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Filamentary superconductivity across the phase diagram of Ba(Fe,Co)\textsubscript{2}As\textsubscript{2}

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We show magnetotransport results on Ba(Fe\textsubscript{1−x}Co\textsubscript{x})\textsubscript{2}As\textsubscript{2} (0.0 ≤ x ≤ 0.13) single crystals. We identify the low-temperature resistance step at 23 K in the parent compound with the onset of filamentary superconductivity (FLSC), which is suppressed by an applied magnetic field in a similar manner to the suppression of bulk superconductivity (SC) in doped samples. FLSC is found to persist across the phase diagram until the long-range antiferromagnetic order is completely suppressed. A significant suppression of FLSC occurs for 0.02 < x < 0.04, the doping concentration where bulk SC emerges. Based on these results and the recent report of an electronic anisotropy maximum for 0.02 ≤ x ≤ 0.04 [Chu et al., Science 329, 824 (2010)], we speculate that, besides spin fluctuations, orbital fluctuations may also play an important role in the emergence of SC in iron-based superconductors.

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I. INTRODUCTION

The origin and mechanism of unconventional superconductivity (SC) remain central issues in modern condensed-matter physics. Similarly to copper-oxide and heavy-fermion superconductors, the unconventional SC in iron-based superconductors arises in close proximity to antiferromagnetism (AFM). The parent compound of the iron-based superconductors is an AFM metal. Upon doping or applying pressure, the AFM order is suppressed and gives way to SC. However, unlike the copper-oxide and heavy-fermion superconductors, which mostly are d-wave superconductors, the pairing symmetry in this new class of superconductors is more complicated. It can be either $s_{±±}$ or $s_{±}$. There is also evidence for nodal superconductivity. Therefore the origin of iron pnictide SC is still of intense debate.

Some studies have shown the importance of spin fluctuations, while others have shown the importance of orbital fluctuations to the emergence of SC. Specifically, on one hand, experimental and theoretical studies show evidence that the electron pairing is mediated by antiferromagnetic spin fluctuations and that SC nucleates at antiphase domain boundaries suggesting that spin fluctuations play an important role in the emergence of SC. On the other hand, there is evidence that the multiorbital band structure near the Fermi energy in iron pnictides could enhance orbital fluctuations that become important to the emergence of SC. Particularly, it has been shown that the uneven occupation of the d orbitals makes the orthorhombic crystal structure more energetically favorable, thus inducing a structural phase transition at $T_c$. However, the large electronic anisotropy revealed in Ba(Fe\textsubscript{1−x}Co\textsubscript{x})\textsubscript{2}As\textsubscript{2} precisely where the crystal’s C\textsubscript{4} rotational symmetry is broken cannot be explained based on the 1% lattice distortion at $T_c$. In fact, the presence of a C\textsubscript{4} structural to C\textsubscript{2} electronic symmetry transition in the quasiparticle interference maps of Ca(Fe\textsubscript{1−x}Co\textsubscript{x})\textsubscript{2}As\textsubscript{2} has been shown. This electronic symmetry transition could be a result of orbital ordering due to the above-mentioned inequivalent occupation of the $d_{xz}$ and $d_{yz}$ orbitals.

Based on these results, we found that it is imperative to perform measurements that could simultaneously reveal the effect of spin and orbital fluctuations on the emergence of SC in iron-based superconductors. In this paper, we reveal through magnetoresistivity studies the presence of filamentary superconductivity (FLSC) over a wide range of the phase diagram of Ba(Fe\textsubscript{1−x}Co\textsubscript{x})\textsubscript{2}As\textsubscript{2} (0 ≤ x ≤ 0.13), from the parent AFM compound to the optimally doped region, and its disappearance in the overdoped region where the AFM order is suppressed. We show that FLSC, which might have the same origin as the bulk SC, coexists with AFM as a result of competing SC and AFM orders. According to our previous work, FLSC and AFM fluctuations are closely correlated. The suppression of the temperature $T_{fj}$ where FLSC sets in is over the same doping range where a maximum in the orbital order emerges. This might suggest that the orbital and SC orders are also competing orders. Hence we speculate that both spin and orbital fluctuations may play important roles for the occurrence of SC in iron-based superconductors.

