMENTAL RESULTS

We grew single crystals of NaFe$_{1-x}$Co$_x$As by self-flux method as described before$^{29}$. The sample quality has been characterized by various techniques, which found that bulk superconductivity appears in the doping range of $0.012 \leq x \leq 0.115$. Our inelastic neutron scattering experiments were carried out over the entire doping range as shown by vertical arrows in Fig. 1(a). The measurements were performed on the HB-1 and HB-3 thermal triple-axis spectrometers at High Flux Isotope Reactor, Oak Ridge National Laboratory, and SPINS cold triple-axis spectrometer at the NIST Center for Neutron Research. Pyrolytic graphite (PG) monochromator and analyzer were used with fixed final neutron energies at $E_f = 14.7$ meV and $E_f = 5$ meV for thermal and cold neutron measurements, respectively. The corresponding energy resolutions are $\Delta E \approx 1.2$ meV and $\Delta E \approx 0.15$ meV, respectively, at the AF ordering elastic position. Several pieces of crystals co-aligned with a total mass of $\sim 10$ g and the mosaic of $\sim 3^\circ$ were used in each experiment. The wave vector $Q$ at $(q_x, q_y, q_z)$ in $\AA^{-1}$ is

![Image](attachment:image.png)

FIG. 1: (Color online) (a) The electronic phase diagram of NaFe$_{1-x}$Co$_x$As, where the arrow indicates the Co-doping levels studied in this work. The grey shaded area marks the Co-doping dependence of $T_c$. The region with AF order is represented by the green shaded area. The open circles are energies of the first resonance $E_r$, and the filled circles and stars are energies of the second resonance $E_{r2}$. The yellow shaded area indicates approximate range of $T_c$ via the empirical relation $E_r/k_B T_c = 4 \sim 6$. The dashed grey lines show the doping dependence of $E_r$, and the dotted lines are the $E_r$ at $T_c$. (b-d) The schematic energy dependence of the resonance for three characteristic Co-doping levels, including underdoped (UD), optimally doped (OD), and highly overdoped (HD). (e-g) Schematic plots of the Fermi surfacing for the above three compositions. The color indicate different orbitals. The anisotropic superconducting gap $\Delta$, on the electron pockets in the underdoped compounds become isotropic on the overdoped side$^{16}$.

![Image](attachment:image.png)

FIG. 2: (Color online) The neutron resonances in NaFe$_{1-x}$Co$_x$As as a function of increasing $x$, obtained as the difference of the energy-scans above and below $T_c$ at the wave vectors $Q_{AF} = (1,0,L)$ and $Q_{AF} = (1,0,0)$ with $L = 0.5,1.5$ (a-e) and $Q = (1,0,L)$ with $L = 0.1$ (f-j). (a,f) $x = 0.012$ (UD); (b,g) $x = 0.0135$ (UD); (c,h) $x = 0.015$ (UD); (d,i) $x = 0.0175$ (UD); (e,j) $x = 0.025$ (OP). The plots are obtained directly by subtracting the superconducting state energy scan from those in the normal state without correcting for background, as is commonly done for determining the energy of the resonance$^{19-21}$. The solid lines are fits with two Gaussians. The vertical dashed lines denote the low-energy resonance at $E_r1$ in (a-d) and (f-i). The negative intensity below the resonance indicates the opening of a spin gap below $T_c$. The vertical arrows indicate the peak positions of the high-energy resonance $E_{r2}$ at $Q_{AF} = (1,0,0)$ with $L = 0.5,1.5$. The energy resolutions are $\Delta E \approx 1.2$ meV and $\Delta E \approx 0.15$ meV, respectively, at the AF ordering elastic position.

![Image](attachment:image.png)
nance occurs at slightly different energies at the AF zone \( Q = 0 \times \) in the superconducting state. For this purpose, energy resonance as the intensity gain of magnetic scattering 4,19 below and above 3 K. The samples are aligned in the \([H, 0, L]\) scattering zone, where the resonance occurs at the AF wave vector \( Q = (1, 0) \), consistent with the Fermi surface nesting wave vector shown in Fig. 1(e)27,28. Some measurements are carried out in the \([H, K, 0]\) scattering plane.

