Electron-doping evolution of the low-energy spin excitations in the iron arsenide BaFe$_{2-x}$Ni$_x$As$_2$ superconductors

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We use elastic and inelastic neutron scattering to systematically investigate the evolution of the low-energy spin excitations in the iron arsenide superconductor BaFe$_{2-x}$Ni$_x$As$_2$ as a function of nickel doping $x$. In the undoped state, BaFe$_2$As$_2$ exhibits a tetragonal-to-orthorhombic structural phase transition and simultaneously develops a collinear antiferromagnetic (AF) order below $T_N = 143$ K. Upon electron-doping of $x = 0.075$ to induce bulk superconductivity with $T_c = 12.3$ K, the AF ordering temperature reduces to $T_N \approx 58$ K. We show that the appearance of bulk superconductivity in BaFe$_{1.925}$Ni$_{0.075}$As$_2$ coincides with a dispersive neutron spin resonance in the spin excitation spectra, and a reduction in the static ordered moment. For optimally doped BaFe$_{1.9}$Ni$_{0.1}$As$_2$ ($T_c = 20$ K) and overdoped BaFe$_{1.65}$Ni$_{0.15}$As$_2$ ($T_c = 15$ K) superconductors, the static AF long-range order is completely suppressed and the spin excitation spectra are dominated by a resonance and spin-gap at lower energies. We determine the electron-doping dependence of the neutron spin resonance and spin gap energies, and demonstrate that the three-dimensional nature of the resonance survives into the overdoped regime. If spin excitations are important for superconductivity, these results would suggest that the three-dimensional character of the electronic superconducting gaps are prevalent throughout the phase diagram, and may be critical for superconductivity in these materials.

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1. INTRODUCTION

An experimental determination of the doping evolution of the spin excitations in iron arsenide superconductors is important for a comprehensive understanding of the role of magnetism in the superconductivity of these materials. Like high-transition temperature (high-$T_c$) copper oxides, the parent compounds of iron arsenide superconductors exhibit static antiferromagnetic (AF) long-range order with a collinear spin structure. Although there is currently no consensus on a microscopic mechanism for superconductivity, spin excitations have been postulated by several theories to play a crucial role in the electron pairing and superconductivity of these materials. In one class of unconventional microscopic theories for superconductivity, electron pairing in iron arsenide superconductors is mediated by quasiparticle excitations between sign reversed hole pockets around the $\Gamma$ point and the electron Fermi pockets around the $M$ point as shown in the inset of Fig. 1(a). If this is indeed the case, spin excitations in the superconducting state should have a collective mode called the neutron spin resonance, whose energy is at (or slightly less than) the addition of the hole and electron superconducting gap energies $E = |\Delta(k + Q) + \Delta(k)|$, where $Q$ is the AF ordering wavevector connecting the hole and electron Fermi pockets at the $\Gamma$ and $M$ points, respectively. Although recent inelastic neutron scattering experiments have found the neutron spin resonance for different iron-based superconductors consistent with this picture, a surprising result has been that the mode in the optimally doped BaFe$_{1.9}$Ni$_{0.1}$As$_2$ ($T_c = 20$ K) has three-dimensional character with clear dispersion along the $c$-axis, quite different from the two-dimensional nature of the resonance in copper oxide superconductors. If spin excitations are important for superconductivity in iron-arsenides, it would be interesting to systematically investigate the doping evolution of the resonance in BaFe$_{2-x}$Ni$_x$As$_2$, and determine if the three-dimensional nature of the mode is a general phenomenon or specific only to the optimally doped materials. Furthermore, since spin waves in the AF ordered parent compounds of (Ba,Sr,Ca)Fe$_2$As$_2$ have rather large anisotropy spin gaps at the AF zone center and the optimally doped superconducting samples are generally gapless, it would be important to see how spin waves in the parent compounds evolve as electrons are doped into the FeAs planes.

