Dispersive Spin Fluctuations in the near optimally-doped superconductor
\( \text{Ba(Fe}_{1-x}\text{Co}_x\text{)As}_2 \) \((x=0.065)\)

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Inelastic neutron scattering is used to probe the collective spin excitations of the near optimally-doped superconductor \( \text{Ba(Fe}_{1-x}\text{Co}_x\text{)As}_2 \) \((x=0.065)\). Previous measurements on the antiferromagnetically ordered parents of this material show a strongly anisotropic spin-wave velocity. Here we measure the magnetic excitations up to 80 meV and show that a similar anisotropy persists for superconducting compositions. The dispersive mode measured here connects directly with the spin resonance previously observed in this compound. When placed on an absolute scale, our measurements show that the local- or wavevector-integrated susceptibility is larger in magnitude than that of the ordered parents over the energy range probed.

I. INTRODUCTION

The recently discovered ferropnictide superconductors 1–6 show critical temperatures exceeding 50 K. They are interesting materials, both in their own right and because they provide valuable insights into other classes of superconductor. Theoretical calculations 2, 4 suggest that the electron-phonon coupling is too weak in the ferropnictides to produce the high transition temperatures which are observed experimentally. The ferropnictides have some similarities to the cuprates in that they are quasi-two-dimensional and have antiferromagnetic parent compounds. Superconductivity in the ferropnictides can be induced 2, 3 from their antiferromagnetic parents by various means: electron or hole doping via chemical substitution; isovalent substitution of the iron or arsenic or by the application of pressure. In the widely-studied AFe2As2 (A=Ca,Sr,Ba) family (known as the “122” family) 2, 3, this can be via chemical substitution at the Fe or As sites or the application of pressure. For example, in BaFe2As2, we obtain superconductivity through electron doping by Co or Ni substitution at the Fe site; by hole doping with K at the Ba site; or by isovalent substitution of Fe by Ru or As by P.

The proximity of superconductivity to antiferromagnetism suggests that the pairing mechanism in the doped ferropnictides is related to the spin degrees of freedom. Theories based on various models of the magnetic excitations have been proposed 7–13. In order to take such theories forward, the collective magnetic excitations in the superconducting region of the phase diagram need to be characterized and the underlying interactions understood. Thus, in this paper, we report an inelastic neutron scattering (INS) study of the magnetic response, up to 80 meV, of a near optimally-doped superconducting \((T_C=23 \text{ K})\) composition \( \text{Ba(Fe}_{0.935}\text{Co}_{0.065})\text{As}_2 \). This composition appears not to be magnetically ordered. We compare our results with similar measurements on the antiferromagnetic parent compounds 14–17 of the 122 series and find that (i) the magnetic excitations are stronger than in the parent compounds and (ii) the anisotropy in the spin-wave velocity observed in the parent compounds persists in this superconductor.

II. EXPERIMENTAL DETAILS

Single crystals of \( \text{Ba(Fe}_{0.935}\text{Co}_{0.065})\text{As}_2 \) were grown by a self-flux method 18. 15 crystals were co-aligned on a thin Al plate using x-rays and neutron diffraction. Our neutron measurements were made on an mosaic of total mass 0.3 g. Resistivity and magnetization measurements identified the superconducting transition temperature \( T_C(\text{onset}) = 23 \text{ K} \). Elastic neutron scattering revealed no evidence of magnetic order at this doping level and temperatures down to 2 K. We used the MAPS instrument at the ISIS spallation source. MAPS is a low-background direct-geometry time-of-flight chopper spectrometer with position sensitive detectors 19. A pulse of spallation neutrons spread over a time of about 5–10 \( \mu \text{s} \) is produced when a pulse of protons hits a Ta target adjacent to a water moderator. Neutrons with the required energy \((E_i = 60, 80, \text{ or } 140 \text{ meV in the present experiment})\) are then selected by an appropriately phased Fermi chopper (rotating at 100 Hz in the present experiment). The Fermi chopper is 10 m from the neutron source and opens for about 45 \( \mu \text{s} \). The neutrons then scatter from the sample \((12 \text{ m from source})\) and are detected in position sensitive 3He detectors at 6 m from the sample. The detection time of the neutron is used to determine its energy transfer. Data are averaged of a range of energies to improve the experimental statistics. The energy ranges of integration are given when we quote neutron energy transfers in the text. Data were placed on an absolute scale \((\text{barn sr}^{-1} \text{ f.u.}^{-1})\) by comparing the count rate with...
that from a plate of vanadium.

