Electron doping evolution of the magnetic excitations in BaFe$_{2-x}$Ni$_x$As$_2$

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We use inelastic neutron scattering (INS) spectroscopy to study the magnetic excitations spectra throughout the Brillouin zone in electron-doped iron pnictide superconductors BaFe$_{2-x}$Ni$_x$As$_2$ with $x = 0.096, 0.15, 0.18$. While the $x = 0.096$ sample is near optimal superconductivity with $T_c \approx 20$ K and has coexisting static incommensurate magnetic order, the $x = 0.15, 0.18$ samples are electron-overdoped with reduced $T_c$ of 14 K and 8 K, respectively, and have no static antiferromagnetic (AF) order. In previous INS work on undoped ($x = 0$) and electron optimally doped ($x = 0.1$) samples, the effect of electron-doping was found to modify spin waves in the parent compound BaFe$_2$As$_2$ below ~100 meV and induce a neutron spin resonance at the commensurate AF ordering wave vector that couples with superconductivity. While the new data collected on the $x = 0.096$ sample confirms the overall features of the earlier work, our careful temperature dependent study of the resonance reveals that the resonance suddenly changes its $Q$-width below $T_c$ similar to that of the optimally hole-doped iron pnictides BaFe$_{0.67}$K$_{0.33}$Fe$_2$As$_2$. In addition, we establish the dispersion of the resonance and find it to change from commensurate to transversely incommensurate with increasing energy. Upon further electron-doping to overdoped iron pnictides with $x = 0.15$ and 0.18, the resonance becomes weaker and transversely incommensurate at all energies, while spin excitations above ~100 meV are still not much affected. Our absolute spin excitation intensity measurements throughout the Brillouin zone for $x = 0.096, 0.15, 0.18$ confirm the notion that the low-energy spin excitation coupling with itinerant electron is important for superconductivity in these materials, even though the high-energy spin excitations are weakly doping dependent.

I. INTRODUCTION

Understanding the origin of superconductivity in strongly correlated electron materials is at the forefront of modern condensed matter physics.$^{12}$ Since high-transition temperature (high-$T_c$) superconductors such as copper oxides and iron pnictides are derived from electron or hole-doping to their antiferromagnetic (AF) order parent compounds,$^{12}$ much efforts over the past 27 years have been focused on determining the role of short-range spin excitations in the superconductivity of these materials.$^{12}$ From inelastic neutron scattering (INS) experiments on copper oxide superconductors, it is well established that the low-energy ($E \leq 100$ meV) spin excitations persist throughout the doping-induced superconductivity dome and vanish when superconductivity ceases to exist in the overdoped regime.$^5$ While INS failed to detect high-energy spin excitations near the AF ordering wave vector (0.5, 0.5) in overdoped copper oxides,$^3$ recent resonant inelastic X-ray scattering experiments find that high-energy ($\geq 100$ meV) spin excitations in reciprocal space near the origin are almost independent of hole-doping across the superconductivity dome.$^2$ In the case of iron pnictides (Fig. 1(a)) INS experiments on single crystals of electron-doped BaFe$_{2-x}$T$_x$As$_2$ (where $T =$ Co, Ni) have mapped out the doping evolution of the spin excitations.$^{34,35}$ In the undoped state, BaFe$_2$As$_2$ exhibits nearly simultaneous tetragonal-to-orthorhombic lattice distortion and collinear AF order below $T_N \approx 138$ K [see left inset of Fig. 1(a)].$^3$ In the AF ordered state, BaFe$_2$As$_2$ forms randomly distributed orthorhombic twin domains rotated 90° apart. As a consequence, the low-energy spin waves from the two separate domains are centered around the AF ordering wave vectors $Q_{\text{AF}} = (\pm 1, 0)$ and $(0, \pm 1)$, respectively, in reciprocal space [see right inset in Fig. 1(a)]. INS experiments using time-of-flight (TOF) chopper spectrometer at the ISIS spallation neutron source in UK have measured spin waves of BaFe$_2$As$_2$ in absolute units throughout the Brillouin zone and determined the spin-wave dispersions along the two high symmetry directions as shown in the solid lines of Figs. 1(e) and 1(f).$^{34}$

