Doping Dependence of Spin Dynamics in Electron-Doped Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)As\(_2\)

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The spin dynamics in single crystal, electron-doped Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)As\(_2\) has been investigated by inelastic neutron scattering over the full range from undoped to the overdoped regime. We observe damped magnetic fluctuations in the normal state of the optimally doped compound \((x = 0.06)\) that share a remarkable similarity with those in the paramagnetic state of the parent compound \((x = 0)\). In the overdoped superconducting compound \((x = 0.14)\), magnetic excitations show a gap-like behavior, possibly related to a topological change in the hole Fermi surface (Lifshitz transition), while the imaginary part of the spin susceptibility \(\chi''\) prominently resembles that of the overdoped cuprates. For the heavily overdoped, non-superconducting compound \((x = 0.24)\) the magnetic scattering disappears, which could be attributed to the absence of a hole Fermi-surface pocket observed by photoemission.

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I. INTRODUCTION

One major difference between conventional and high-\(T_c\)-cuprate superconductors is the proximity to a competing magnetically ordered state in the latter, and it has long been believed that magnetic fluctuations could replace the role of phonons in mediating an electron-pairing interaction. This mechanism could give rise to more tightly bound Cooper pairs, elevating the transition temperature. The recent discovery of iron pnictide superconductors\(^1\) with \(T_c\) exceeding 50 K (Ref. 2) in close proximity to antiferromagnetic order reinvigorates this idea.

For the parent compounds of the cuprates, the magnetic properties are well described by the two-dimensional (2D) quantum non-linear sigma model\(^3\,^4\) and magnetic order is driven by a large instantaneous 2D correlation length, weak interlayer coupling, and spin anisotropies.\(^5\) For the iron pnictides, on the other hand, there is still much debate over the nature of the magnetism.\(^6\) For example, it remains controversial whether the stripe-type antiferromagnetic order in the parent compounds is stabilized by the spin-density-wave instability due to Fermi-surface nesting or by anisotropic in-plane exchange interactions due to 3d orbital ordering.\(^6\) More importantly, spin fluctuations in the doped compounds, which are arguably a key to understand the pairing mechanism, are largely unexplored. In particular, the question remains whether the normal-state spin fluctuations are simply governed by the Fermi-surface topology, or other effects, such as orbital fluctuations. Indeed, it has been proposed that orbital-spin fluctuations in a multiband ground state could give rise to the superconducting pairing.\(^7\) Measurements of magnetic fluctuations in the normal state should provide vital information to resolve these issues.

Hence our goal is to investigate the change in the spin dynamics as a function of doping and elucidate the interconnection with the band structure. For the electron-doped compounds, recent ARPES and transport studies clearly show the disappearance of the hole pockets around the antiferromagnetic zone center, i. e., a Lifshitz transition.\(^8\) This occurs for an electron doping \(x_L\) of Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)As\(_2\) \((0.15 < x_L < 0.3)\) from ARPES, whereas \(x_L \sim 0.1\) from transport measurements.\(^9\)\(^10\) The doping dependence of the spin dynamics, however, has not been comprehensively studied on single-crystal samples by neutron scattering. Previous work focused on powder samples\(^1\) or on the spin resonance in the optimally doped and underdoped compounds.\(^10\)\(^11\) Here we investigate the spin dynamics in single-crystal, electron-doped Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)As\(_2\) for \(x = 0, 0.06, 0.14,\) and 0.24, ranging from the parent compound to the heavily overdoped regime, with a particular emphasis on magnetic fluctuations in the normal state. We find that the magnetic fluctuations in the paramagnetic state of the parent compound are remarkably similar to those in the normal state of the optimally doped compound. On the other hand, the spin dynamics in the overdoped regime is drastically different, and resembles that in the overdoped cuprates. As the cobalt content increases well into the heavily overdoped regime, magnetic scattering disappears, coinciding with the absence of superconductivity.
II. EXPERIMENTAL DETAILS