In this article, we report our inelastic neutron scattering studies of the low-energy spin excitations in electron-doped BaFe$_{2-x}$Ni$_x$As$_2$ with $x = 0.075, 0.15$ [Fig. 1(a)], and compare and contrast them with the spin excitations...
Ni-doping, the structural and magnetic phase transition temperatures of BaFe$_{1.925}$Ni$_{0.075}$As$_2$ become near 97 K and 91 K, respectively [41]. In addition, three dimensional spin waves in BaFe$_2$As$_2$ change into quasi-two-dimensional spin excitations for BaFe$_{1.96}$Ni$_{0.04}$As$_2$ with no evidence for the neutron spin resonance [41] or bulk superconductivity [4]. By increasing the Ni-doping $x$ to 0.075 to form BaFe$_{1.925}$Ni$_{0.075}$As$_2$ [Fig. 1(a)], bulk superconductivity appears at $T_c = 12.3$ K [Fig. 1(b)] and the Néel temperature of the material is now reduced to $T_N \approx 58$ K [Figs. 1(d) and 1(e)]. Our inelastic neutron scattering experiments show the presence of a three-dimensional neutron spin resonance with distinct energies at the AF wavevectors $Q = (0.5, 0.5, 0)$ and $(0.5, 0.5, 1)$, quite similar to that of the optimally doped BaFe$_{1.9}$Ni$_{0.1}$As$_2$ [21, 22]. The intensity gain of the mode below $T_c$ is compensated by opening a pseudo spin gap at lower energies and reduction in the static AF order [see inset in Fig. 1(d)].

To study the doping evolution of the resonance, we observed at optimal doping and in the lightly/undoped regime. Before Ni-doping, BaFe$_2$As$_2$ exhibits simultaneous structural and magnetic phase transitions below $T_s = T_N = 143$ K, changing the crystal lattice symmetry from the high-temperature tetragonal to the low-temperature orthorhombic phase [3]. Upon doping electrons via either Co or Ni substitution for Fe, the structural and magnetic phase transitions are separated [39, 40]. For $x = 0.04$
also carried out inelastic neutron scattering experiments on overdoped BaFe$_{1.85}$Ni$_{0.15}$As$_2$ \cite{24} and found that the energy of the mode is approximately proportional to $T_c$. Our elastic neutron scattering measurements indicate that the static antiferromagnetism has been completely suppressed, while the neutron spin resonance in the superconducting state exhibits similar dispersion along the c-axis as the underdoped and optimally doped materials. This suggests that the three-dimensional nature of the resonance energy and its associated superconducting gap energy $\Delta$ are prevalent throughout the superconducting electronic phase diagram. These results can also provide information needed for calculating the electron-doping dependence of the AF coupling between the layers, and estimating the doping dependence of the superconducting gap energy.

In two recent inelastic neutron scattering experiments on underdoped BaFe$_{1.906}$Co$_{0.094}$As$_2$ ($T_c = 15$ K) \cite{24} and BaFe$_{1.92}$Co$_{0.08}$As$_2$ ($T_c = 11$ K) \cite{25}, static AF order was found to coexist with superconductivity and cooling below $T_c$‘s in these samples induced a weak neutron spin resonance in the magnetic excitation spectra at the expense of elastic magnetic scattering \cite{24, 25}. For BaFe$_{2-x}$Ni$_x$As$_2$, bulk superconductivity appears only when $x \geq 0.05$ \cite{4}. To compare the electronic phase diagram of BaFe$_{2-x}$Ni$_x$As$_2$ with Co-doped materials and see the effect of superconductivity on the spin excitations, we chose to study underdoped BaFe$_{1.925}$Ni$_{0.075}$As$_2$ (where Ni concentration is nominal) and overdoped BaFe$_{1.85}$Ni$_{0.15}$As$_2$ superconductors. The temperature dependence of the susceptibility in Figures \ref{fig:1}(b) and \ref{fig:1}(c) show $T_c$‘s of 12.3 K and 15.5 K for BaFe$_{1.925}$Ni$_{0.075}$As$_2$ and BaFe$_{1.85}$Ni$_{0.15}$As$_2$, respectively, consistent with the overall electronic phase diagram from heat capacity measurements \cite{4}.

