Spin gap and magnetic resonance in superconducting BaFe$_{1.9}$Ni$_{0.1}$As$_2$

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We use neutron spectroscopy to determine the nature of the magnetic excitations in superconducting BaFe$_{1.9}$Ni$_{0.1}$As$_2$ ($T_c = 20$ K). Above $T_c$, the excitations are gapless and centered at the commensurate antiferromagnetic wave vector of the parent compound, while the intensity exhibits a sinusoidal modulation along the $c$-axis. As the superconducting state is entered a spin gap gradually opens, whose magnitude tracks the $T$-dependence of the superconducting gap observed by angle resolved photoemission. Both the spin gap and magnetic resonance energies are temperature and wave vector dependent, but their ratio is the same within uncertainties. These results suggest that the spin resonance is a singlet-triplet excitation related to electron pairing and superconductivity.

PACS numbers: 74.25.Ha, 74.70.-b, 78.70.Nx

The magnetic scattering in the high-transition-temperature (high-$T_c$) copper oxide superconductors is characterized by strong spin correlations in the vicinity of the antiferromagnetic (AF) wave vector of the magnetically ordered parent materials, and a spin 'resonant' magnetic excitation whose energy scales with $T_c$ and whose intensity develops like the superconducting order parameter. As the superconducting state is entered a spin gap gradually opens, whose magnitude tracks the $T$-dependence of the superconducting gap observed by angle resolved photoemission. Both the spin gap and magnetic resonance energies are temperature and wave vector dependent, but their ratio is the same within uncertainties. These results suggest that the spin resonance is a singlet-triplet excitation related to electron pairing and superconductivity.

We use neutron spectroscopy to determine the nature of the magnetic excitations in superconducting BaFe$_{1.9}$Ni$_{0.1}$As$_2$ ($T_c = 20$ K) because these samples have excellent superconducting properties. In the absence of Ni-doping, BaFe$_2$As$_2$ is a nonsuperconducting metal that orders antiferromagnetically with a spin structure shown in Fig. 1a. Because of the unit cell doubling along the orthorhombic $a$-axis and $c$-axis spin arrangement, magnetic Bragg reflections occur at wave vectors $Q = (1,0,1)$ and $(1,0,3)$ type positions and are absent at $Q = (1,0,0)$ and $(1,0,2)$ because of the unit cell doubling along the orthorhombic $a$-axis and $c$-axis spin arrangement. Previous neutron scattering experiments on hole-doped Ba$_{0.68}$K$_{0.32}$Fe$_2$As$_2$ powder samples and single crystals of BaFe$_{1.84}$Co$_{0.16}$As$_2$ ($T_c = 22$ K) show that the effect of superconductivity is to induce a neutron spin resonance at energies of $\sim 5k_BT$, remarkably similar to the doping dependence of the resonance in high-$T_c$ copper oxides and heavy fermions. Measurements on single crystals of BaFe$_{1.9}$Ni$_{0.1}$As$_2$ ($T_c = 20$ K) suggest that the resonance actually exhibits dispersion along the $c$-axis, and occurs at distinctively different energies at the three-dimensional (3D) AF ordering wave vector $Q = (1,0,1)$ and at $Q = (1,0,0)$. We note that in the parent materials the spin wave dispersions in the Fe-based superconductors are anisotropic and clearly 3D in nature, as opposed to the purely two-dimensional spin wave dispersion on the parent cuprates. For the cuprates the spin fluctuations in the superconducting regime are again purely 2D, while the iron-based superconductors appear to exhibit anisotropic 3D behavior like their parents.
The neutron scattering measurements were carried out on the SPINS cold and BT-7 thermal triple-axis spectrometers at the NIST Center for Neutron Research. We label the momentum transfer \( Q = (q_x, q_y, q_z) \) as \( (H, K, L) = (q_x/2\pi, q_y/2\pi, q_z/2\pi) \) reciprocal lattice units (rlu) using the orthorhombic magnetic unit cell of the parent undoped compound (space group \( Fmmn \), \( a = 5.564, b = 5.564, \) and \( c = 12.77 \) Å) for easy comparison with previous spin wave measurements on the parent compounds, even though the actual crystal structure is tetragonal \([32\), 34, 35]). Many single crystals were co-aligned to obtain a total mass of \( \sim 1.2 \) grams. The in-plane and out-of-plane mosaics of the aligned crystal assembly are \( 1.3^\circ \) and \( 4.3^\circ \) full width at half maximum (FWHM), respectively \([24]\). For the experiment, the BaFe\(_{1.9}\)Ni\(_{0.1}\)As\(_2\) crystal assembly was mounted in the \([H, 0, L]\) zone inside a liquid He cryostat. The final neutron wave vector was fixed at either \( E_f = 5 \) meV with a cold Be filter or at \( E_f = 14.7 \) meV with a PG filter in front of the analyzer.