Figure 1(a) shows the schematic phase diagram of electron-doped BaFe$_{2-x}$Ni$_x$As$_2$ as determined from transport and neutron diffraction experiments.$^{34,35}$ In previous work$^{30}$ the evolution of the low-energy spin excitations was found to qualitatively follow the Fermi surface nesting picture and arise from quasiparticle excitations between the hole and electron Fermi pockets near $\Gamma$ and $M$ points, respectively.$^{30,30}$ By comparing spin waves of the parent compound with spin excitations of the optimally electron-doped superconductor BaFe$_{1.9}$Ni$_{0.1}$As$_2$ in absolute units, it was found that
FIG. 1: (Color online) (a) The schematic electronic phase diagram of BaFe$_{2-x}$Ni$_x$As$_2$, where the arrows at $x = 0.096, 0.15, 0.18$ indicate doping levels studied in this paper$^{24,25}$. The Inserts show the in-plane magnetic structure in real space and Brillouin zone in reciprocal space. (b,c,d) DC magnetic susceptibility indicates nearly 100% diamagnetic volume for all three measured dopings with $T_c = 20$ K, 14 K and 8 K. (e,f) The dispersions of spin excitations along the $[1,K]$ and $[H,0]$ directions for BaFe$_{2-x}$Ni$_x$As$_2$ with $x = 0.096, 0.15, 0.18$. The solid and dash lines are spin wave dispersions in the parent compound BaFe$_2$As$_2$ ($x = 0$)$^{27}$. Electron doping on BaFe$_2$As$_2$ affects only the low-energy spin excitations by broadening the spin waves below 80 meV and forming a low-energy ($E_r \approx 7$ meV) neutron spin resonance below $T_c$, but has no impact on spin waves above 100 meV$^{29}$. From systematic triple-axis IN$^{28}$ and nuclear magnetic resonance$^{43}$ measurements of the low-energy spin excitations in BaFe$_{2-x}$Co$_x$As$_2$, the suppression of superconductivity in electron-overdoped BaFe$_{2-x}$T$_x$As$_2$ is found to be associated with vanishing low-energy spin excitations. Although these re-
tide superconductors, and complements the earlier work on the electron optimal $x = 0.1$ superconductor, and the $x = 0.3$ electron-overdoped nonsuperconductor. Consistent with earlier work, we find that the low energy spin excitations in $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ are transversely elongated ellipses around the commensurate AF order wave vector. For the $x = 0.096$ sample near optimal superconductivity, a neutron spin resonance appears at $E_r = 7$ meV below $T_c$, and the mode forms transversely incommensurate spin excitations at higher energies. While the energy of the resonance is weakly temperature dependent, the transverse and radial widths of the mode show a superconductivity-induced narrowing below $T_c$. For samples at the overdoped side $x = 0.15$, superconductivity induces a transversely incommensurate resonance at $E_r = 6.5$ meV. On increasing electron-doping further to $x = 0.18$, low-energy spin excitations have a broad commensurate component independent of superconductivity and a transversely incommensurate resonance below $T_c$ at $E_r = 5.5$ meV. By comparing TOF INS data in $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ with $x = 0.096, 0.15, 0.18$, we establish the wave vector and energy dependence of the spin excitations throughout the Brillouin zone from optimally electron-doped to electron over-doped iron pnictides. Our results are consistent with the idea that superconductivity in iron pnictides requires the low-energy spin excitation-itinerant electron interaction, and indicate an intimate connection between spin excitations and superconductivity.

II. EXPERIMENT

We carried out INS experiments using the MERLIN TOF chopper spectrometer at the Rutherford-Appleton Laboratory, UK. For the experiments, sizable single crystals of $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ grown by self-flux method were co-aligned on several aluminum plates by hydrogen-free glue with both in-plane and out-of-plane mosaic less than $3^{\circ}$. The total mass of our samples is 41 grams for $x = 0.096$, 45 grams for $x = 0.15$, 25 grams for $x = 0.18$, respectively. Using orthorhombic crystalline lattice unit cell for easy comparison with the spin wave results of $\text{BaFe}_2\text{As}_2$, we define the wave vector $\mathbf{Q}$ 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 (r.l.u.), where $a \approx b \approx 5.60$ Å, and $c = 12.77$ Å. The samples are loaded inside a standard closed-cycle Helium refrigerator with incident beam parallel to the $c$-axis. To probe spin excitations at different energies, we chose neutron incident beam energies of $E_i = 20, 25, 30, 50, 80, 250, 450$ meV with corresponding Fermi chopper frequencies of $\omega = 150, 200, 200, 400, 500, 550, 600$ Hz, respectively. To facilitate comparison with spin waves in $\text{BaFe}_2\text{As}_2$, spin excitations in doped materials are normalized to the absolute units (mbarn/s/meV/f.u.) using a vanadium standard. The neutron scattering cross section $S(Q, E)$ is related to the imaginary part of the dynamic suscep-

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**FIG. 2:** (Color online) Comparison of the dispersions of low-energy spin waves in $\text{BaFe}_2\text{As}_2$ with the FWHM of spin excitations in $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ with $x = 0.096, 0.15, 0.18$ along the $[1, K]$ and $[H, 0]$ directions. (a,b) The solid lines show spin wave dispersions of $\text{BaFe}_2\text{As}_2$. The grey, blue, and brown regions show the FWHM of low-energy spin excitations of $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ with $x = 0.096, 0.15, 0.18$ along the $[1, K]$ and $[H, 0]$ directions, respectively. (c,d) Energy dependence of the FWHM along the transverse $[1, K]$ and longitudinal $[H, 0]$ directions as determined from TOF INS measurements. The FWHM of peak along the $[1, K, 3]$ direction from the triple-axis experiments is also shown in (c).