We grew single crystals of BaFe$_{2-x}$Ni$_x$As$_2$ with $x = 0.075, 0.15$ using the self-flux method \cite{3}. Our neutron scattering experiments were carried out on the HB-3, HB-1 thermal triple-axis spectrometers at the high-flux isotope reactor (HFIR), Oak Ridge National Laboratory \cite{33}: the BT-7 thermal triple-axis spectrometer at the NIST Center for Neutron Research \cite{22}; and the PANDA cold triple-axis spectrometer at the Forschungszentrum J"ulich (FZJ), Heinz Maier-Leibnitz (FZM-III), TU M"unchen \cite{21}. We defined the wave vector $Q$ at ($q_x$, $q_y$, $q_z$) as
\[ (H, K, L) = (q_a/2\pi, q_b/2\pi, q_c/2\pi) \] reciprocal lattice units (rlu) using the tetragonal nuclear unit cell, where \( a = 3.89 \text{ Å}, b = 3.89 \text{ Å}, \) and \( c = 12.77 \text{ Å}. \) We co-aligned about 6 grams for each of the \( z = 0.075, 0.15 \) samples of BaFe_{2-x}Ni_{x}As_2 in the \([H, H, L]\) horizontal scattering plane, and put our samples inside either a closed cycle refrigerator or a liquid He cryostat.

For thermal triple-axis measurements on HB-1, HB-3, and BT-7, we used pyrolytic graphite (PG) as monochromator and analyzer with typical collimations of open-40°-S-40°-120°′. The final neutron energy was chosen to be either \( E_f = 13.5 \text{ meV} \) or \( E_f = 14.7 \text{ meV} \) with a PG filter before the analyzer. For cold triple-axis measurements on PANDA, we chose final neutron energy of \( E_f = 5.0 \text{ meV} \) with a cooled Be filter in front of the analyzer. We used both horizontal and vertical focusing PG monochromator and analyzer with no collimators. We also used a \( E_f = 13.5 \text{ meV} \) setup with a PG filter in one of the PANDA measurements.

III. RESULTS AND DISCUSSIONS

We first describe our elastic and quasielastic neutron scattering results on the underdoped BaFe_{1.925}Ni_{0.075}As_2. Consistent with earlier results on underdoped BaFe_{1.906}Co_{0.094}As_2 \[24] and BaFe_{1.92}Co_{0.08}As_2 \[25], the AF structure of the BaFe_{1.925}Ni_{0.075}As_2 sample reported here is identical to the undoped parent compound but with a Néel \( T_N \approx 58 \text{ K} \) [Fig. 1(c)]. The temperature dependence of the quasielastic scattering at \( Q = (0.525, 0.525, 1) \) and \( E = 1.5 \text{ meV} \) shows a clear kink below \( \sim 58 \text{ K} \), thus confirming the Néel temperature of the system. The inset in Figure 1(d) shows the expanded temperature dependence of the AF Bragg peak intensity at \( Q = (0.5, 0.5, 3) \). The scattering decreases with decreasing temperature at the onset of \( T_c \), suggesting that the static moment competes with superconductivity similar to the Co-doped materials \[24, 25\].