We first probe the wave vector dependence of the low-energy spin fluctuations. Figures 2a and 2b show \([H, 0, 3]\) and \([1, 0, L]\) scans at \( E = 1 \) meV through the 3D \((1, 0, 3)\) Bragg peak position below and above \( T_c \). We see that the spin excitations observed above \( T_c \) vanish at low \( T \). Fig. 2c shows the intensity of the scattering above \( T_c \) as a function of wave vector along the \( c \)-axis, using the low \( T \) data as background, and reveals the intrinsic wave vector modulation of the intensity of the normal state spin fluctuations. The solid curve is a fit to the data using \( \Delta S(Q, \omega)/(24 K - 2 K) = A F(Q)^2 \sin^2(\pi L/2) + C \), where \( F(Q) \) is the magnetic form factor of Fe\(^{2+}\) and \( C \) is constant. These data are consistent with previous work on BaFe\(_{1.9}\)Ni\(_{0.1}\)As\(_2\) which showed that the spin fluctuation intensity has a \( c \)-axis modulation at \( E = 8.5 \) meV, and a gap in the superconducting state \([24]\). For comparison, Fig. 2d shows the magnetic scattering through the \([1, 0, 1]\) position in the superconducting state at the resonance energy of \( E = 7 \) meV, and the magnetic scattering above \( T_c \). We note that in the undoped AF state, the spin wave spectrum in BaFe\(_2\)As\(_2\) has a gap of 9.8 meV \([35]\), while in the normal state of the doped system we find that the spin fluctuation spectrum is gapless.

The behavior of the low energy spin excitations as a function of temperature is shown in Fig. 3, which summarizes the BT-7 and SPINS data around \( Q = (1, 0, 1) \). Figure 3a shows wave vectors \([H, 0, 1]\) scans through the \( Q = (1, 0, 1) \) position above and below \( T_c \) at \( E = 2 \) meV. A clear Gaussian peak centered at \( Q = (1, 0, 1) \) in

FIG. 1: (color online). (a) Schematic diagram of the Fe spin structure in the BaFe\(_2\)As\(_2\), which has magnetic Bragg peaks at \( Q = (1, 0, 1), (1, 0, 3) \), etc. For our experiment on BaFe\(_{1.9}\)Ni\(_{0.1}\)As\(_2\), we use the same unit cell for easy comparison. (b) Temperature dependence of the spin gap as determined from energy scans (Fig. 3c) and the temperature dependence of the scattering at \( Q = (1, 0, 1) \) (Fig. 3d). The solid curve represents the temperature dependence of the BCS gap function. (c, d) Schematic of the magnetic response and spin gaps at \( Q = (1, 0, 0), (1, 0, 1) \), respectively. Measurements at \( Q = (1, 0, 3) \) showed similar behavior as those at \( Q = (1, 0, 1) \).

FIG. 2: (color online). Examples of constant energy scans around the \((1, 0, 3)\) position for \( E = 1 \) meV obtained with \( E_f = 5 \) meV above and below \( T_c \) on SPINS. (a) \( Q \)-scan along the \([H, 0, 3]\) direction for \( E = 1 \) meV at 24 K and 2 K. A clear peak centered at \((1, 0, 3)\) at 24 K disappears at 2 K, indicating the opening of a spin gap. (b) Similar scan along the \([1, 0, H]\) direction showing a peak centered at \((1, 0, 3)\) that disappears below \( T_c \). (c) Using scattering at 2 K as background scattering, we determine the normal state \( L \)-modulation of the spin fluctuations by subtracting the 2 K data from 24 K data. It is clear that spin fluctuations are 3D and have similar modulations along the \( c \)-axis as spin waves. (d) \( Q \)-scan in the superconducting state through the magnetic resonance position, and above \( T_c \) near \((1, 0, 1) \).
the normal state vanishes below $T_c$, demonstrating that the spin gap $\Delta_{\text{spin}} > 2$ meV. Figure 3b plots the signal and background scattering along the $[1, 0, L]$ direction for $E = 2$ meV at 30 K, where we find that the normal state scattering also peaks at 3D AF wave vector positions. To determine the spin gap value at $Q = (1, 0, 1)$, we carried out temperature dependent measurements at 2 K, 15 K, and 30 K using SPINS. We find a clear reduction in scattering (net negative values in the subtraction) below 3 meV and 1.5 meV at 2 K and 15 K, respectively. These results show that the maximum magnitude of the spin gap at 2 K and 15 K, respectively.

