Inelastic Neutron-Scattering Measurements of a Three-Dimensional Spin Resonance in the FeAs-Based BaFe$_{1.9}$Ni$_{0.1}$As$_2$ Superconductor

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We use inelastic neutron scattering to study magnetic excitations of the FeAs-based superconductor BaFe$_{1.9}$Ni$_{0.1}$As$_2$ above and below its $T_c$ (=20 K). In addition to gradually open a spin gap at the in-plane antiferromagnetic ordering wave vector $(1, 0, 0)$, the effect of superconductivity is to form a three-dimensional resonance with clear dispersion along the $c$ axis. The intensity of the resonance develops like a superconducting order parameter, and the mode occurs at distinctively different energies at $(1, 0, 0)$ and $(1, 0, 1)$. If the resonance energy is associated with the superconducting gap energy $\Delta$, then $\Delta$ is dependent on the wave vector transfers along the $c$ axis. These results suggest that one must be careful in interpreting the superconducting gap energies obtained by surface sensitive probes such as scanning tunneling microscopy and angle resolved photoemission.

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Understanding the interplay between spin fluctuations and superconductivity in high-transition-temperature (high-$T_c$) superconductors is important because spin fluctuations may mediate electron pairing for superconductivity. In the case of high-$T_c$ copper oxides, it is now well documented that the spin fluctuation spectrum is dominated by a collective excitation known as the resonance mode centered at the antiferromagnetic (AF) ordering wave vector $Q = (1/2, 1/2)$ [1–5]. A similar mode has also been observed in heavy fermion superconductors UPd$_2$Al$_3$ [6] and CeCoIn$_5$ [7]. Although the intensity of the mode behaves like an order parameter below $T_c$, the energy of the mode is dispersionless for wave vector transfers $(Q)$ along the $c$ axis and directly tracks $T_c$ [2–5], thus suggesting that the mode is an intrinsic property of the two-dimensional (2D) CuO$_2$ planes and intimately associated with superconductivity. For FeAs-based superconductors [8–11], the presence of static AF ordering in their parent compounds [with spin structure of Fig. 1(a)] [12–16] and the remarkably similar doping dependent phase diagram to that of the high-$T_c$ copper oxides [13] suggest that AF spin fluctuations may also play an important role in the superconductivity of these materials. Indeed, recent neutron-scattering measurements on spin fluctuations of powder samples of superconducting Ba$_{0.8}$K$_{0.2}$Fe$_2$As$_2$ ($T_c = 38$ K) [17] and crystalline electric field excitations of Ce in CeFeAsO$_{0.84}$F$_{0.16}$ ($T_c = 41$ K) [18] found clear evidence for resonant-like magnetic intensity gain below $T_c$ at $\hbar\omega \sim 14$ and 18.7 meV, respectively. However, the Ce crystalline electric field measurements give no information on the $Q$ dependence of the scattering [18]. Although the resonant-like scattering in Ba$_{0.8}$K$_{0.2}$Fe$_2$As$_2$ occurs near the AF ordering wave vector, the powder nature of the experiment impedes to distinguish whether the resonant scattering is centered at the three-dimensional (3D) AF wave vector $Q = (1, 0, 1)$ of its parent compound [14–16] or simply at a 2D AF in-plane wave vector $Q = (1, 0, 0)$ [17].

FIG. 1 (color online). (a) Schematic diagram of the Fe spin ordering in the BaFe$_2$As$_2$ and we use the same unit cell for BaFe$_{1.9}$Ni$_{0.1}$As$_2$ for easy comparison. (b) Reciprocal space probed in our experiment. (c) Resistivity and magnetic susceptibility measurements of $T_c$. (d,e) Elastic neutron-scattering $L$ scans through $(1, 0, 1)$ and $(1, 0, 3)$ magnetic Bragg peaks at 30 K, showing no evidence of static long-range AF order [15,16].
In this Letter, we report the results of inelastic neutron-scattering studies of spin fluctuations in single crystals of superconducting BaFe$_{1-x}$Ni$_x$As$_2$ [$T_c = 20$ K, Fig. 1(c)] [11]. We show that the effect of superconductivity is to gradually open a low-energy spin gap and also to induce a resonance at energies above the spin gap energy. Although the intensity of the resonance develops below $T_c$ similar to that of the resonance in high-$T_c$ copper oxides, the mode actually has a dispersion along the $c$ axis, and occurs at distinctively different energies at $Q = (1, 0, 1)$ and $(1, 0, 0)$ in contrast with the cuprates. If the resonance energy in FeAs superconductors is associated with the superconducting gap energy $\Delta$, then $\Delta$ should be 3D in nature and depend sensitively on the $Q$ values along the $c$ axis.