To see if there is a neutron spin resonance mode in underdoped BaFe_{1.925}Ni_{0.075}As_2 and to compare its c-axis dispersion with optimally doped BaFe\(_{1.9}\)Ni\(_{0.1}\)As\(_2\) \[21\], we carried out constant-\( Q \) scans at \( Q = (0.5, 0.5, 0) \) and \( (0.5, 0.5, 1) \) above and below the superconducting transition temperature \( T_c \). Figure 2(a) shows the raw data collected on the HB-3 triple-axis spectrometer at the signal \( Q = (0.5, 0.5, 0) \) and background \( Q = (0.7, 0.7, 0) \) positions. There is clear intensity gain at \( Q = (0.5, 0.5, 0) \) near \( E = 7 \text{ meV} \) below \( T_c \) at the expense of spectral weight loss below \( \sim 4 \text{ meV} \). The temperature difference spectrum between 2 K and 20 K in Fig. 2(b) confirms the presence of the mode at \( E = 7 \text{ meV} \) below \( T_c \) and a reduction in spectral weight below 4 meV. The open squares in Fig. 2(a) show the energy dependence of the background scattering at \( Q = (0.7, 0.7, 0) \).

**FIG. 5.** (color online) (a) \( Q \)-scans along the \((H,H,1)\) direction at \( E = 2.5 \text{ meV} \) above and below \( T_c \) for BaFe\(_{1.925}\)Ni\(_{0.075}\)As\(_2\). (b) Temperature difference plot shows that the scattering at \( Q = (0.5, 0.5, 1) \) and \( E = 2.5 \text{ meV} \) decreases below \( T_c \). (c) Identical \( Q \)-scans across \( T_c \) at the resonance energy of \( E = 5.5 \text{ meV} \). The scattering enhances below \( T_c \). (d) Temperature difference plot between 2 K and 20 K, showing clear field-induced scattering below \( T_c \) at \( Q = (0.5, 0.5, 1) \).

Figures 3 summarizes constant-energy scans at \( E = 7 \text{ meV} \) and 3 meV along the \([H,H,0]\) direction. At the resonance energy, the scattering shows a well-defined peak centered at \( Q = (0.5, 0.5, 0) \) that increases in intensity below \( T_c \) [Fig. 3(a)]. Figure 3(b) shows the temperature difference plot which confirms that the intensity gain below \( T_c \) in Fig. 2(a) occurs at \( Q = (0.5, 0.5, 0) \). Similarly, constant-energy scans at \( E = 3 \text{ meV} \) above and below \( T_c \) in Fig. 3(c) reveal clear normal state magnetic scattering that is not completely suppressed (gapped) below \( T_c \), at least with the energy resolution afforded with these thermal triple-axis measurements. Figure 2(c) shows our esti-
The scattering increases in intensity below $T_c$ of 12.3 K.

FIG. 6. (color online) (a) Temperature dependence of the $E = 7$ meV scattering at $Q = (0.5, 0.5, 0)$ for BaFe$_{1.925}$Ni$_{0.075}$As$_2$. The scattering increases in intensity below $T_c$.

The motion of the energy dependence of the dynamic susceptibility $\chi''(Q, \omega)$ above and below $T_c$, obtained by subtracting the background and correcting for the Bose population factor using $\chi''(Q, \omega) = [1 - \exp(-\hbar\omega/k_BT)]S(Q, \omega)$, where $E = \hbar\omega$. While the normal state susceptibility appears to increase linearly with energy, superconductivity rearranges the spectrum, creating a (pseudo) spin gap below 4 meV and a neutron spin resonance at $E = 7$ meV for in-phase spin fluctuations along the $c$-axis ($L = 0$).

To investigate the behavior for the out-of-phase spin fluctuations along the $c$-axis, we plot in Fig. 4(b) constant-$Q$ scans at $Q = (0.5, 0.5, 1)$ above and below $T_c$. Figure 4(b) shows the temperature difference plot, and a comparison of Fig. 4(b) and Fig. 2(b) immediately reveals that the neutron spin resonance has moved from $E = 7$ meV at $Q = (0.5, 0.5, 0)$ to $E = 5$ meV at $Q = (0.5, 0.5, 1)$. Note in particular that for $Q = (0.5, 0.5, 0)$ there is essentially no change with temperature for the scattering at 5 meV (Fig. 2), where the maximum in intensity occurs at the $Q = (0.5, 0.5, 1)$ position. This is compelling evidence that the neutron spin resonance is dispersive for both underdoped and optimally doped BaFe$_{2-x}$Ni$_x$As$_2$.