Energy scattering increases in intensity. The overall behavior of the data is remarkably similar to that in the optimally hole-doped La$_{1-\delta}$Sr$_\delta$CuO$_4$ [22] and electron-doped Nd$_{1.85}$Ce$_{0.15}$CuO$_4$ [29]. However, it is also clear that the spin-gap occurs at a lower energy at $Q = (1, 0, 1)$ than for $Q = (1, 0, 0)$, which is quite different than the cuprates [22]. Figure 4c,d presents the data in the form of the dynamic susceptibility $\chi''(Q, \omega)$, which is related to $S(Q, \omega)$ through the (removal of the) detailed balance factor; $\chi''(Q, \omega) = (1 - \exp(-\hbar\omega/k_BT))S(Q, \omega)$. Recall that the thermal population factor increases with decreasing temperature, and this function is divided into $S(Q, \omega)$ to obtain $\chi''(Q, \omega)$ [with $\chi''(Q, \omega = 0) = 0$]. The filled circles are $\chi''(Q, \omega)$ obtained from Q-scans as a consistency check. Upon entering the superconducting state, the spectral weight is rearranged, with the suppression of low energy spin fluctuations and the appearance of the neutron spin resonance at energies above the spin gap. The present data are consistent with the reported spin resonance values of $E = 9$ meV for $Q = (1, 0, 0)$ and $E = 7$ meV for $Q = (1, 0, 1)$ [24]. We estimate that the intensity of the resonance is approximately compensated by the opening of the spin gap below the resonance.
To quantify the magnitude of the spin gaps at \( Q = (1, 0, 0) \) and \( (1, 0, 1) \) in the superconducting state, we follow previous work [32] and fit the data with

\[
S(Q, \omega) = \frac{A E^{T}}{(\Gamma^{2} + (\hbar \omega)^{2})(1 - \exp(-\hbar \omega/k_{B}T))},
\]

where \( E^{T} = Re[(\hbar \omega - \Delta + i\Gamma_{s})(\hbar \omega + \Delta + i\Gamma_{s})]^{1/2} \), \( A \) is the amplitude, \( \Delta \) is the spin gap, \( \Gamma \) is the inverse lifetime of the spin fluctuations with \( \hbar \omega \gg \Delta \), \( E^{T} \) is an odd function of \( E = \hbar \omega \), and \( \Gamma_{s} \) is the inverse lifetime of the fluctuations at the gap edge. The solid curves are the results of these fits. In the normal state, this functional form does not provide an adequate fit over the entire energy range, and we restricted it to lower energies (as indicated by the extent of the curve for those data). We find \( \Delta = 0 \) for both \( Q = (1, 0, 0) \) and \( (1, 0, 1) \). On cooling into the superconducting state, Eq. (1) can be used over the entire energy range of the data, and the least-squares fit to the \( Q = (1, 0, 0) \) data (solid curves in Figs. 4a and 4c) yields \( A = 56.7 \pm 7.9 \), \( \Gamma = 13 \pm 6.5 \) meV, \( \Delta = 4.3 \pm 0.8 \) meV, \( \Gamma_{s} = 0 \pm 0.73 \) meV. Similarly, for \( Q = (1, 0, 1) \) we find \( A = 55.5 \pm 14.5 \), \( \Gamma = 5 \pm 0.7 \) meV, \( \Delta = 2.5 \pm 0.08 \) meV, \( \Gamma_{s} = 0 \pm 0.53 \) meV (solid curves in Figs. 4b and 4d). The results of this analysis show that the superconducting spin gap values for \( Q = (1, 0, 0) \) and \( (1, 0, 1) \) are distinctively different.

The present measurements, as well as the previous data on this material [24], demonstrate that the resonance occurs at \( E = 9 \) meV for \( Q = (1, 0, 0) \), which has a spin gap \( \Delta = 4.3 \pm 0.8 \) meV. For \( Q = (1, 0, 1) \), the resonance is at the lower energy of \( E = 7 \) meV, and the spin gap also occurs at the lower energy of \( \Delta = 2.5 \pm 0.08 \) meV. Therefore these two energy scales track one another, with a ratio that is the same within the uncertainties of the experiments. This is the expected behavior for the singlet-triplet transition of a Cooper pair [26].

We summarize in Figs. 1b-1d the key results of our experiments. The measured temperature dependence of the spin gap at \( Q = (1, 0, 1) \) is shown in Fig. 1b. The solid curve shows the prediction of a simple BCS gap function near \( T_{c} \), \( \Delta(T) = A(1 - (T/T_{c}))^{1/2} \), which describes the data fairly well. Figures 1c and 1d plot schematically the spin gap and resonance at \( Q = (1, 0, 0) \) and \( (1, 0, 1) \). The two energies exhibit the same dependence on wave vector. In ARPES experiments [27], two isotropic superconducting gaps with values of 7 meV and 4.5 meV were observed for BaFe\(_{1-x}\)Co\(_{x}\)As\(_{2}\) with \( T_{c} = 25.5 \) K. Comparison with the \( Q = (1, 0, 0) \) neutron measurements suggests that the resonance energy at \( Q = (1, 0, 0) \) is indeed less than twice the superconducting gap energy. These results are consistent with the idea that the resonance is a bond state related to singlet-triplet excitations of Cooper pairs, with a superconducting gap that varies with the momentum transfer along the c-axis [30].

We thank Songxue Chi, Jun Zhao, and Leland Harriger for coaligning some of the single crystals used in the present experiment. This work is supported by the U.S. DOE BES No. DE-FG02-05ER46202, NSF DMR-0756568, and in part by the U.S. DOE, Division of Scientific User Facilities. The work at Zhejiang University is supported by the NSF of China.

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