Results are consistent with the presence of a large spin gap ($\sim 50$ meV) in the electron-overdoped nonsuperconducting $\text{BaFe}_{1.7}\text{Ni}_{0.3}\text{As}_2$; it is still unclear how spin excitations gradually evolve from optimally doped superconductor to electron-overdoped nonsuperconductor. Since spin excitations may mediate electron pairing for superconductivity, it would be important to determine the temperature and electron-doping evolution of spin excitations in $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$ across the superconductivity dome [Fig. 1(a)].

In this article, we report triple-axis and TOF INS studies of temperature and doping dependence of spin excitations in $\text{BaFe}_{2-x}\text{Ni}_x\text{As}_2$. For this work, we chose Ndoping concentrations of $x = 0.096, 0.15$, and $0.18$ with superconducting transition temperatures of $T_c = 20$ K [Fig. 1(b)], $14$ K [Fig. 1(c)], and $8$ K [Fig. 1(d)], respectively. This range of Ndoping covers the nearly optimally electron-doped to electron-overdoped iron pnict-
tibility $\chi''(Q, E)$ by correcting for the Bose population factor via $\chi''(Q, E) = 1/(1 - \exp(-E/(k_B T))) \chi''(Q, E)$, where $k_B$ is the Boltzmann’s constant. We can then calculate the local dynamic susceptibility by using $\chi''(E) = \int \chi''(Q, E) dQ/\int dQ$ (in units of $\mu_B^2/\text{eV/f.u.}$), where $\chi''(Q, E) = (1/3)tr(\chi''_{\alpha\beta}(Q, E))$.

In addition to the TOF INS measurements on MERLIN, we also took data on the $x = 0.096$ compound using the TAIPAN thermal neutron triple-axis spectrometer at the Bragg Institute, Australian Nuclear Science and Technology Organization (ANSTO). The measurements were carried out on $\sim$29 grams co-aligned single crystals using the $[H, 0, 3H] \times [0, -K, 0]$ scattering plane. TAIPAN uses double focusing pyrolytic graphite monochromator and vertical focusing analyzer with a pyrolytic graphite filter before the analyzer and a fixed final neutron energy of $E_f = 14.87$ meV. In $[H, K, 3H]$ scattering plane, we performed transverse scans along the $[1, K, 3]$ direction for energies up to 25 meV. The BaFe$_{2-x}$Ni$_x$As$_2$ samples with $x = 0.096, 0.18$ for TOF and triple-axis experiments are aligned using a Photonic Sciences X-ray Laue camera and co-aligned by using TAIPAN triple-axis spectrometer at ANSTO and ALF crystal alignment facility at ISIS. The BaFe$_{1.85}$Ni$_{0.15}$As$_2$ samples for TOF experiments are co-aligned using E3 neutron four-circle diffraction spectrometer at Canadian Neutron Beam Center in Chalk River, Canada.

### III. RESULTS

We first describe the evolution of spin excitation dispersions in BaFe$_{2-x}$Ni$_x$As$_2$. Figure 1(e) and (f) show the overall dispersions along the $Q = [1, K]$ and $[H, 0]$ directions for $x = 0.096, 0.15$ and 0.18 compared with the parent compound $x = 0$ (black solid lines). While the spin excitations at low-energy below $\sim$100 meV become slightly more dispersive upon Ni doping, the high energy spin excitations are not much affected by electron doping, similar to those in the heavily electron overdoped BaFe$_{1.7}$Ni$_{0.3}$As$_2$. These results suggest that the effective magnetic exchange couplings $J$ are not much affected by electron-doping in BaFe$_{2-x}$Ni$_x$As$_2$ for $x \leq 0.3$.

To further study the effect of electron-doping to the low energy ($E < 60$ meV) spin excitations, we fit the wave vector dependence of the spin excitations along the $[1, K]$ and $[H, 0]$ directions by Gaussian on a linear background to estimate their full-width-at-half-maximum (FWHM) in BaFe$_{2-x}$Ni$_x$As$_2$ with $x = 0.096, 0.15, 0.18$. For comparison, we also probe the low-energy spin excitations on BaFe$_{2-x}$Ni$_x$As$_2$ with $x = 0.096$ using TAIPAN triple-axis spectrometer along the $Q = [1, K, 3]$ direction. The solid black lines in Figs. 2(a) and 2(b) are spin wave dispersions of BaFe$_{2-x}$As$_2$ estimated using the previous obtained in-plane effective magnetic exchange couplings and appropriate spin anisotropy gap values. The shaped area in Figs. 2(a) and 2(b) show the FWHM of spin excitations for BaFe$_{2-x}$Ni$_x$As$_2$ with $x = 0.096, 0.15, 0.18$ along the $[1, K]$ and $[H, 0]$ directions, respectively. Figure 2(c) shows energy dependence of the spin excitation widths along the $[1, K]$ direction. Within the probed energy range ($3 \leq E \leq 60$ meV), the widths of spin excitations increase monotonically with increasing energy and the electron-doping level $x$ for these three samples. For BaFe$_{1.904}$Ni$_{0.096}$As$_2$, the spin excitation widths determined from the triple-axis experiments are slightly smaller than that from the TOF measurements due to the differences in the instrumental resolutions in these two techniques. Figure 2(d) shows the energy dependence of the spin excitation widths along the $[H, 0]$ direction, which are almost independent of electron-doping above 30 meV. Thus the low-energy spin excitations are transversely elongated upon doping and become broader than spin waves in the undoped compound.