Figure 5 shows constant-energy scans along the $[H, H, 1]$ direction and the temperature difference data between 2 K and 20 K for $E = 2.5, 5.5$ meV. Similar to the $[H, H, 0]$ scans in Fig. 3, we find that superconductivity only reduces but does not completely suppress the magnetic scattering at $E = 2.5$ meV [Figs. 5(a) and 5(b)]. Similarly, the scattering near the resonance energy at $E = 5.5$ meV shows a clear increase below $T_c$. To test if the intensity gain at $E = 7$ and $Q = (0.5, 0.5, 0)$ is responding to superconductivity, we show in Fig. 6 the temperature dependence of the scattering, which clearly increases below $T_c$ consistent with the temperature dependence of the neutron spin resonance [19, 22, 24, 26].

To determine the $c$-axis modulation of the spin excitations at different temperatures and energies, we show in Figs. 7, 8, and 9 constant-energy scans along the $Q = (0.5, 0.5, L)$ (signal) and $(0.7, 0.7, L)$ (background) directions at $E = 3$ meV and various temperatures. (a) The scattering at 2 K and 20 K shows well-defined peaks centered at $L = \pm 1$. Fourier transform of these peaks give $c$-axis spin-spin correlation length of ~14 Å. (b) Signal and background Scattering at 2 K. The solid line is Gaussian fit to the data. The dashed line shows the expected magnetic scattering at $Q = (0.5, 0.5, 3)$ assuming Fe$^{2+}$ form factor. The absence of clear peaks at $L = \pm 3$ suggests that magnetic scattering in BaFe$_{1.925}$Ni$_{0.075}$As$_2$ damps out much faster than expected. (c) Similar scans at 50 K and 70 K. (d) The solid lines in the Figure show the effect of Bose population factor as a function of increasing temperature if one normalizes the magnetic scattering above background at 2 K. The magnetic intensity changes in the system clearly does not obey the Bose statistics, indicating that the scattering is not spin waves.

FIG. 7. (color online) Constant-energy scans along the $(0.5, 0.5, L)$ (signal) and $(0.7, 0.7, L)$ (background) directions at $E = 3$ meV and various temperatures. (a) The scattering at 2 K and 20 K shows well-defined peaks centered at $L = \pm 1$. Fourier transform of these peaks give $c$-axis spin-spin correlation length of ~14 Å. (b) Signal and background Scattering at 2 K. The solid line is Gaussian fit to the data. The dashed line shows the expected magnetic scattering at $Q = (0.5, 0.5, 3)$ assuming Fe$^{2+}$ form factor. The absence of clear peaks at $L = \pm 3$ suggests that magnetic scattering in BaFe$_{1.925}$Ni$_{0.075}$As$_2$ damps out much faster than expected. (c) Similar scans at 50 K and 70 K. (d) The solid lines in the Figure show the effect of Bose population factor as a function of increasing temperature if one normalizes the magnetic scattering above background at 2 K. The magnetic intensity changes in the system clearly does not obey the Bose statistics, indicating that the scattering is not spin waves.
above the background. Gaussian fits to these peaks give a intensity but the counting below to be affected [Fig. 7(c)]. To test if the magnetic scattering at 50 K. The solid line shows the expected $L$-dependence of the magnetic scattering assuming simple Fe$^{2+}$ form factor, which clearly fails to describe the data.

