Antiferromagnetism is relevant to high temperature (high-$T_c$) superconductivity in copper oxides and iron arsenides because superconductivity arises from electron- or hole-doping of their static antiferromagnetic (AF) ordered parent compounds \cite{Harriger1, Schneidewind1, Li1, Zhao1, Wei1, Lu1, Dong1, Zhou1, Zhao2, Hu1, Li1, Schneidewind2}. In the case of cuprates, spin waves of the parent materials can be very well described by a local moment Heisenberg Hamiltonian and spin excitations in optimally doped superconductors are dominated by a neutron spin resonance centered at the AF ordering wavevector $[1, 2, 3]$. For undoped iron arsenides such as AF$_{2}$As$_{2}$ ($A=$Ba, Sr, Ca) with a spin structure of Fig. 1a \cite{Cry, Nima}, spin waves consist of a large anisotropy gap at the AF zone center $|\Delta (1, 0, 1)| \approx 9.8$ meV and excitations extend up to $\approx 200$ meV \cite{Cry, Schneidewind2, Li1, Zhao1, Wei1}. For optimally doped superconductors \cite{Cry, Schneidewind2, Li1, Zhao1, Wei1}, the gapped spin wave excitations were replaced by a gapless continuum of scattering in the normal state and a neutron spin resonance below $T_c$ \cite{Cry, Schneidewind2, Li1, Zhao1, Wei1}. Since spin fluctuations may play a crucial role in the electron pairing and superconductivity of iron arsenides \cite{Cry, Schneidewind2, Li1, Zhao1, Wei1}, it is imperative to determine how the spin dynamics of the undoped AF parent compounds evolve as they are tuned toward optimally doped superconductivity by electron or hole doping.

In the undoped state, BaFe$_2$As$_2$ exhibits simultaneous structural and magnetic phase transitions below $T_c = T_N = 143$ K, changing the crystal lattice symmetry from the high-temperature tetragonal to low-temperature orthorhombic phase \cite{Cry}. Upon Co-doping to induce electrons onto the FeAs plane, the combined AF and structural phase transitions were split into two distinct transitions and the electronic phase diagram in the lower Co-doping region displays coexisting static AF order with the superconductivity \cite{Pratt1, Pratt2}. Although recent neutron scattering experiments confirmed that the upper transition is structural and the AF order occurs at lower temperature \cite{Pratt1, Pratt2, Zhaol}, its microscopic origin is still unknown. More importantly, it is unclear what happens to the spin waves of BaFe$_2$As$_2$ when electrons are doped into these materials. While Pratt et al. \cite{Pratt1} reported gapless normal state spin excitations for BaFe$_{1.96}$Co$_{0.04}$As$_2$, measurements on BaFe$_{1.96}$Co$_{0.08}$As$_2$ suggest gapped normal state excitations \cite{Pratt2}. On cooling below $T_c$, both materials reveal a reduction in the static ordered AF moment and the appearance of a spin resonance \cite{Pratt1, Pratt2}.

To compare with the results obtained on Co-doped BaFe$_2$As$_2$ \cite{Pratt1, Pratt2, Zhaol, Pratt3, Pratt4}, we carried out neutron scattering experiments on Ni-doped BaFe$_{1.96}$Ni$_{0.04}$As$_2$ ($T_c \approx 15$ K, Figs. 1c,1d) \cite{Zhaol}. In contrast to the results on Co-doped materials \cite{Pratt1, Pratt2}, we find that the static AF order and spin excitations in BaFe$_{1.96}$Ni$_{0.04}$As$_2$ do not respond to the occurrence of superconductivity. Instead, the effect of electron-doping is to significantly reduce the $c$-axis exchange coupling and change the three-dimensional (3D) spin waves of BaFe$_2$As$_2$ into quasi two-dimensional (2D). These results suggest that the separated structural/magnetic phase transition and the appearance of bulk superconductivity upon doping may be associated with the diminishing spin anisotropy gap and the 3D to 2D transition of the spin excitations.