To directly compare the evolution of spin excitations as a function of increasing electron-doping $x$, we show in Figs. 3 and 4 TOF INS measurements for $x = 0.096, 0.15, 0.18$ obtained on MERLIN using identical setup. The scattering intensity is normalized to absolute units of m barn/sr/meV/f.u. using a vanadium standard and the dashed boxes mark the AF Brillouin zone for the magnetic unit cell with single Fe$^{2+}$. For energies below 70 meV ($E = 8 \pm 1$ meV, Figs. 3(a), 3(c), 3(i); $16 \pm 2$ meV, Figs. 3(b), 3(f), 3(j); $48 \pm 4$ meV, Figs. 3(c), 3(g), 3(k); $60 \pm 10$ meV, Figs. 3(d), 3(h), 3(l), for $x = 0.096, 0.15$, and 0.18, respectively), spin excitations are transversely elongated ellipses centered around the in-plane AF ordering wave vectors $Q_{AF} = (\pm 1, 0)$ and $(0, \pm 1)$ due to the two twinned domains. The excitations become more transversely elongated and decrease in intensity with increasing $x$. On increasing energies to $E = 96 \pm 10$ meV [Figs. 4(a), 4(c), and 4(i) for $x = 0.096, 0.15$, and 0.18, respectively], spin excitations starts to split transversely away from the AF ordering wave vectors and become less doping dependent. For energies $E = 129 \pm 10$ meV [Figs. 4(b), 4(f), and 4(j), $E = 181 \pm 10$ meV [Figs. 4(c), 4(g), and 4(k)], and $E = 225 \pm 10$ meV [Figs. 4(d), 4(h), and 4(l)], spin excitations become rather similar, and are almost electron-doping independent.

Figure 5 compares the background subtracted scattering for the $E_i = 450, 250$, and 80 meV data projected in the wave vector ($Q = [1, K]$) and energy space for BaFe$_{2-x}$Ni$_x$As$_2$ with $x = 0.096, 0.15$, and 0.18. These incident beam energies were chosen to probe spin excitations at different energies. Figures 5(a), 5(d), and 5(g) show the $E_i = 450$ meV data for the $x = 0.096, 0.15$, and 0.18 samples, respectively. Similar data with $E_i = 250$ and 80 meV are shown in Figs. 5(b), 5(e), 5(h), and 5(c), 5(f), 5(j), where the solid lines are spin wave dispersions for BaFe$_{2-x}$As$_2$. While magnetic scattering clearly decreases with increasing doping at energies below 60 meV, they are virtually unchanged for energies above 100 meV, consistent with results in Figs. 3 and 4. To quantitatively determine the evolution of spin excitations for BaFe$_{2-x}$Ni$_x$As$_2$ with $x = 0.096, 0.15$, and 0.18, we show in Figs. 6 and 7 constant-energy cuts at different
Energies along the $[1, K]$ and $[H, 0]$ directions, respectively. At $E = 5 \pm 1$ [Fig. 6(a)] and $8 \pm 1$ meV [Fig. 6(b)], the commensurate spin excitations at $x = 0.96$ become weaker and transversely incommensurate on moving to $x = 0.15, 0.18$. For energies of $E = 16 \pm 2$ [Fig. 6(c)], $48 \pm 4$ [Fig. 6(d)], and $60 \pm 10$ [Fig. 6(e)] meV, the electron-doping induced spin excitation intensity reduction becomes smaller. Finally, there are no significant
difference between spin excitations of the parent compound and $x = 0.096, 0.15, 0.18$ at $E = 96 \pm 10$ [Fig. 6(f)], $129 \pm 10$ [Fig. 6(g)], $181 \pm 10$ [Fig. 6(h)], and $225 \pm 10$ meV [Fig. 6(i)]. Figures 7(a)-7(d) show the comparison of $[H, 0]$ scans for the $x = 0.096, 0.15,$ and $0.18$ samples at $E = 16 \pm 2, 48 \pm 4, 96 \pm 10,$ and $129 \pm 10$ meV. While the electron-doping evolution of the spin excitation intensity is consistent with cuts along the $[1, K]$
direction, they are commensurate at all energies probed.

To illustrate further the electron-doping evolution of the spin excitations in the overdoped regime, we compare constant-\(Q\) cuts in spin excitations of BaFe\(_{2-x}\)Ni\(_x\)As\(_2\) with \(x = 0, 0.096, 0.15,\) and 0.18 in Fig. 8\((a)\). The arrows in the inset of Fig. 8\((a)\) show the directions of the constant-\(Q\) cuts. At wave vectors near the Brillouin zone center at \(Q = (1, 0, 0.5)\) and \((1, 0, 2)\), electron-doping clearly suppresses the low-energy spin excitations. On increasing the wave vector to \(Q = (1, 0.35)\) and \((1, 0.5)\), there are much less difference in spin excitations of undoped and doped materials. Spin excitations form a broad peak near 100 meV in electron-overdoped samples similar to spin waves in parent compound.