We grew high quality BaFe$_{19}$Ni$_{11}$As$_2$ single crystals (each with mosaicity $<0.5^\circ$) using the flux method [11]. Figure 1(c) shows resistivity and magnetic susceptibility data of a typical crystal showing an onset $T_c$ of 20.2 K with a transition width less than 1 K. We coaligned 21 single crystals on a flat Al plate to obtain a total mass of about 0.6 grams. The in-plane mosaic of the aligned crystal assembly is about 1.3$^\circ$ and the out-of-plane mosaic is less than 4.3$^\circ$ full width at half maximum. Our neutron-scattering experiments were performed on the PANDA cold triple-axis spectrometer at the Forschungsneuquelle Heinz Maier-Leibnitz (FRM II), TU München, Germany. We used pyrolytic graphite (0,0,2) as monochromator and analyzer without any collimator. 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 [14–16] of the parent undoped compound (space group Fmmm, $a = 5.564$, $b = 5.564$, and $c = 12.77$ Å). We choose this reciprocal space notation (although the actual crystal structure is tetragonal) for easy comparison with previous spin-wave and elastic measurements on the parent compound, where magnetic Bragg peaks and low-energy spin waves are expected to occur around $(1, 0, 0)$ and $(1, 0, 3)$ rlu positions [see Fig. 1(b)] [19–21]. For the experiment, the BaFe$_{19}$Ni$_{11}$As$_2$ crystal assembly was mounted in the [$H$, $0$, $L$] zone inside a closed cycle refrigerator. The final neutron wave vector was fixed at either $k_f = 1.55$ Å$^{-1}$ with a cold Be filter or at $k_f = 2.662$ Å$^{-1}$ with a pyrolytic graphite filter in front of the analyzer.

We first searched for possible static AF order in our samples. For undoped BaFe$_2$As$_2$, magnetic Bragg peaks are expected at the $(1, 0, 1)$ and $(1, 0, 3)$ positions for the spin structure of Fig. 1(a) [14]. In addition, the low-temperature spin waves have an anisotropy gap of about 9.8 meV [21]. Our elastic $Q$ scans through these expected AF Bragg peak positions are featureless [Figs. 1(d) and 1(e)], confirming the absence of static long-range order above 30 K. Figure 2 summarizes constant-energy scans at 3 K (well below $T_c$) and at 30 K (above $T_c$) at $\hbar\omega = 2, 6$, and 8.5 meV carried out with $k_f = 1.55$ Å$^{-1}$.

Although these probed energies are well below the 9.8 meV spin gap energy in the parent compound [21], we observe at 30 K clear peaks centered at the in-plane AF wave vector $(0, 0, 0)$ for $\hbar\omega = 2$ and 6 meV, and half of a peak at $\hbar\omega = 8.5$ meV due to kinematic constraints [Figs. 2(a)–2(c)]. Fourier transforms of the Gaussian peaks in Figs. 2(a) and 2(b) gave the minimum dynamic spin correlation lengths of $\xi = 16 \pm 4$ and $21 \pm 4$ Å for $\hbar\omega = 2$ and 6 meV, respectively. The spin-spin correlations extend only to several chemical unit cells and are much smaller than the $\xi = 80 \pm 10$ Å at $\hbar\omega = 1.5$ meV obtained for electron-doped cuprate superconductor Pr$_{0.88}$La$_{0.12}$Ce$_{0.15}$CuO$_4$ [4]. On cooling from 30 to 3 K, the Gaussian peak at $\hbar\omega = 2$ meV vanishes and suggests the opening of a spin gap [Figs. 2(a)]. In contrast, the Gaussian peaks at $\hbar\omega = 6$ meV hardly change across $T_c$ [Fig. 2(b)], whereas the scattering at $(1, 0, 0)$ for $\hbar\omega = 8.5$ meV actually increases below $T_c$ [Fig. 2(c)]. These results are similar to those for electron-doped Nd$_{1.85}$Ce$_{0.15}$CuO$_4$ [5], and immediately suggest that the opening of a low-energy spin gap below $T_c$ is compensated by intensity gain above the gap energy. The low-temperature $(1, 0, L)$ scan at $\hbar\omega = 8.5$ meV shows two broad peaks centered at $(1, 0, -1)$ and $(1, 0, 1)$ corresponding to the 3D AF ordering wave vector [14–16].