II. EXPERIMENTAL DETAILS

High quality single crystals of Ba(Fe\textsubscript{1−x}Co\textsubscript{x})\textsubscript{2}As\textsubscript{2} were grown using the FeAs flux method. Powder x-ray-diffraction measurements were done and the amount of impurities is below the sensitivity of the machine. Typical dimensions of the single crystals are $2 \times 0.5 \times 0.06$ mm$^3$. The in-plane resistivity $\rho$ was measured using the electrical contact configuration of the flux transformer geometry and multiple electrodes were fabricated on each sample by bonding Au wires to the crystal
with H2OE epoxy paste. The current I was applied in the 
ab plane and the magnetic field H (up to 14 T) was applied 
parallel to the c axis of the crystals.

III. RESULTS AND DISCUSSION

The samples studied in this work have an actual Co 
concentration 0.0 \leq x \leq 0.13. The values of the overdoped 
samples were determined based on the value of the resistive 
Tc. To determine the actual Co concentration for all the other 
samples we extracted the structural Tc and antiferromagnetic 
TNY phase transition temperatures from the derivative of the 
resistivity curve [inset to Fig. 1(b)]. Specifically, upon cooling 
from high temperatures \(d\rho/dT\) shows a change in slope, 
followed by a sharp dip as indicated by the arrows. We used 
these two features to determine Tc and TNY, respectively, in 
correlation to neutron and x-ray measurements.15 Knowing 
these transition temperatures for each sample, we determined 
the actual x value from the temperature-doping (T-x) phase 
diagram of Fig. 3 (Ref. 16).

Figure 1(a) depicts the reduced resistivity \(\rho/\rho(300 \text{K})\) 
data for the parent compound (no Co doping) measured up 
to 14 T, while its inset shows the zero-field data measured 
over a wide temperature range (2–300 K). Notice the presence 
of a small step (decrease) in the data of the main panel 
which shifts to lower temperatures with increasing magnetic 
field (see arrows). Based on a recent report on undoped 
antiferromagnetic CaFe2As2,5 we identify the temperature 
\(T_{fl}\) (\(H_{fl} = 23.5 \text{K} \) in the zero-field resistivity data) of this step in 
resistivity with the onset of filamentary superconductivity.

Figure 1(b) shows the H-T phase diagrams generated from 
the field dependence of \(T_{fl}\) of the x = 0 sample (magenta 
squares) and an almost optimally doped (x = 0.06) single 
crystal with bulk \(Tc = 25.1 \text{K} \) (green circles). [Its zero-
field-resistivity data are shown in the inset to Fig. 1(a).] A 
comparison of these H-T phase diagrams shows the striking 
similarity between them, with the zero-field \(T_{fl}\) (x = 0) and 
\(Tc\) (x = 0.06) within two degrees of each other, and their 
similar suppression by magnetic field. The H-T data of the 
undoped and optimal doped samples can scale together (data 
not shown), which might suggest that FLSC and bulk SC have 
a common origin. Linear fits of these two sets of data give 
upper critical field \(H_{c2}(0) = 30.4 \text{T} \), \(Tc = 19.5 \text{K} \), and a slope 
of \(-1.56 \text{T/K}\) for the x = 0 sample, and \(H_{c2} = 50.3 \text{T} \), \(Tc = 22.9 \text{K} \), and a slope of \(-2.2 \text{T/K}\) for the x = 0.06 sample. A 
fit with the Ginzburg-Landau (GL) expression for the upper 
critical field, \(H_{c2}(T) = H_{c2}(0)\left[1 - (T/Tc)^2\right]^{1/2}\), yields 
\(H_{c2}(0) = 25.9 \text{T} \) and \(Tc = 19.7 \text{K} \) for the x = 0 sample, and 
\(H_{c2}(0) = 43.9 \text{T} \) and \(Tc = 22.9 \text{K} \) for the x = 0.06 sample. The 
dashed curve is a fit to the Werthamer-Helfand-Hohenberg 
(WHH) relation, \(H_{c2}(0) = -0.7Tc(\rho_{c2}/\rho_{c2}), \) which gives 
\(H_{c2}(0) = 21.3 \text{T} \) for the x = 0 sample, and \(H_{c2}(0) = 35.3 \text{T} \) 
for the x = 0.06 sample. The red solid curves show the 
high-temperature tails for both dopings, typical in iron-based 
superconductors.