FIG. 8. (color online) Constant-energy scans along the $(0.5,0.5,L)$ (signal) and $(0.7,0.7,L)$ (background) directions at the resonance energy of $E = 5.5$ meV and various temperatures. (a) The $c$-axis scattering at 70 K. (b) The signal and background scattering at 50 K. The data again show clear peaks at $L = \pm 1$ but much damped peaks at $L = \pm 3$. (c) Signal and background scattering at 2 K and 20 K. Superconductivity clearly enhances magnetic scattering at $L = 0$ and $L = \pm 1$. The spin-spin correlation length is again about 14 Å and is weakly temperature dependent in the probed temperature range (2 K to 70 K).

similar to that of $f(Q_z) = F^2(Q)[S_0+S_1\exp(-(L-L_0)^2/2\sigma^2)]$, where $F(Q)$ is the magnetic form factor \cite{43}, $L = Q_z c/2\pi$, $L_0 = \pm 1,\pm 3,\cdots$, $\sigma$ is the width of the Gaussian which gives the correlation length of the spin excitations along the $c$-axis, $S_0$ and $S_1$ are fitting parameters for constant magnetic rod scattering along any $L$ and maximum magnetic intensity at odd values of $L$, respectively.

Figure 7 shows the temperature dependence of the magnetic scattering at $E = 3$ meV along the $c$-axis. Starting from 2 K and 20 K in Fig. 7(a), we see two clear peaks centered around $Q = (0.5,0.5,1)$ and $(0.5,0.5,-1)$ above the background. Gaussian fits to these peaks give a $c$-axis spin correlation length of $\sim 14$ Å [Fig. 7(b)]. Upon increasing temperature to 50 K and 70 K, the magnetic peaks at $Q = (0.5,0.5,1)$ and $(0.5,0.5,-1)$ reduce in intensity but the $c$-axis spin-spin correlations appear not to be affected [Fig. 7(c)]. To test if the magnetic scattering below $T_N$ simply follows the Bose population factor as expected for simple spin waves excitations, we show in Fig. 7(d) the temperature dependence of the magnetic scattering normalized to the scattering at 2 K. The observed magnetic scattering clearly does not obey the Bose population factor, suggesting that the spin excitations in the doped materials are not simple spin waves, in contrast to the (undoped) parent compounds.

To probe the $L$-dependence of the magnetic scattering at the neutron spin resonance energies, we show in Figs. 8 and 9 constant-energy scans at $E = 5.5$ meV and 7 meV, respectively. Consistent with constant-energy scan data at $E = 3$ meV (Fig. 7), the magnetic scattering is still centered at $L = \pm 1,\pm 3$ positions and superconductivity has a relatively small effect on the overall magnetic scattering. Comparison of Figs. 2, 4, and 8, 9 reveals that the neutron spin resonance so clearly illustrated in the temperature difference scattering is a rather subtle effect that occurs at both $L = 0$ and $L = 1$ [Figs. 8(c) and 9(a)]. On warming to 50 K and 70 K, the magnetic scattering decreases but the $c$-axis spin correlation length of $\sim 14$ Å appears to be fairly temperature independent. The solid lines in Fig. 8(a) and 9(b) show Gaussian fits to the data which indicates that the decrease in the magnetic scattering along the $c$-axis direction falls off faster than just the Fe$^{2+}$ form factor \cite{43}. If we assume that the spins prefer to lie in the $a$-$b$ plane as is the case for the AF undoped system, an additional reduction in intensity is expected due to the neutron spin-Fe-spin orientation factor and this brings the curve in reasonable agreement with the data.

Having described our comprehensive measurements on the underdoped BaFe$_{1.925}$Ni$_{0.075}$As$_2$, we now discuss in—