To determine the size of the superconducting spin gap and confirm the intensity gain at $\hbar\omega = 8.5$ meV below $T_c$, we carried out energy scans at $Q = (1, 0, 0)$ below and above

![FIG. 2 (color online). Constant-energy scans around the (1, 0, 0) and (1, 0, 1) positions for $\hbar\omega = 2$, 6, and 8.5 meV obtained with $k_f = 1.55$ Å$^{-1}$. (a–c) $Q$ scan along the [$H$, $0$, $0$] direction for $\hbar\omega = 2$, 6, and 8 meV at 30 and 3 K. The inset in (a) shows the temperature difference plot and a Gaussian fit to the data. The missing low-$Q$ data for scans in (b) and (c) are due to kinematic constraint. (d) $Q$ scan along the [$1$, $0$, $L$] direction for $\hbar\omega = 8.5$ meV at 3 K. Note two clear peaks centered at $(1, 0, -1)$ and $(1, 0, 1)$, respectively. The dashed-line peak is the low-temperature spin waves of BaFe$_2$As$_2$ at $\hbar\omega = 10$ meV from Fig. 2f in [21].]
The results suggest that the spin gap in reduction below 15 K is not due to simple Bose statistics. Figure 3(c) shows the energy dependence of the scattering at \( Q = (1,0,0) \) and \( h\omega = 2 \text{ meV} \). The solid line shows the expected magnetic intensity change due to the Bose population factor; it is clear that the intensity reduction below 15 K is not due to simple Bose statistics. The results suggest that the spin gap in \( \text{BaFe}_{1.9}\text{Ni}_{0.1}\text{As}_2 \) opens gradually with decreasing temperature until it reaches about 4 meV at 3 K (confirmed by recent measurements), remarkably similar to the spin gap behavior of electron-doped \( \text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4 \) [5,22].

Although the results displayed in Figs. 1–3 using \( k_f = 1.55 \text{ Å}^{-1} \) are suggestive of a resonance below \( T_c \), kinematic constraints did not allow us to carry out measurements for energies above \( h\omega = 9 \text{ meV} \) at \( Q = (1,0,0) \). To determine the energy location of the possible mode, we collected additional data with \( k_f = 2.662 \text{ Å}^{-1} \). Figure 4(a) shows the energy scan raw data at \( Q = (1,0,0) \) below and above \( T_c \). Inspection of the data reveals that the low-temperature scattering enhances dramatically around \( h\omega = 9.0 \text{ meV} \) compared to the normal state scattering. Since Bose population factor does not contribute much to magnetic scattering intensity for \( h\omega > 5 \text{ meV} \) between 3 and 30 K, the (low minus high) temperature difference scattering represents the net magnetic intensity gain at low temperature. Subtracting the 30 K data from the 3 K data reveals a clear localized mode near 9.0 meV [Fig. 4(b)]. Gaussian fit to the data gives a peak position \( h\omega = 9.1 \pm 0.4 \text{ meV} \), a peak width \( 3.3 \pm 0.9 \text{ meV} \), and an integrated area \( 346 \pm 82 \) per 20 minutes [Fig. 4(b)]. Since spin excitations at \( h\omega = 8.5 \text{ meV} \) peak at \( (1,0,-1)/(1,0,1) \) and have clear \( c \) axis modulation.

![FIG. 3 (color online). (a) Energy scans at \( Q = (1,0,0) \) from 2 to 9 meV at 30 and 3 K with \( k_f = 1.55 \text{ Å}^{-1} \). The broad peak in energy near 6 meV was also seen in the background scattering collected at the (1,3,0) position (not shown) and were not intrinsic properties of the material. (b) Intensity difference between the 3 and 30 K data at \( Q = (1,0,0) \). The negative scattering below 4 meV indicates the opening of a spin gap, while positive scattering above 6 meV suggests a magnetic intensity gain below \( T_c \). (c) Temperature dependence of the scattering obtained at \( Q = (1,0,0) \) and \( h\omega = 2 \text{ meV} \) with the vertical arrow indicating \( T_c \). The solid line shows the expected \( T \)-dependent scattering due to the Bose population factor.](image)