All single-crystal samples were grown from a self-flux using the Bridgman method described in Ref. 25. The cobalt content was determined by energy dispersive x-ray analysis (EDX) using a scanning electron microscopy. Magnetic susceptibility of the ray analysis (EDX) using a scanning electron microscopy. The latter in the overdoped regime. For the place the former close to the optimally doped regime and 26 K and 7 K, respectively. The transition temperatures pounds exhibits superconducting transitions (onset) at 9.5 meV and 9 meV, respectively. For $x = 0$, background was estimated from constant-energy scans at $\hbar \omega = 3, 5, 8, \text{and } 15 \text{ meV}$. In (a), open red circles and open blue squares denote background calculated from constant-energy scans at 140 K and 250 K, respectively. For $x = 0.06$ and 0.14 background was measured away from the peak positions at $Q = (1, 2, 0, 1)$ and $(3, 0, 0)$, respectively. For $x = 0.24$, both constant-Q and constant-energy scans do not show scattering intensity above background, which was measured at $Q = (1.5, 0, 1)$. The dotted lines in (a), (b), and (c) denote the fitted background.

III. MAGNETIC FLUCTUATIONS IN THE NORMAL STATE

The scattering intensity can be written as $S_T(Q, \omega) = \langle n(\omega, T) + 1 \rangle \chi''(\mathbf{q}, \omega)$, where $n(\omega, T)$ is the Bose factor and $\chi''$ is the imaginary part of the spin susceptibil-
According to the theory of nearly antiferromagnetic metals, \( \chi''(q, \omega) \) is given by

\[
\chi''(q, \omega) = \frac{\chi_0(T)\Gamma(T)\hbar\omega}{(\hbar\omega)^2 + \Gamma(T)^2 \cdot (1 + D^2q_a^2 + F^2(q_a^2 + q_b^2))^2},
\]

where \( \Gamma \) is the damping constant, \( D \) and \( F \) represent the magnetic correlation lengths along the out-of-plane and in-plane directions, respectively, \( \chi_0 \) represents the isothermal susceptibility, and \( q = (q_a, q_b, q_c) \) is a wave vector away from an antiferromagnetic zone center. Background was estimated from a series of constant-energy scans or a constant-\( Q \) scan taken away from the antiferromagnetic wave vector and fit to a polynomial function of both momentum transfer and energy transfer. The coefficients of this polynomial were initially assumed to be temperature dependent.

### A. Undoped \( x = 0 \)

For the parent compound \( (x = 0) \), our analysis of the background shows weak temperature dependence in the measuring temperature range. We, therefore, assume temperature-independent background in the fitting (see Figure 2). The background-subtracted scattered intensity \( S_T(Q, \omega) \) was then converted to \( \chi'' \). Representative constant-\( Q \) scans, constant-energy scans, \( S_T(Q, \omega) \), and \( \chi'' \) measured in the paramagnetic state are shown in Figures 1(a), 2, and 4(a)-(b), respectively. \( \chi'' \) was fitted to the theory of nearly antiferromagnetic metals according to Eq. (1). In the fitting procedure, \( \chi_0 \) was constrained to obey the Curie law, since the data at high energy \( (\hbar\omega \gtrsim 15 \text{ meV}) \), which are required to uniquely determine \( \Gamma \) and \( \chi_0 \) at high temperatures, are not available. Error bars correspond to three times the standard deviation and in Figure 5 indicate large uncertainty at high temperatures, where \( \Gamma \) lies beyond the measuring energy range. We found that \( D \) and \( F \) do not change significantly within the measuring temperature range between 140 K and 250 K, and the difference lies within the uncertainties. Therefore, we were unable to determine their temperature-dependence and hence their values were fixed at 2.6(5) Å and 20(6) Å, respectively. We note that the uncertainties were obtained from fitting the 140 K data and that they could become larger at high temperatures (see Figure 2). We have previously reported these anisotropic magnetic fluctuations in the paramagnetic state of the parent compound at 145 K\(^{27} \). We have refitted those data using Eq. (1) (not shown), and have found that the lineshapes can be well described by Eq. (1) using the same parameters \( D \) and \( F \), while \( \Gamma \) at 145 K was obtained from the linear relation mentioned below (also see Figure 5).