FIG. 9. (color online) $L$-dependence of the magnetic scattering for BaFe$_{1.925}$Ni$_{0.075}$As$_2$. (a) Identical scans as Fig. 8 except we now change the excitation energy to $E = 7$ meV. Comparison of the 2 K and 20 K data here indicates that the magnetic scattering enhancement below $T_c$ at $L = 0$ is larger than that at $L = 1$. (b) Signal and background scattering at 50 K. The data again show clear dependence of the magnetic scattering assuming simple Fe$^{2+}$ form factor, which clearly fails to describe the data.
elastic neutron scattering experiments on the overdoped BaFe$_{1.85}$Ni$_{0.15}$As$_2$ [Figs. 1(a), 1(c)], where the static AF order is completely suppressed. These measurements were carried out on the PANDA cold triple-axis spectrometer. Figure 10 summarizes the constant-$Q$ scans at $Q = (0.5, 0.5, 1)$ and $(0.5, 0.5, 0)$ below and above $T_c$. Using $E_f = 5$ meV, we find in Figs. 10(a) and 10(b) that the neutron spin resonance occurs at $E = 6$ meV for $Q = (0.5, 0.5, 1)$. Similar scans at $Q = (0.5, 0.5, 0)$ reveal clear scattering intensity enhancement above $E = 5$ meV [Figs. 10(c) and 10(d)]. However, kinematic constraints with the $E_f = 5$ meV spectrometer configuration did not allow a conclusive determination of the resonance energy. Figure 10(e) shows identical scans carried out with $E_f = 13.5$ meV. Inspection of Figs. 10(e) and 10(f) indicates that the resonance energy at $Q = (0.5, 0.5, 0)$ is now shifted to $E = 8$ meV. If we assume that the negative scattering in the temperature difference spectra of Figs. 10(b) and 10(d) gives the onset of the spin gap (assuming background scattering is temperature independent between 2 K and 20 K), these results suggest that the spin gap at $Q = (0.5, 0.5, 0)$ is larger than that at $Q = (0.5, 0.5, 1)$, consistent with the previous conclusions.
FIG. 12. (color online) Summary of electron-doping dependence of the neutron spin resonance energies at \(Q = (0.5, 0.5, 0)\) and \((0.5, 0.5, 1)\) as a function of \(T_c\). The data for BaFe\(_{2−x}\)Ni\(_x\)As\(_2\) were from Refs. 21, 22, 24, 44 and present work. The data for BaFe\(_{2−x}\)Ni\(_x\)As\(_2\) were from Refs. 21, 24–26. The solid lines are linear fits to the data.

To determine if the low-energy spin excitations have a clean spin gap like those of the optimally doped BaFe\(_{1.925}\)Ni\(_{0.15}\)As\(_2\) [21, 22], we took constant-energy scans along the \([H, H, 0]\) and \([H, H, 1]\) directions above and below \(T_c\) for \(E = 1\) meV. Figures 11(a) and 11(b) show that spin excitations of BaFe\(_{1.85}\)Ni\(_{0.15}\)As\(_2\) are gapless in the normal state for both \(L = 0\) and 1 rlu, but open a clean gap below \(T_c\) at \(E = 1\) meV. These results are similar to the optimally Ni-doped material, but are clearly different from underdoped BaFe\(_{1.925}\)Ni\(_{0.075}\)As\(_2\) where there are no clean spin gap at \(E = 2.5\) meV [Fig. 5(a)]. Figures 11(e) and 11(f) show wave-vector scans at the expected resonance energies for \(Q = (0.5, 0.5, 0)\) and \((0.5, 0.5, 1)\). In both cases, we find clear intensity enhancement below \(T_c\). The temperature dependent scattering at \(Q = (0.5, 0.5, 0)\) and \(E = 8\) meV in Fig. 11(g) shows clear order-parameter-like intensity increase below \(T_c\), thus confirming the neutron spin resonance [19–22, 24–26].