Having established the electron-doping evolution of the overall spin excitations spectra, we now describe the effect of superconductivity on the low-energy spin excitations. From previous work, we know that a neutron spin resonance appears in the superconducting state of iron pnictide suggests that the mode energy decreases on warming to \(T_c\) and is coupled with the decreasing superconducting gap energy. Very recently, a sharp neutron spin resonance has been identified in superconducting...
NaFe$_{0.935}$Co$_{0.045}$As ($T_c = 18$ K) iron pniictide. Here, the resonance energy is again found to be weakly temperature dependent similar to the mode in Fe$_{1+y}$Te$_{1-x}$Se. In order to probe the detailed temperature dependence of the resonance in BaFe$_{1.944}$Ni$_{0.06}$As$_2$, we carried out TOF INS measurements on MERLIN with $E_i = 30$ meV at many temperatures below and above $T_c$. Following previous practice, we used the $T = 25$ K data as background and assumed that the net intensity gain near the AF ordering wave vector at lower temperatures is the resonance. Since an incident beam energy of $E_i = 30$ meV corresponds to $L \approx 1$ r.l.u. near the resonance energy of $E_r \approx 7$ meV, we can simultaneously probe the wave vector and energy dependence of the mode below $T_c$. Figures 9(a)-9(f) show the wave vector dependence of the temperature differences (the low-temperature data subtracts the data at 25 K) in spin excitations, $S(Q,E,T)-S(Q,E,T=25 \text{ K})$ with $E_r = 7 \pm 1$ meV, at $T=5, 11, 14, 16, 20 \text{ K}$, respectively. At $T = 5$ K, the superconductivity-induced resonance forms a transversely elongated ellipse in the $[H,K]$ plane centered at $Q_{AF} = (1,0)$ [Fig. 9(a)]. On warming to $T = 11$ K [Fig. 9(b)], 14 K [Fig. 9(c)], and 16 K [Fig. 9(d)], the resonance becomes weaker and broader along both the $[H,0]$ and $[1,K]$ directions. The resonance becomes almost indistinguishable from the background at $T = 20$ K [Fig. 9(f)].

Figures 9(g)-9(l) show the net magnetic scattering above the $T = 25$ K background projected onto the $[1,K]$ and energy space at different temperatures. At $T = 5$ K, we see a clear neutron spin resonance centered at $E_r = 7 \pm 1$ meV and $Q_{AF} = (1,0)$ [Fig. 9(g)]. Although the intensity of the resonance becomes progressively weaker on warming up to temperatures $T = 11$ K [Fig. 9(h)], 14 K [Fig. 9(i)], and 16 K [Fig. 9(j)], its peak position in energy appears to be fixed at $E \approx 7$ meV. On further warming to $T = 18$ K [Fig. 9(k)], one can still see a weak resonance near $E_r \approx 7$ meV. It becomes impossible to discern any magnetic signal at $T = 20$ K above the $T = 25$ K background scattering [Fig. 9(l)].

To quantitatively determine the temperature evolution of the resonance, we cut the images in Figs. 9(g)-9(l) along the energy direction by integrating wave vectors $0.8 < H < 1.2$ and $-0.2 < K < 0.2$ r.l.u. around $Q_{AF} = (1,0)$. Figure 10(a) shows the outcome at temperatures in Fig. 9(g)-9(l) and additional data taken at $T = 19$ K and 22 K. At all temperatures below $T_c = 20$ K, we see a well-defined resonance showing as positive scattering above background near $E = 7$ meV. There are no statistical differences in magnetic scattering for temperatures between $T = 20, 22$ K and 25 K. Figure 10(b) shows the wave vector cuts along the $[1,K]$ direction with energy-integration of $E = 7 \pm 1$ meV and $Q$-integration from $0.8 < H < 1.2$ at different temperatures. There are
FIG. 6: (Color online) Constant-energy cuts in the spin excitations of BaFe$_{2-x}$Ni$_x$As$_2$ along the [1, K] direction at different energies corresponding to those in Fig. 3 and Fig. 4, where the wave vector integration ranges are 0.9 < H < 1.1 for the K cuts and −0.1 < K < 0.1 for the H cuts. The solid lines are Gaussian fitting results for each doping and spin waves in the parent compound BaFe$_2$As$_2$.

well-defined peaks centered at the commensurate AF ordering wave vector for all probed temperatures. The solid lines are Gaussian fits to the data, which give peak intensity and FWHM of the spin excitations. Figure 10(c) shows similar wave vector cuts along the [H, 0] direction with Gaussian fits. The superconductivity-induced effects on wave vector dependence of the resonance along the [1, K] and [H, 0] directions are shown in Figs. 10(d) and 10(e), respectively. The data are peaked around the Q$_{AF}$ = (1, 0) wave vector and the solid lines are Gaussian fits on zero backgrounds.