At each temperature, the solid lines in Figures 1(a), 2 and 4(a)-(b) denote the global fits to several constant-energy scans and the constant-\( Q \) scan, convoluted with the four dimensional resolution function. The resulting fit parameter \( \Gamma \) is linearly proportional to temperature, e.g., \( \Gamma(T) = \alpha \cdot T \), where \( \alpha = 0.16(6) \text{ meV/K} \) as shown by the solid line in Figure 5. Note that \( \Gamma \) remains finite at the ordering temperature \( T_N = 136 \text{ K} \), and we do not observe any divergence of the correlation length at \( T_N \). Therefore, unlike the parent compounds of the cuprates, the observed magnetically ordered state in BaFe\(_2\)As\(_2\) is most likely not driven by the spin dynamics of the paramagnetic phase, but may be explained in light of the...
interconnection between the lattice and magnetic interactions.

**B. Optimally doped** \( x = 0.06 \)

For the optimally-doped compound \( (x = 0.06) \), the antiferromagnetic order is completely suppressed, and superconductivity emerges for \( T < T_c = 26 \) K. In the superconducting state, we observe the broad inelastic scattering centered at \( \hbar \omega = 9.6 \) meV (Figures 1(b) and 1(c)-(d)), in agreement with earlier reports,\(^{21,28}\) where the peak is attributed to the resonance mode. As temperature increases above \( T_c \), the inelastic peak is replaced by quasielastic magnetic fluctuations. Similar to the parent compound, the imaginary part of the spin susceptibility can be well described by the theory of nearly antiferromagnetic metals [Eq. (1)], which is shown by the solid lines in Figures 1(b), 3, and 1(c)-(d). The magnetic correlation length \( D \) (\( F \)) is equal to 2.4(6) Å [19(3) Å] at 30 K and 0.9(9) Å [12(3) Å] at 100 K. If measured at the same temperature, the correlation lengths measured in the optimally doped compound are shorter than those measured in the parent compound, which could be due to the change in the spin concentration upon doping. The in-plane magnetic correlation length is consistent with the earlier report on the \( x = 0.075 \) compound.\(^{28}\) We note that the out-of-plane magnetic correlation length was not measured in Ref. 28. This anisotropic magnetic fluctuations observed in the paramagnetic state of the parent compound and in the normal state of the optimally doped compound are reminiscent of the anisotropic exchange interactions measured in the ordered state of the parent compound.\(^{27}\)

We observe a marked similarity in the magnetic fluctuations in the normal state of the optimally doped compound and those in the magnetic state of the parent compound. More importantly, the temperature dependence of \( \Gamma \) follows the same linear relation as that observed in the parent compound (Figure 5). As a comparison, Inosov et al.\(^{28}\) reported that the temperature dependence of \( \Gamma \) follows a similar linear form \( \Gamma(T) = \alpha \cdot (T + \Theta) \) in the \( x = 0.075 \) compound \( (T_c = 25 \) K), where \( \alpha = 0.14(4) \) meV/K and \( \Theta \), the Curie-Weiss temperature, is equal to 30(10) K. The fact that the imaginary
part of the spin susceptibility in the parent and optimally
doped compounds can be well described by the theory of
nearly antiferromagnetic metals suggest that the mag-
netic fluctuations in the normal state of the parent and
optimally doped compounds could have a common ori-
gin, and are likely related to the presence of the quasi-
two-dimensional hole and electron pockets observed by
photoemission;\textsuperscript{15} more evidences of this interconnection
will be presented below.