To systematically investigate the electron-doping evolution of the double resonance in the underdoped regime27, we first focus on a series of compositions from \( x = 0.012 \) to \( x = 0.0175 \) [Fig. 2(a)-(d),(f)-(i)]. Similar to previous neutron scattering work4,19, we define resonance as the intensity gain of magnetic scattering in the superconducting state. For this purpose, energy scans are carried out at fixed wave vectors below and above \( T_c \), and the net intensity gain of the scattering below \( T_c \) is ascribed to the resonance. In the case of NaFe\(_{1-x}\)Co\(_x\)As, previous work has shown that the resonance occurs at slightly different energies at the AF zone center \( Q_{AF} = (1, 0, L) \) with \( L = 0.5, 1.5 \) and boundary with \( L = 0.1 \)27. We have therefore carried out systematic measurements at these two wave vectors for all Co-doping levels. Figure 2(a) and 2(f) shows the outcome for NaFe\(_{1-x}\)Co\(_x\)As with \( x = 0.012 \), when the system first becomes near the bulk superconducting phase15. The temperature difference plot shows a resonance peak at \( E_{r1} = 3.75 \) meV for \( L = 0.5 \) and \( E_{r1} = 4.5 \) meV for \( L = 0 \) with the corresponding spin gaps of \( E_g \approx 3 \) and 4 meV, respectively. To further confirm the existence of the resonance, we carried out momentum and temperature dependence measurements on SPINS. Figure 3(a) shows constant-energy scans at \( E_{r1} = 3.75 \) meV along the \([H, 0, 0.5]\) direction, which reveals clear intensity gain below \( T_c \) at \( Q_{AF} \). For an energy below the resonance at \( E = 0.75 \) meV, AF spin fluctuations are completely suppressed below \( T_c \), suggesting the opening of a spin gap in the superconducting state [Fig. 3(b)]. Temperature dependence of the elastic magnetic scattering is shown in Fig. 3(c). Similar to previous work on underdoped superconducting iron pnictides30,31, we see clear evidence for AF order below \( T_N \approx 35 \) K and the suppressive effect of superconductivity on AF order. Figure 3(d) shows temperature dependence of scattering at \( E_{r1} = 3.75 \) meV and \( Q_{AF} = (1, 0, 0.5) \). Based on these results, we find clear evidence for the resonance in the \( x = 0.012 \) compound.

At higher doping levels, \( x = 0.0135 \) [Fig. 2(b, g)], \( x = 0.015 \) [Fig. 2(c, h)], and \( x = 0.175 \) [Fig. 2(d, i)], a second resonance mode with a broad width appears at a higher energy \( E_{r2} \). As the superconducting transition temperature \( T_c \) increases with increasing Co-doping, \( E_{r2} \) also increases, whereas \( E_{r1} \) stays at almost the same energy for \( Q_{AF} = (1, 0, 0.5) \). These results suggest that the energy of the first resonance is not directly associated with \( k_B T_c \). Furthermore, we note that the spectral weight of the low-energy resonance gradually shifts to the
of the susceptibility for $x = 0.045$ and $x = 0.08$. For Co-doping levels above $x = 0.025$, resonances are not dispersive along the $L$ direction. (b) Temperature dependence of the susceptibility for $x = 0.045$ and $x = 0.08$. The superconducting volume fraction of the $x = 0.08$ sample is significantly lower than that of the electron doping $x = 0.045$ [Fig. 5(b)]$^{28}$, we would expect a reduction in the superconducting gap amplitude $2\Delta = \Delta_h + \Delta_e$ as well$^{33}$. If the resonance is a bound-state below the particle-hole continuum $2\Delta$, there should be a corresponding reduction in the mode energy on moving from $x = 0.045$ to $x = 0.08$. Figure 5(a) compares temperature difference plots of the energy scans below and above $T_c$ for $x = 0.045$ to $x = 0.08$. While there is a clear resonance in both samples, the resonance for $x = 0.08$ shows a much broader width compared to that of $x = 0.045$ even considering the differences in instrumental energy resolution in these two experiments. In addition, the two resonances have almost the same peak energy at $E_r = 7$ meV, despite the large reduction in $T_c$ for $x = 0.045$ to $x = 0.08$. To confirm that the intensity gain below $T_c$ in the $x = 0.08$ sample is indeed the resonance, we show in Fig. 5(c) temperature dependence of the scattering at $E_r = 7$ meV. For both $x = 0.045$ to $x = 0.08$ samples, there are clear superconducting order parameter like intensity gain below $T_c$’s, a hallmark of the resonance. Figure 5(d) and 5(e) shows constant-energy scans above background below and above $T_c$ along the $[H,0,0]$ and $[1,K,0]$ directions, respectively, for $x = 0.08$. The red solid lines are similar wave vector scans for the $x = 0.045$ sample$^{28}$. These results confirm the temperature difference plots, showing that intensity gain of below $T_c$ in Fig. 5(a) and 5(c) is indeed from the resonance. Although $x = 0.08$ sample is not a 100% bulk superconductor [Fig. 5(b)], the differences between the superconducting and normal state should still represent the effect of superconductivity to the magnetic excitations. Based on the properties of the resonance in the $x = 0.045$ to $x = 0.08$ samples shown in Fig. 5, we conclude that the mode energy $E_r$ does not scale linearly with $T_c$ or $\Delta$. The ratios $E_r/k_BT_c$ and $E_r/2\Delta$ in the $x = 0.08$ composition are well above the values proposed in the universal relations [see Fig. 1(a)]. Furthermore, we find that while the resonance for both samples are centered at the AF ordering wave vector, the $x = 0.08$ sample has considerable broader $Q$-width along the $H$ and $K$ directions.