Using parameters obtained from fits to the spin excitations spectra in Fig. 10, we can determine the temperature dependence of the resonance energy, intensity, and Q-widths along the [1, K] and [H, 0] directions. These results can be compared with temperature dependence of the superconducting gaps determined from other methods. From angle resolved photoemission spectroscopy experiments, it is well known that the electron-doped BaFe$_{2-x}$Te$_2$As$_2$ iron pnictides have the large isotropic superconducting gaps $\Delta_h$ located on the hole Fermi surface near the zone center position $\Gamma$ and the small gap $\Delta_e$ on one of the electron Fermi surfaces near $M$ point. The temperature dependence of the superconducting gaps decrease with increasing temperature and vanish at $T_c$. The pink solid line in Figure 11(a) shows temperature dependence of the sum of the electron and hole Fermi surface superconducting gaps $\Delta_e + \Delta_h$ obtained from point-contact andreev reflection measurements on BaFe$_{1.9}$Ni$_{0.1}$As$_2$. By comparing the temperature dependence of the resonance in the color contour plot and the solid points with the pink solid line, we see that the energy position of the resonance is weakly temperature dependent and does not follow the temperature dependence of the sum of the electron and hole pocket superconducting gaps. This is similar to the temperature dependence of the resonance in superconducting iron chalcogenide Fe$_{1+y}$Te$_{1-x}$Se$_2$ and NaFe$_{0.935}$Co$_{0.045}$As iron pnictide.

Figure 11(b) shows the temperature dependence of the
The solid lines are spin wave cuts in parent compound, $E = 96.10$ meV. The solid lines are identical cuts, from spin waves in BaFe$_{2-x}$Ni$_x$As$_2$. The spin excitations are commensurate at all energies probed. The resonance increases below $K < 0.15$, the spectral weight of the resonance mode is prevalent in both the electron underdoped BaFe$_{1.904}$Ni$_{0.096}$As$_2$ superconductor, where the transversely elongated spin excitations become slightly narrower below $T_c$. In a recent INS experiment on superconducting BaFe$_{1.928}$Ni$_{0.074}$As$_2$ $(T_c = 17$ K$)$, the resonance at $E_r = 6$ meV was found to have spin wave like dispersion across the transverse direction. In our TOF INS measurements for BaFe$_{1.904}$Ni$_{0.096}$As$_2$, this would correspond to a dispersive resonance along the transverse $[1, K]$ direction in Fig. 9(g). To see if we can detect the possible dispersion of the resonance, we cut the temperature difference plot in Fig. 9(g) along the $[1, K]$ direction in 1 meV interval. The outcome in Fig. 12 shows that the resonance indeed disperses outward for energies above $\sim 10$ meV along the transverse direction. This result, combined with earlier observation of incommensurate resonance in electron overdoped BaFe$_{1.85}$Ni$_{0.15}$As$_2$ indicate that the transversely dispersive resonance mode is prevalent in both the electron underdoped BaFe$_{2-x}$Ni$_x$As$_2$. At present, it is unclear how to understand the wave vector narrowing of the resonance below $T_c$ [Figs. 11(c) and 11(d)] at $E = 7$ meV and the dispersion of the mode at higher energies from Fermi surface nesting point of view.}

Having described the temperature, wave vector and energy dependence of the low-energy spin excitations in BaFe$_{1.904}$Ni$_{0.096}$As$_2$, we now discuss similar TOF INS measurements for BaFe$_{1.85}$Ni$_{0.15}$As$_2$. In previous triple-axis and TOF INS measurements on BaFe$_{1.85}$Ni$_{0.15}$As$_2$, an incommensurate neutron spin resonance has been identified. Figure 13(a) shows the temperature difference of spin excitations between 5 K and 20 K projected onto the energy and $[1, K]$ plane. By integrating wave vectors from $0.8 < H < 1.2$ and $-0.2 < K < 0.2$, we plot the energy dependence of the resonance in Figure 13(b). The mode energy is now at $E_r = 6.5$ meV compared with $E_r = 7$ meV for BaFe$_{1.904}$Ni$_{0.096}$As$_2$. Figure 13(c) shows a wave vector cut along the $[1, K]$ direction at $E = 6.5 \pm 1$ meV, which confirm the transverse incommensurate nature of the resonance. A similar cut along the $[H, 0]$ direction indicates that the mode is commensurate along the longitudinal direction [Fig. 13(d)].
FIG. 9: (Color online) (a-f) The wave vector dependence of the resonance in BaFe$_1$)$_{0.904}$Ni$_{0.096}$As$_2$ at $T = 5$ K, 11 K, 14 K, 16 K, 18 K and 20 K after subtracting the normal state data at 25 K. (g-l) Energy dependence of the two-dimensional slices along the $Q = [1, K]$ direction for the resonance at different temperatures. The mode essentially disappears around 20 K, but its peak positions are weakly temperature dependent.