The magnetic cross section of an isotropic paramagnet is given by

\[ \frac{d^2\sigma}{d\Omega \, dE} = \frac{2(\gamma r_c)^2}{\pi^2 \mu_B^2} \frac{k_f}{k_i} |F(Q)|^2 \left( \frac{\chi''(q, \omega)}{1 - \exp(-\hbar \omega/kT)} \right), \]

where \((\gamma r_c)^2 = 0.2905\) barn sr\(^{-1}\), \(k_i\) and \(k_f\) are the incident and final neutron wavevectors and \(|F(Q)|^2\) is the isotropic magnetic form factor for a Fe\(^{2+}\) orbital. We use Eq. 1 to convert the measured cross section to the energy- and momentum-dependent susceptibility \(\chi''(q, \omega)\). Ba(Fe\(_{0.935}\)Co\(_{0.065}\))\(_2\)As\(_2\) has the tetragonal crystal structure shown in Fig. 2(a) with lattice parameters \(a = 3.955\) Å and \(c = 12.95\) Å. We use the reciprocal space notation to label wavevectors \(Q = h \mathbf{a}^* + k \mathbf{b}^* + \ell \mathbf{c}^*\). In this notation, the antiferromagnetic ordering wavevector of BaFe\(_2\)As\(_2\) is \((1/2, 1/2, 0)\). The data reported in this paper were collected with \(\mathbf{c}^*\) parallel to \(k_i\). Under these conditions, there is a coupling of \(l\) and \(\omega\). We give the \(l\) value corresponding to each \(\omega\) in the figures and captions.

III. RESULTS

Figure 2 shows typical \(q\)-dependent images of \(S(q, \omega) = (k_i/k_f) d^2\sigma/d\Omega dE\) in the \((h, k)\) plane for various energy transfers obtained from the MAPS spectrom-
La$_2$-$_x$Sr$_x$CuO$_{23-y}$ and more recently FeTe$_{1-x}$Se$_x$[24]. The Sato-Maki cross-section is of the form:

$$\chi''(q,\omega) = \chi_0(\omega) \frac{\kappa^4(\omega)}{[\omega^2(\omega) + R(q)]^2}$$

(6)

with

$$R(q) = \frac{1}{4\delta^2} \left\{ \left[ (h-h_0)^2 + (k-k_0)^2 - \delta^2 \right]^2 + \frac{\lambda}{4} \left[ (h-h_0)^2 - (k-k_0)^2 \right]^2 \right\},$$

where $Q_0 = (h_0, k_0)$ is the nearest reciprocal lattice point to $Q$ with odd $(h + k)$. The location of the peaks in the response is controlled by $\delta$, and its shape by $\kappa$ and $\lambda$. For example, to obtain a peak centered at $Q=(1/2,1/2)$, $\delta = 1/\sqrt{2}$. We found that the Sato-Maki function could give a good description of our data except at the highest energies $E \approx 80$ meV. Fits of the SM-function are shown in Figs. 4-8. The values of $\chi''(\omega)$ computed from the SM fits are statistically indistinguishable from those obtained using the SW cross-section.

IV. DISCUSSION

The magnetic response has been studied by neutron scattering in a number of superconducting ferropnictide[24-28] including Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$[26,29]. Previous experiments on the superconducting 122 systems have either been on powder samples[25] or performed over a smaller energy range[26,29] than the present experiment. One of the early results from INS on the iron-based superconductors was the observation of a ‘spin-resonance’[25]. The spin-resonance is most easily interpreted in terms of a sign difference in the superconducting gap $\Delta(k)$ between different parts of the Fermi surface[22] and is consistent with a $s^\pm$ gap function. At the lowest energies probed by the present experiment we are able to observe the spin-resonance. Fig. 5 shows this most directly: There is a peak in the local susceptibility $\chi''(\omega)$ near 10 meV at $T = 7$ K which is suppressed on raising the temperature to 26 K. The resonance is strongest near $q = (1/2,1/2)$ as shown in Fig. 2(a) and Fig. 3(d,h). As the energy is increased above the resonance energy, magnetic excitations disperse in an anisotropic manner (see Fig. 4). Thus our results are consistent with an upwardly dispersing mode which is strongest near $q = (1/2,1/2)$. This is at least qualitatively consistent with theoretical predictions for a $s^\pm$ state[30]. Interestingly, this contrasts with the behavior in YBa$_2$Cu$_3$O$_{6+x}$, where the resonance mode disperses downwards in energy[30-32].

An important result from our experiment is the observation of dispersive anisotropic spin fluctuations up to 80 meV. Neutron scattering studies on the antiferromagnetic parents of the 122 series, BaFe$_2$As$_2$[14,15] and CaFe$_2$As$_2$[16,17], have observed spin-wave excitations up to about 200 meV. Unfortunately, single crystal measurements up to 80 meV only exist for CaFe$_2$As$_2$. The magnetic excitations have been analyzed using a spin-wave model based on localized moments. Ba(Fe$_{0.935}$Co$_{0.065}$)$_2$As$_2$ is close to magnetic order at low temperature. In order to parameterize our data we use a damped spin-wave cross-section with an energy gap. In our case, the gap $\Delta$ in Eq. 3 is due to the superconductivity (and the formation of the resonance described above).