Using the self-flux method \cite{Zhaol}, we grew a $\sim 1$ gram single crystal of BaFe$_{1.96}$Ni$_{0.04}$As$_2$ with an in-plane and out-of-plane mosaic of 1.74° and 2.20° full-width at half maximum (FWHM), respectively. We defined the wave vector $Q$ at $(q_x, q_y, q_z)$ as $(H, K, L) = (q_xa/2\pi, q_yb/2\pi, q_zc/2\pi)$ reciprocal lattice units (rlu) using the orthorhombic magnetic unit cell (space group Fmmm), where $a = 5.5$ Å,
FIG. 1: (color online). (a) Diagram of the parent compound BaFe$_2$As$_2$ with Fe spin ordering and magnetic exchange couplings depicted. We use the same unit cell for BaFe$_{1.96}$Ni$_{0.04}$As$_2$. (b) Electronic phase diagram from Ref. 7. (c) Temperature dependence of the Meissner and shielding signals on a small crystal (field cooled $4\pi\chi = -0.001$ at 4.5 K). (d) Temperature dependence of the structural distortion of the lattice as determined by tracking the width of the $(200)$ nuclear Bragg peak using $\lambda/2$ scattering without Be filter. (e) Magnetic order parameter determined by $Q$-scans around $(101)$ magnetic Bragg peak above background. The solid line shows order parameter fit using $\phi^2 \propto (1 - T/T_N)^{2B}$ with $T_N = 91.3 \pm 0.7$ K and $\beta = 0.3 \pm 0.02$.

$B = 5.4$ Å, and $c = 12.77$ Å. We performed our neutron scattering experiment on the PANDA cold triple-axis spectrometer at the Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II), TU Munchen, Germany as described earlier. Our sample was aligned in the $[H, 0, L]$ zone inside a closed cycle refrigerator.

Figures 1c and 1d show the resistivity and susceptibility data. The resistivity shows clear anomalies near 97 K and 91 K before superconductivity sets in below $\sim$15 K (Fig. 1c). Although the presence of superconductivity below $T_c \approx 15$ K is confirmed in the susceptibility measurement (Fig. 1d), the weak Meissner effect suggests superconducting volume fraction of less than 0.2%.

Similar to Co-doped BaFe$_2$As$_2$, spin waves have an anisotropy gap about 8 meV at $Q = (1, 0, 1)$ [$\Delta(1, 0, 1) = 8$ meV. For optimally Co and Ni doped materials, spin excitations are gapless in the normal state and superconductivity-induced spin gaps open below $T_c$. Figure 2a shows the constant-$Q$ scans at the $Q = (1, 0, 1)$ (signal) and $Q = (1.2, 0, 1)$ (background)
2D nature of the excitations. The spin gap is not affected by superconductivity. Fig. 4h shows Q-scans along the $[H,0,0]$ direction at different temperatures. The scattering shows no anomaly across $T_c$ but clearly peak at $T_N$. (b) Temperature dependence of the scattering at 4 meV and $Q = (1,0,0)$ again peaks at $T_N$.

Further evidences for 2D spin excitations in BaFe$_{1.96}$Ni$_{0.04}$As$_2$ are summarized in Fig. 3. Assuming spin excitations in BaFe$_{2-x}$Ni$_x$As$_2$ can be described by an effective Heisenberg Hamiltonian, the spin anisotropy gaps at $Q = (1,0,1)$ and $Q = (1,0,0)$ are $\Delta(1,0,1) = 2S[(J_{1a} + 2J_2 + J_c + J_s)^2 - (J_c + J_{1a} + 2J_2)^2]^{1/2}$ and $\Delta(1,0,0) = 2S[(2J_{1a} + 4J_2 + J_c)(2J_c + J_s)]^{1/2}$, respectively [11, 12, 14, 15]. Here $S$ is the magnetic spin ($= 1$); $J_{1a}$, $J_2$, $J_c$, $J_s$ are effective in-plane nearest-neighbor, next nearest-neighbor, c-axis, and magnetic single ion anisotropy couplings, respectively (Fig. 1a). For BaFe$_{2}$As$_2$, we estimate $\Delta(1,0,1) = 7.8$ meV and $\Delta(1,0,0) = 20.2$ meV assuming $J_{1a} = 36$, $J_2 = 18$, $J_c = 0.3$, $J_s = 0.106$ meV [12, 14, 15]. Upon electron doping to form BaFe$_{1.96}$Ni$_{0.04}$As$_2$, these spin gap values have been reduced to $\Delta(1,0,1) = 2$ meV and $\Delta(1,0,0) = 4$ meV (Figs. 2b and 3b). Since such electron-doping hardly changes the in-plane Q-scan widths compared to that of the undoped BaFe$_{2}$As$_2$ (Figs. 2e-g, 3e,3g) [12, 14], it should only slightly modify the in-plane exchange couplings. Assuming that $J_{1a}$ and $J_2$ are unchanged in BaFe$_{1.96}$Ni$_{0.04}$As$_2$, the observed $\Delta(1,0,1) = 2$ meV and $\Delta(1,0,0) = 4$ meV would correspond to $J_c = 0.01$ meV and $J_s = 0.007$ meV, suggesting a rapid suppression of c-axis exchange coupling and magnetic single ion anisotropy with electron doping.