Finally, we summarize in Figure 12 the electron-doping dependence of the neutron spin resonance at \(Q = (0.5, 0.5, 0)\) and \((0.5, 0.5, 1)\) as a function of \(T_c\) for both BaFe\(_{2−x}\)Ni\(_x\)As\(_2\) [21, 22, 41, 44] and BaFe\(_{2−x}\)Co\(_x\)As\(_2\) [20, 24, 26]. For copper oxide high-\(T_c\) superconductors, one of the hallmarks of the resonance is that its energy is proportional to \(T_c\) over a very wide temperature range [31, 32]. Since BaFe\(_{2−x}\)Ni\(_x\)As\(_2\) has two resonances at distinctively different energies, its dispersion along the \(c\)-axis is related to the superconducting gap \(\Delta_0\) and its deviation \(\delta\) via \(E(Q_z) \sim 2\Delta_0 - 2\delta|\sin(Q_z/2)|\) [21]. The observation of a linear relationship for both mode energies and \(T_c\) suggests that \(\delta/\Delta_0 = \omega(0.5, 0.5, 0) - \omega(0.5, 0.5, 1))/\omega(0.5, 0.5, 0)\) is approximately 0.28 and weakly Ni-doping dependent. Therefore, the ratio of interplane \((J_{//})\) and intraplane \((J_{\perp})\) AF coupling, \(J_{\perp}/J_{//}\), is weakly electron-doping dependent assuming that the values of \(\Delta_0\) and \(\delta\) are proportional to \(J_{//}\) and \(J_{\perp}\), respectively.

We now discuss the physical interpretation of the above results. In the theory of spin-fluctuation-mediated superconductivity [4, 12], the electron pairing arises from sign-reversed S-wave interband scattering between hole pockets centered at the \(\Gamma\) point and electron pockets at the \(M\) points [inset in Fig. 1(a)] [15, 18]. One of the consequences of such electron-hole pocket excitations is to induce a resonance peak at the AF ordering wave vector \(Q = (0.5, 0.5, 0)\) in the spin excitations spectrum. In the strictly two-dimensional model, the energy of the resonance is at (or slightly less than) the addition of hole \((\Delta_0^h)\) and electron \((\Delta_0^e)\) superconducting gap energies \((\Delta_0 = \Delta_0^h + \Delta_0^e)\). Our previous finding of three-dimensionality of the resonance in optimally doped BaFe\(_{1.925}\)Ni\(_{0.15}\)As\(_2\) [21, 22] suggests that the superconducting gap energy \(\Delta_0\) should be three-dimensional as well and sensitive on the \(Q\) values along the \(c\)-axis. The new results reported in the present paper on underdoped BaFe\(_{1.925}\)Ni\(_{0.075}\)As\(_2\) and overdoped BaFe\(_{1.85}\)Ni\(_{0.15}\)As\(_2\) confirm the earlier conclusion, and reveal that the three-dimensional nature of the superconducting gap is prevalent throughout the superconducting dome. If spin excitations are mediating the electron pairing for superconductivity, these results would suggest that the AF exchange coupling along the \(c\)-axis \((J_{\perp})\) contributes significantly to the electron pairing. Although the overall spin excitations as a function of increasing electron doping transform into quasi two-dimensional spin excitations rather rapidly as demonstrated by the disappearing anisotropic spin gaps at \(Q = (0.5, 0.5, 0)\) and \((0.5, 0.5, 1)\) with increasing Ni-doping [44], the superconductivity-induced resonance retains its three-dimensional character even in the overdoped regime. This means that the superconducting electronic gaps in the iron-arsenic based materials are three-dimensional and quite different from that of the copper oxide superconductors.

IV. CONCLUSIONS

In summary, we have determined the doping evolution of the low-energy spin excitations in BaFe\(_{2−x}\)Ni\(_x\)As\(_2\) for both underdoped and overdoped superconductors. In underdoped BaFe\(_{1.925}\)Ni\(_{0.075}\)As\(_2\) we find that the appearance of bulk superconductivity is associated with the appearance of a weak three-dimensional neutron spin resonance. The spectral weight gain of the resonance below \(T_c\) is a rather small portion of the overall normal state magnetic scattering, and is compensated by opening a weak pseudo spin gap and reduction in static magnetic moment. Our Ni-doping dependent investigation of the spin gap and neutron spin resonance reveals that the three-dimensional nature of the mode found earlier for the optimally doped sample is a universal property.
of Ni-doped superconductors. These results in turn suggest that AF spin excitations between the layers are also important for the superconductivity of these materials.

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