We find that Eqs. 2-3 provide a good description of the...

FIG. 2: (color online) (a)-(e) Constant-energy slices through the magnetic excitations in Ba(Fe$_{0.935}$Co$_{0.065}$)$_2$As$_2$ as observed on MAPS. (f)-(j) Fits to a phenomenological spin-wave cross-section (Eqs. 2-3). (k)-(o) Fits to a phenomenological Sato-Maki cross sections (Eq. 6) [e.g. for (a) $\kappa = 0.21 \pm 0.09$, $\delta = 0.6 \pm 0.2$, $\lambda = 5 \pm 2$]. The incident energies used were 60 meV [(a)-(b)], 140 meV [(c)-(e)] and the corresponding $l$ values were $l=1$ [(a)-(b)], 2.5 (c), 4(d) and 5.5 (e). The $E_i=140$ meV data were collected using a proton beam current of 175 $\mu$A for 80 hours. A constant background has been subtracted from each plot.
data (as illustrated by Figs. 2–3) with $\Gamma/E = 0.15$ (as per Ref. 17). This method of analysis produces spin-wave velocities of $v_\parallel = 580 \pm 60$ meVÅ, $v_\perp = 230 \pm 30$ meVÅ and $\Delta = 10 \pm 0.5$ meV. Using the published exchange constants, we obtain spin-wave velocities for the parent compound CaFe$_2$As$_2$ of $v_\parallel = 513$ meVÅ or 494 meVÅ$^{17}$ and $v_\perp = 370$ meVÅ$^{18}$ or 348 meVÅ$^{17}$. If the magnetic interactions are described using a Heisenberg Hamiltonian

$$H = J_1 \sum_{\langle jk \rangle} \mathbf{S}_j \cdot \mathbf{S}_k + J_2 \sum_{\langle jk \rangle} \mathbf{S}_j \cdot \mathbf{S}_k,$$  \hfill (7)$$

where the sums are over nearest-neighbor and next-nearest-neighbor pairs (see Fig. 1), we can convert the spin-wave velocities to effective exchange constants based on spin-wave theory in the ordered parent antiferromagnets. Using the relations $v_\parallel = S_\perp \sqrt{2a(J_1 + 2J_2)}$ and $v_\perp = S_\perp \sqrt{2a/(2J_2 + J_1)(2J_2 - J_1)}$ (see Refs. 14, 16, 17, 32), we obtain $J_1 = 43 \pm 7$ and $J_2 = 30 \pm 3$ meV. It should be noted that this is not the only possible analysis, but it is the simplest. It may also be possible to interpret the data in terms of the $J_{1\alpha \beta}$ model$^{17,33}$ which has been used to describe the ordered parent CaFe$_2$As$_2$. However $|J_{1\alpha} - J_{1\beta}|$ is certainly less for Ba(Fe$_{0.935}$Co$_{0.065}$)$_2$As$_2$.

In addition to comparing the spin-wave velocities with the parent compounds, we can also compare the strength of the magnetic response. Fig. 4 shows the wavevector-averaged or local susceptibility $\chi''(\omega)$ defined by Eq. 4. In the case of Ba(Fe$_{0.935}$Co$_{0.065}$)$_2$As$_2$, we have averaged over $0 \leq h < 1$ and $0 \leq k < 1$, and $\pm 0.25$ around various odd, even and non-integer $l$ values (see caption to Fig. 4). As mentioned above, points with different $l$ appear to follow the same trend, suggesting that there is no $l$-dependence to this partially averaged quantity. Thus, the graph represents the true $\chi''(\omega)$. For comparison, we...
have computed $\chi''(\omega)$ for CaFe$_2$As$_2$ using the exchange constants and $S_{\text{eff}}$ from Ref. 17. This result is shown as the dashed line in Fig. 5. We note that the response is slightly larger in Ba(Fe$_{0.935}$Co$_{0.065}$)$_2$As$_2$ over the energy range investigated here. The same phenomenon occurs in the cuprates where, for example, the spin fluctuations are stronger in optimally-doped La$_{2-x}$Sr$_x$CuO$_4$ than in La$_2$CuO$_4$. One might understand this increase at lower energies as being due to a shift in spectral weight from higher energies (above the window of the present experiment) and the loss of the peak due to magnetic order present in the ordered compounds.