C. Overdoped $x = 0.14$

In contrast to the parent and optimally doped com-
ounds, the spin dynamics in the overdoped, supercon-
ducting compounds ($x = 0.14$) shows the depletion of
the scattering intensity in the low-energy region and gap-
like excitations around 10 meV (Figures 4(c) and 4(e)–
(f)). Its peak profile cannot be fit to either Gaussian
or Lorentzian line shapes. Furthermore, the scatter-
ing intensity exhibits weak temperature dependence, and
the in-plane magnetic correlations are much shorter than
those of the parent and optimally doped compounds as
shown in the inset of Figure 4(c). The imaginary part
of the spin susceptibility [Figure 4(f)] displays linear
energy-dependence at low energy and a sharp drop at
high energy. With the exception of the gap-like behav-
ior, these magnetic excitations share many characteristics
with those observed in the overdoped cuprates.\textsuperscript{29}

The origin of the magnetic excitations in the over-
doped, superconducting compound ($x = 0.14$) is unclear
at the present moment. We note that $x = 0.14$ is a
higher doping than the neck-collapsing Lifshitz transition
($x \sim 0.1$), and is indeed very close to the hole-pocket-
vanishing Lifshitz point.\textsuperscript{14} Thus, it is likely that the dom-
inant part of the hole band lies below the Fermi level. Re-
cent theoretical calculations show that in such a case the
imaginary part of the spin susceptibility is strongly sup-
pressed giving rise to the pseudogap behavior,\textsuperscript{30} and such
a pseudogap was observed in NMR measurements in the
electron overdoped regimes of LaFeAsO$_{1-x}$F$_x$ (Ref. 31)
and Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$.\textsuperscript{32} Even though the gap-like be-
havior observed in our neutron scattering suggests that
the majority of the hole Fermi surface already diminishes
at $x = 0.14$, superconductivity with lower $T_c = 7$ K is
still observed. This superconductivity may then be of the
nodal type, where pairing is formed between electrons on
the same electron Fermi surface around the zone corners,
as has been theoretically proposed as one alternative to
the $s_\pm$ mechanism.\textsuperscript{33} A recent heat transport experiment
indeed indicates that the superconducting gap shows a
tendency to be strongly anisotropic as the electron dop-
ing increases in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$,\textsuperscript{34} although the mea-
surements were made only up to $x = 0.114$ so a direct
comparison with our result ($x = 0.14$) is not possible at
present.

D. Heavily overdoped $x = 0.24$

For the heavily overdoped, non-superconducting com-
 pound ($x = 0.24$), Figure 4(d) shows the suppression of
the magnetic scattering. NMR measurements on the
$x = 0.26$ compound,\textsuperscript{22} and neutron scattering measure-
ments on electron-doped LaFeAsO$_{1-x}$O$_x$ in the heavily
overdoped regime\textsuperscript{18} reveal the suppression of the spin
fluctuations consistent with our result. In this regime,
both photoemission measurements and first-principles
calculations point to the disappearance of a hole Fermi-
surface pocket.\textsuperscript{12,15} Our result, therefore, further sug-
gests the correlation between the electronic band struc-
ture and magnetism, and supports the scenario that the
magnetic fluctuations in the underdoped and optimally
doped regimes, which serve as a precursor to supercon-
ductivity, originate from quasiparticle scattering across
the electron and hole pockets.\textsuperscript{25}

IV. CONCLUSION

We have studied the spin dynamics in electron-doped
Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ at four cobalt concentrations. We ob-
serv e a striking similarity between the magnetic fluctua-
tions in the paramagnetic state of the parent compound
and those in the normal state of the optimally doped
compound, and the suppression of the magnetic signal in
the heavily overdoped regime, in which superconductiv-
dity disappears. These two results suggest that magnetism
and superconductivity are strongly correlated. On the
other hand, magnetic excitations in the vicinity of the
hole-pocket-vanishing Lifshitz point are markedly differ-
ent from those in the underdoped regime, with the emer-
gence of a spin gap and much weaker temperature de-
pendence of $\chi''$. These changes in the spin dynamics at
different doping levels likely reflect changes in the elec-
tronic band structure. Further experimental and theo-
retical work is desirable to examine the interconnection
between the band structure, spin fluctuations, and su-
perconducting pairing mechanism.

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