measurements using 8 grams of sample. Using 25 grams of co-aligned single crystals with an incident neutron beam energy of $E_i = 20$ meV along the c-axis, we can now detect clear low-energy spin excitations at the AF wave vector positions on MERLIN. Figures 14(a) and 14(b) show spin excitation images projected onto the energy and $[1, K]$ plane at $T = T_c - 3 = 5$ K and $T = T_c + 2 = 10$ K, respectively. Consistent with the behavior of spin excitations at other Ni-doping levels, we see plumes of scattering stemming from $Q_{AF} = (1, 0)$. In the normal state (10 K), spin excitations are commensurate and centered at $Q_{AF} = (1, 0)$ from $E = 4$ meV to 9 meV [Fig. 14(b)]. On cooling to below $T_c$ (5 K), the scattering is enhanced between $E = 5$ meV and 7 meV [Fig. 14(a)]. The temperature difference plot in Figure 14(c) reveals evidence for incommensurate spin excitations.

Figures 14(d) and 14(e) show wave vector dependence of the spin excitations in the $[H, K]$ plane at the resonance energy $E_r = 5.5 \pm 1$ meV below and above $T_c$, respectively. In the normal state (10 K), spin excitations form transversely elongated ellipse commensurate with the underlying lattice [Fig. 14(e)]. On cooling to below $T_c$ (5 K), spin excitations at transversely incommensurate positions are enhanced [Fig. 14(d)]. The temperature differences between 5 K and 10 K reveal transversely incommensurate spin excitations marked by dashed circles [Fig. 14(f)].

To further probe the wave vector, energy, and temperature dependence of the magnetic excitations in BaFe$_{1.82}$Ni$_{0.18}$As$_2$, we show in Fig. 15(a) the energy dependence of the spin excitations near the AF ordering position integrated within the range of $-0.2 < K < 0.2$ and $0.8 < H < 1.2$ r.l.u. below and above $T_c$. The data reveals a small enhancement of the scattering below $T_c$ for energies around $E_r = 5.5$ meV. Figure 15(b) shows the temperature difference between 5 K and 10 K, and one can see a very weak resonance near $E_r = 5.5$ meV. Figure 15(c) shows cuts along the $[1, K]$ direction at $E = 5.5 \pm 1$ meV and $0.9 < H < 1.1$. The red circles are data at 10 K showing a commensurate peak centered at $Q_{AF} = (1, 0)$. The blue squares are identical cut at 5 K, which have more scattering at the incommensurate positions. The brown diamonds are the temperature difference plot which again reveal the incommensurate neutron spin resonance. Figure 15(d) shows similar cuts along the $[H, 0]$ direction. The scattering peaks at the commensurate AF ordering position and has no observable changes across $T_c$, as confirmed by the temper-
and $0.18$, using method described before \( \chi \) for BaFe\(_{1.904}\)Ni\(_{0.096}\)As\(_2\). Figure 16(a) shows the comparison of the energy dependent \( \chi''(E) \) for \( x = 0.0, 0.096, 0.15 \) and $0.18$, using method described before\( [20, 33] \). The solid line is the result for BaFe\(_{2-x}\)Ni\(_x\)As\(_2\).\(^{29}\) The energy dependence of the local susceptibility for BaFe\(_{1.904}\)Ni\(_{0.096}\)As\(_2\) is almost identical to that of BaFe\(_{1.9}\)Ni\(_{1}\)As\(_2\).\(^{29}\). On increasing the electron doping levels to $x = 0.15$ and $0.18$, we see a significant suppression of the local dynamic susceptibility for energies below $\sim$80 meV. Instead of forming clear peak at the resonance energy as in the case of BaFe\(_{1.904}\)Ni\(_{0.096}\)As\(_2\), the energy dependent \( \chi''(E) \) increases linearly with increasing energy and the superconductivity-induced resonance is not a visible peak in BaFe\(_{2-x}\)Ni\(_x\)As\(_2\) with $x = 0.15$ and $0.18$ [Fig. 16(a)]. For spin excitation energies above $\sim$80 meV, electron-doping to BaFe\(_2\)As\(_2\) appears to have little effect on the local dynamic susceptibility. These results are consistent with the notion that Fermi surface nesting and itinerant electrons are controlling the low-energy spin excitations while high-energy spin excitations arise from the local moments.\(^{[20, 108]}\) Upon further doping to electron-overdoped nonsuperconductor for BaFe\(_{2-x}\)Ni\(_x\)As\(_2\) with
$\Delta E \geq \Delta E_{0}$. Future work in this area might shed light on the relationship between the low-energy spin excitations and Fermi surface nesting.