In Ref. [32], it was argued that spin anisotropy for
BaFe$_1.92$Co$_{0.08}$As$_2$ is similar to that of the BaFe$_2$As$_2$, meaning that the reduction in spin gap at $Q = (1,0,0)$ arises mostly from reduced $J_1$ and $J_2$. Assuming the best fitted values of $S(J_{1a} + 2J_2) = 32$ meV and $SJ_c = 0.34$ meV [22], we expect $\Delta(1,0,1) = 5.5$ meV and $\Delta(1,0,0) = 14.2$ meV with $SJ_s = 0.106$ meV. These values are clearly different from the observation. Even if we assume all exchange couplings to reduce by 50% upon electron-doping with $S(J_{1a} + 2J_2) = 32$ meV, $SJ_c = 0.15$ meV, and $SJ_s = 0.05$ meV, we still find $\Delta(1,0,1) = 3.8$ and $\Delta(1,0,0) = 10$ meV. This suggests that the large reduction in the $\Delta(1,0,0)$ gap values upon electron doping is due to the reduced $J_c$ and three-dimensionality of the system.

To determine the temperature dependence of $\Delta(1,0,0)$, we show in Fig. 3c the observed scattering at the signal $Q = (1,0,0)$ and background $(1,4,0,0)$ positions at several temperatures. Figure 3d plots the estimated $\chi''(Q,\omega)$. Comparing Fig. 3d with Fig. 3b, the 4 meV spin gap $\chi''(Q,\omega)$ at 18 K vanishes upon warming to above 60 K. These results are confirmed by $Q$-scans at 4 meV along the $[H,0,0]$ direction (Fig. 3e). While scans at 2 K and 18 K are featureless, the scattering at 86 K and 100 K shows clear peaks centered at $Q = (1,0,0)$. For $Q$-scans at 6 meV, the scattering shows well-defined peaks at all temperatures (Fig. 3g). Converting these data into $\chi''(Q,\omega)$ in Fig. 3h confirms the results of Fig. 3d.

Finally, we show in Fig. 4a the temperature dependence of the 1 meV scattering at the $Q = (1,0,0)$ (signal) and $Q = (1,4,0,0)$ (background) positions. While the background scattering only increases slightly with increasing temperature and shows no anomaly across $T_N$, the scattering at $Q = (1,0,0)$ clearly peaks at $T_N$. $Q$-scans along the $[H,0,0]$ direction at 1 meV confirm these results (the inset of Fig. 4a). Temperature dependence of the scattering at 4 meV and $Q = (1,0,1)$ show similar behavior (Fig. 4b). These results suggest that the disappearing $\Delta(1,0,1)$ and $\Delta(1,0,0)$ gaps near $T_N$ arise from critical scattering associated with the static AF order.

To understand the separated structural and magnetic phase transitions for BaFe$_{1-2x}$Co$_x$Ni$_{0.4}$As$_2$, we note that in an effective $J_1$-$J_2$-$J_c$ model [22, 23], the separation of the lattice and magnetic transition temperatures is controlled by the value of $J_c$. There is only one transition temperature when $J_c$ is large. A finite separation between the two transition temperatures occurs when $J_c/J_2$ is reduced to the order of 10$^{-3}$. Our experimental result of $J_c/J_2 \sim 0.5 \times 10^{-3}$ is consistent with this picture. To quantitatively estimate the reduced $T_N$ due to the smaller $J_c$, we note that $T_N \sim J_2/[\ln(J_2/J_c)]$ [22]. Let $J_0$ be the magnetic exchange values for the parent compounds, we can write $T_N^0/T_N = a[\ln(a) + \ln(b) + \ln(c)]/\ln(c)$, where $a = J_0/J_c$, $b = J_0/J_c$, $c = J_0/J_c$. Using the experimental values of the exchange coupling parameters determined earlier, we obtain $(J_2/J_2^0) = (1/a) \sim 0.87$, which is self-consistent with our suggestion that upon doping, the coupling between layers $J_c$ is dramatically reduced while the change of the in-plane magnetic exchange coupling is small. These results provide a natural and consistent interpretation for our experimental observations.

In summary, we have shown that the most dramatic effect of electron-doping in BaFe$_2$As$_2$ is to transform the 3D anisotropic spin waves into 2D spin excitations. These results suggest that reduced dimensionality in spin excitations of iron arsenides is important for the separated structural/magnetic phase transition and the occurrence of high-$T_c$ superconductivity in these materials.

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