III. DISCUSSION AND CONCLUSION

Figure 6 summarizes the Co-doping evolution of the resonance in NaFe$_{1-x}$Co$_x$As. The open circles in Fig. 6(a) shows that the energy of the first resonance $E_{r1}$ is essentially $T_c$ independent. If the double resonance originates from the superconducting gap anisotropy in the underdoped regime$^{16,27,34}$, one would expect that $E_{r1}$ decreases with increasing doping, contrary to the observation. On the other hand, these results may indicate that the first resonance is coupled with the static AF order and spin waves as suggested theoretically$^{35}$. If this is indeed the case, one would expect that an uniaxial pressure used to detwin the sample would separate the double resonance, where the first resonance associated with spin waves ($E_{r1}$) should appear at $Q_{AF} = (\pm 1,0)$ but not at $(0, \pm 1)$, while the second resonance ($E_{r2}$)
which causes the hole pockets to shrink and the electron pockets to expand, as illustrated in Fig. 1(e-g). As the mismatch between the electron and hole pockets increases with doping, the resonance peak obtains more contributions from the scattering momenta that are away from the AF order wave vector (1, 0), and therefore shows a broader peak in the momentum space. This is reminiscent of the wave vector dependence of the resonance in BaFe$_{2-x}$Ni$_x$As$_2$ family of materials, where the mode becomes transversely incommensurate in the electron-overdoped regime, except here the scattering is commensurate in the entire measured doping range. With electron overdoping and sinking of the hole pocket below Fermi surface, the low-energy spin excitations vanish together with the suppression of superconductivity, very similar to the presence of a large spin gap in electron-overdoped nonsuperconducting BaFe$_{1.7}$Ni$_{0.3}$As$_2$. The second less considered effect is that the Co dopants can also act as local nonmagnetic impurities. In iron pnictides where the superconducting order parameter changes sign between the hole and electron pockets [Fig. 1(e-g)], interband scatterings from these impurities are superconducting pair-breaking. Therefore, as more impurities are introduced with increasing Co-doping, we expect that the superconducting gap to be gradually filled and the critical temperature $T_c$ to be reduced due to these pair-breaking scatterings. However, the spin resonance arises from the superconducting quasiparticles that retain the original gap amplitude $\Delta$. Therefore, the resonance energy $E_r$ is not much affected by these interband nonmagnetic scatterings, and the mode will acquire a larger width in energy due to the broadened quasiparticle peak with increasing impurity concentration. These results are consistent with our experimental observations, suggesting the important roles of the impurity scatterings in determining the energy and wave vector dependence of the resonance. Our study in the overdoped NaFe$_{1-x}$Co$_x$As have demonstrated that the Co dopants introduce two important effects into the system, namely the additional itinerant electrons and local nonmagnetic impurities.

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