Figure 16(b) shows the electron doping dependence of the dynamic spin-spin correlation lengths, obtained by Fourier transform of the $Q = [1, K]$ dependence of the spin dynamic susceptibility. As we can see from the Figure, electron doping from an optimally doped superconductor to electron-overdoped superconductor only appears to shorten the spin-spin correlation length for spin excitations at low-energies, and have little impact to the zone boundary spin excitations. To understand the impact of electron-doping to the total fluctuating magnetic moments, defined as $\langle m^{2} \rangle = \langle m^{2} \rangle = \langle |\chi^{0}(E)|^{2}dE \rangle = \langle 1 - \exp(-E/kT) \rangle$, we show in Fig. 16(c) the electron-doping dependence of $\langle m^{2} \rangle$ for BaFe$_{2-x}$Ni$_{x}$As$_{2}$ with $x = 0.0, 0.096, 0.1, 0.15, 0.18, 0.25$. We used $\langle m^{2} \rangle \approx 3.6 \mu_{B}^{2}/Fe$ for BaFe$_{2}$As$_{2}$ from a recent work with a value slightly larger than the earlier estimation of $\langle m^{2} \rangle \approx 3.17 \mu_{B}^{2}/Fe$. The $\langle m^{2} \rangle$ shows a linear decrease in value with increasing $x$. From the electron doping dependence of the local dynamic susceptibility $\chi^{0}(E)$ in Fig. 16(a), we see that the decreasing total moment $\langle m^{2} \rangle$ with increasing $x$ in BaFe$_{2-x}$Ni$_{x}$As$_{2}$ is due almost entirely to the reduction in spin excitations below $\sim 80$ meV.

Finally, Figure 16(d) shows the total spectral weight of spin resonance and the energy positions at $T = 5$ K, estimated from the superconductivity-induced spin excitation change, as a function of $T_{c}$. The resonance energy is linearly scaling with $T_{c}$, the same as previous results in cuprates and pnictides. As superconductivity ceases to exist for BaFe$_{2-x}$Ni$_{x}$As$_{2}$ with $x \rightarrow 0.25$, superconductivity-induced low-energy resonance also approaches zero, even though the high-energy spin excitations are not much affected. This is consistent with the notion that superconductivity requires itinerant electron-spin excitation coupling and the Fermi surface nesting.
driven low-energy spin excitations are important for superconductivity in electron-doped iron pnictides.

FIG. 14: (Color online) Temperature dependence of the spin excitations in BaFe$_{1.82}$Ni$_{0.18}$As$_2$ measured with $E_i = 20$ meV. (a,b,c) Energy dependence of the two-dimensional slices along the $Q = [1, K]$ direction at $T = 5$ K, 10 K, and their differences, respectively. (d,e,f) Wave vector dependence of the two-dimensional slices in the energy range $E = 5.5 \pm 1$ meV at 5 K and 10 K, and their difference, respectively. The dashed circles mark positions of incommensurate spin fluctuations.

IV. DISCUSSION AND CONCLUSIONS

By comparing the structure, phase diagram, and magnetic excitations in high-$T_c$ copper oxide, iron-based, and heavy Fermion superconductors, Scalapino concludes that spin fluctuation-mediated pairing is the common thread linking different classes unconventional superconductor. Within the framework of this picture, the superconducting condensation energy should be accounted for by the change in magnetic exchange energy $\Delta E_{ex}(T)$ between the normal ($N$) and superconducting ($S$) phases at zero temperature. For an isotropic $t$-$J$ model, $\Delta E_{ex}(T) = 2J[\langle S_{i+x} \cdot S_i \rangle_N - \langle S_{i+x} \cdot S_i \rangle_S]$, where $J$ is the nearest neighbor magnetic exchange coupling and $\langle S_{i+x} \cdot S_i \rangle$ is the magnetic scattering in absolute units at temperature $T$. If there are no changes in magnetic scattering between the normal and superconducting state, spin excitations should not contribute to the superconducting condensation energy. This is consistent with the observation that superconductivity-induced effect in spin excitations becomes very weaker in electron-overdoped iron pnictides with reduced $T_c$. While the total fluctuating moment $\langle m^2 \rangle$ only decreases slightly on moving from the AF parent compound BaFe$_2$As$_2$ to electron-overdoped nonsuperconducting BaFe$_{1.7}$Ni$_{0.3}$As$_2$ [Fig. 16(c)], the changes in resonance intensity appears to correlate with superconducting $T_c$ [Fig. 16(d)]. This suggests that the superconducting transition temperature in electron-doped iron pnictides is associated with the strength of the itinerant electron-low-energy spin excitations coupling or Fermi surface nesting conditions of the
FIG. 16: (Color online) (a) Energy dependence of the local dynamic susceptibility $\chi^a(E)$ for BaFe$_2-x$Ni$_x$As$_2$ with $x = 0, 0.096, 0.15, 0.18$ in the absolute units ($\mu_B^2 / eV/f.u.$). While the high energy spin excitations are doping independent, low-energy spin excitations ($E < 80$ meV) decreases with increasing electron doping. (b) Energy dependence of the dynamic spin-spin correlation lengths ($\xi$) for BaFe$_2-x$Ni$_x$As$_2$ with $x = 0, 0.096, 0.15, 0.18, 0.30$ obtained by Fourier transform of the constant-energy cuts along the [1, $K$] direction. (c) Ni-doping dependence of the total fluctuating moment. (d) The $T_c$ dependence of the resonance energy and its spectral weight.

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