![FIG. 4 (color online). (a) Energy scans at \( Q = (1,0,0) \) from 5 to 13 meV at 30 and 3 K with \( k_f = 2.662 \text{ Å}^{-1} \). The background scattering at \( Q = (1.3,0,-1) \) is weakly \( T \)-dependent between 30 and 3 K. (b) The temperature difference scattering between 3 and 30 K shows a clear resonant peak at \( h\omega = 9.1 \pm 0.4 \text{ meV} \). (c) Energy scans at \( Q = (1,0,-1) \) from 2 to 13 meV at 30 and 3 K. (d) The temperature difference plot confirms that the mode has now moved to 7.0 \pm 0.5 \text{ meV} \). (e) Wave vector dependence of the scattering at 30 and 3 K for \( h\omega = 7 \text{ meV} \), confirming that the resonance intensity gain occurs at \( Q = (1,0,-1) \). (f) Temperature dependence of the scattering at \( h\omega = 7 \text{ meV} \) shows a clear order-parameter-like increase below \( T_c \). The solid line is the best fit to the data using \( I = I_o + k(1-(T/T_o)^\beta) \) yielding \( \beta = 0.5 \) and \( T_o = 20 \text{ K} \). The intensity differences in (c) and (f) are within 2\( \sigma \).](image)
clearly shows that the intensity gain below \( T_c \) confirms that the intensity gain at the resonance occurring at \( Q = (1, 0, -1) \). A Gaussian fit to the temperature difference plot in Fig. 4(d) gives a peak position \( h \omega = 7.0 \pm 0.5 \) meV, a peak width 1.9 \pm 0.7 meV, and an integrated area of 464 \pm 145 per 20 minutes. To further confirm that the intensity gain at \( h \omega = 7 \) meV is indeed the resonance occurring at \( Q = (1, 0, -1) \), we carried constant-energy scans around \((1,0,-1)\) and the outcome clearly shows that the intensity gain below \( T_c \) arises from scattering at the 3D AF ordering position [Fig. 4(e)]. Finally, in Fig. 4(f) we plot the temperature dependence of the scattering at \((1, 0, -1)\) and \( h \omega = 7 \) meV. The scattering increases dramatically below the onset of \( T_c \) and is remarkably similar to that of the resonance in high-\( T_c \) copper oxides [1–5].

If the resonance is a measure of electron pairing correlations in high-\( T_c \) superconductors [23], the observed 3D resonance dispersion in BaFe\(_{1-x}\)Ni\(_{x}\)As\(_2\) would suggest a variation of the superconducting gap \( \Delta \) along the \( c \) axis, similar to those in UPd\(_2\)Al\(_3\) [24]. This is quite different from the high-\( T_c \) copper oxides, where \( \Delta \) is strictly 2D and independent of the \( c \) axis modulations. For FeAs-based superconductors, the resonance may arise from quasiparticle transitions across the sign-revised \( s \)-wave electron \( (\Delta_0) \) and hole \( (\Delta_0^*) \) superconducting gaps in pure two-dimensional models [25–30]. By considering the AF coupling between layers, the gap functions can be naturally modified to \( \Delta_0(k_z) = \Delta_0^0 + \delta \cos(k_z) \) and \( \Delta_0(k_z) = \Delta_0^0 + \delta \cos(k_z) \). For a sign-revised \( s \)-pairing symmetry, \( \Delta_0^0 \sim -\Delta_0^0 \sim -\Delta_0^0 \). Therefore, the dispersion of the resonance along the \( c \) axis is roughly determined by [26]

\[
h \omega(q_z) \sim \min[(|\Delta_0(k_z)| + |\Delta_0(k_z + q_z)|, k_z)] \sim 2\Delta_0^0 - 2\delta \left| \sin \left( \frac{q_z}{2} \right) \right|. \tag{1}
\]

Based on this interpretation, our experimental results suggest \( \delta/\Delta_0^0 = [\omega(1, 0, 0) - \omega(1, 0, -1)]/\omega(1, 0, 0) = 0.26 \pm 0.07 \). If spin fluctuations are responsible for electron pairing and superconductivity, the values \( \Delta_0^0 \) and \( \delta \) are expected to be proportional to the intraplane and interplane AF couplings, \( J_\parallel \) and \( J_\perp \), respectively, which naturally suggests \( \delta/\Delta_0^0 \sim J_\perp/J_\parallel \). The ratio \( \delta/\Delta_0^0 \) determined by our resonance dispersion is a reasonable agreement with the ratio of the AF exchange couplings measured by neutron-scattering experiments in the parent compounds [19–21]. These results suggest that spin fluctuations are also important for superconductivity in FeAs-based superconductors.

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Note added.—After finishing the present work, we became aware of a preprint where the resonance at \( h \omega = 9.5 \) meV was reported near \( Q = (1, 0, 0) \) in superconducting BaFe\(_{1.84}\)Co\(_{0.16}\)As\(_2\) (\( T_c = 22 \) K) [31].

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