It is interesting to compare the present results with those obtained on the cuprates. Firstly, we note that the magnitude of the local susceptibility is similar in optimally doped La$_{2-x}$Sr$_x$CuO$_4$ and Ba(Fe$_{0.935}$Co$_{0.065}$)$_2$As$_2$. Both the cuprates and Ba(Fe$_{0.935}$Co$_{0.065}$)$_2$As$_2$ show a strong response near the $Q=(1/2,1/2)$. In La$_{2-x}$Sr$_x$CuO$_4$ and YBa$_2$Cu$_3$O$_{6+y}$, various dispersive modes and ‘resonance’ features are observed below $E \lesssim 50$ meV. At higher energies, $50 \lesssim E \lesssim 100$ meV, the response in YBa$_2$Cu$_3$O$_{6+y}$ and La$_{2-x}$Sr$_x$CuO$_4$ is quasi-isotropic or has fourfold symmetry about the $(1/2,1/2)$ position. In contrast, the response in Ba(Fe$_{0.935}$Co$_{0.065}$)$_2$As$_2$ is more anisotropic being broader along $(1/2 - \xi, 1/2 + \xi)$ than the $(1/2 + \xi, 1/2 + \xi)$. We discuss possible origins of this anisotropy below.

Discussion about the best way to describe the magnetic interactions in the ferropnictides continues. Model Lindhard calculations of the wavevector-dependent susceptibility $\chi(q,\omega)$ based on the band structure appear to reproduce the $q$-anisotropy in $\chi''(q,\omega)$ observed here at higher energies. In particular, the response is broader along $(1/2 - \xi, 1/2 + \xi)$ rather than the $(1/2 + \xi, 1/2 + \xi)$ for a given energy. It is interesting to note that highly structured magnetic excitations, characteristic of nesting, have recently been observed in FeTe$_{1-x}$Se$_x$. For FeTe$_{0.51}$Se$_{0.49}$, an anisotropic response similar to the one reported here is observed. The authors of Ref. 24 find that the Sato-Maki cross section (Eq. 5) provides a good description of their data over a 10–120 meV energy range. Motivated by this we also fitted our data to the Sato-Maki form (see Fig. 2). While the Sato-Maki function provides a reasonable description of the data, the phenomenological spin-wave cross section provides a better description at higher energies.

V. CONCLUSIONS

In summary, we have used inelastic neutron scattering to probe the collective spin excitations of near optimally doped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ ($x=0.065$). Strongly disper-
sive spin fluctuations are observed up to 80 meV. In the superconducting state, our measurements are consistent with a mode dispersing from the spin resonance near \( Q = (1/2, 1/2) \) to high energy. At higher energies, we observe excitations which are anisotropic as a function of in-plane wavevector. The response is centered on the \( M \) or \( (1/2, 1/2) \) position of the Brillouin zone and is broader in-plane wavevector. The response is centered on the \( Q \) observable excitations which are anisotropic as a function of \( \omega \), with a mode dispersing from the spin resonance near \( \Delta \). Our measurements are consistent with \( \Delta \) varying between zones in this case. The authors of Ref. 17 extended zone scheme in orthorhomic notation. The average shown in Fig. 5 of the present paper corresponds to averaging wavevectors over several orthorhomic Brillouin zones \((0 < h < 2, -1 < k < 1, 0 < l < 2)\). It is necessary to sample all representative wavevectors of \( \chi''(\mathbf{q}, \omega) \) which varies between zones in this case. The authors of Ref. 17 obtain a larger value for \( \chi''(\omega) \) because they average over the Brillouin zone where the response is strongest.

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18. Further details of the MAPS spectrometer are given on the ISIS website. (www.isis.rl.ac.uk).
19. In the present experiment we assume \( \chi''(\mathbf{q}, \omega) \) is isotropic with respect to field direction. Thus the susceptibilities quoted correspond to \( \chi'' = (1/3)(\chi''_{xx} + \chi''_{yy} + \chi''_{zz}) \).
20. The local susceptibility shown in Fig. 4 of Ref. 17 is defined by Eq. 2 but averaged over one Brillouin zone \((0.5 < h < 1.5, -0.5 < k < 0.5, 0.5 < l < 1.5)\) of the extended zone scheme in orthorhomic notation. The average shown in Fig. 5 of the present paper corresponds to averaging wavevectors over several orthorhomic Brillouin zones \((0 < h < 2, -1 < k < 1, 0 < l < 2)\). It is necessary to sample all representative wavevectors of \( \chi''(\mathbf{q}, \omega) \) which varies between zones in this case. The authors of Ref. 17 obtain a larger value for \( \chi''(\omega) \) because they average over the Brillouin zone where the response is strongest.
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