It has been argued that the filamentary SC reported in 
CaFe2As2 suggests that even the nominally pure stoichiometric 
material can spontaneously become electronically inhomoge-
nous at the nanoscale.5 Such an electronic inhomogeneity 
at the nanoscale occurs spontaneously in a nominally uniform 
system due to competing interactions or competing orders (see 
Ref. 17 and references therein). Inhomogeneity is favorable for 
subdominant order to emerge locally, in regions where the 
competing dominant order vanishes. For example, AFM order 
can be present inside the vortex core18 and FLSC nucleates at 
the domain walls in the antiferromagnetically ordered parent 
compound.5 The very similar H-T phase diagrams of undoped 
BaFe2As2 and undoped antiferromagnetic CaFe2As2 (Ref. 5) 
show a common signature for FLSC in these parent compounds 
and therefore imply that the AFM order is also a competing 
order to superconductivity in the BaFe2As2 system.

We note that the step in the low-temperature resistivity 
[see Fig. 1(a)] is not always observed in the undoped 
samples.13,19-21 For example, Tanatar et al., studied five 
different samples of undoped BaFe2As2 and found that only 
two samples showed the partial SC transition.22 We also made 
measurements on a number of single crystals of the parent 
compound and confirmed that this step is observed only for

FIG. 1. (Color online) (a) Temperature T dependence of the 
reduced resistivity \(\rho/\rho(300 \text{K})\) curves for BeFe2As2 measured in 
a current of 1 mA and different applied magnetic fields, i.e., \(H = 
0, 0.5, 1, 2, 4, 6, 10, 14 \text{T} \) (H \(\parallel c\)). Inset: Zero-field resistivity 
curves of BaFe1−xCo2xAs2 (x = 0 and 0.06) measured over a 
wide temperature range (2–300 K). (b) H-T phase diagrams of 
BaFe1−xCo2xAs2 (x = 0 and 0.06). The black solid curves are fits 
of the data to the GL expression for the upper critical field; the dotted 
lines are fits with the Werthamer-Helfand-Hohenberg relationship. 
The red solid curves are guides to the eye. Inset: Reduced resistivity 
\(\rho/\rho(300 \text{K})\) curve and the derivative curve \(d\rho/dT\), for x = 0.03 
sample.
small and thin single crystals, (with a thickness of ∼50–60 μm) with shiny surfaces (after careful cleaving), whereas in the resistivity of larger samples, which contain a number of smaller crystals, this feature is not there. High quality single crystals are therefore better for the detection of filamentary superconductivity. A similar conclusion has been reached by Park et al., in their study of textured SC in CeRhIn5.23

Since the step in resistivity is sometimes hard to detect for reasons discussed above, we performed magnetoresistivity (MR) measurements, which is a more sensitive method to detect the small decrease in resistivity due to FLSC. To determine how FLSC evolves across the phase diagram of Ba(Fe,Co)2As2, we measured the temperature dependence of the magnetoresistivity \( \Delta \rho / \rho(0) \equiv [\rho(14T) - \rho(0)] / \rho(0) \) of single crystals with different Co doping [Fig. 2(a)]. With decreasing temperature, the magnetoresistivity first increases almost linearly, then shows a sudden deviation (kink) at a certain temperature. We associate this sudden change in magnetoresistivity with the appearance of FLSC and identify its temperature as \( T_{fl} \) for all the samples studied in this work since we confirmed that the temperature of this kink in the magnetoresistivity of the parent compound coincides with \( T_{fl} \) and, in addition, FLSC, like SC, is strongly affected by magnetic field. We summarize the doping dependence of \( T_{fl} \) in Figs. 2(b) and 3.

It is worthwhile to mention that we compared samples with the same Co concentration but different thicknesses and found that as the thickness decreases, the signature of FLSC (step in resistivity) is more pronounced. However, the kink in the MR data is at the same \( T_{fl} \), for all samples (regardless their thickness), even for the thick samples that do not show any low-temperature step in their resistivity. Furthermore, the magnitude of MR when approaching \( T_{fl} \) from above is the same regardless the thickness of the sample. Therefore we conclude that, even though the low-temperature step in resistivity is not always detectable, FLSC exists in all samples and can be better detected through the more sensitive MR measurement.

The plot of Fig. 2(b) shows that, in fact, \( T_{fl} \) displays a nonmonotonic dependence on Co doping, with a minimum for \( x \approx 0.02–0.03 \). Interestingly, Chu et al.9 have recently reported a maximum electronic nematic order over the same range of Co concentrations, around the onset of bulk superconductivity \( (x = 0.03) \). This suggests a maximum orbital order around the onset of bulk SC. The fact that \( T_{fl} \) of FLSC shows a minimum over this same doping range may suggest that orbital and SC orders are also competing orders. So orbital fluctuations could also contribute to the pairing mechanism in iron-based superconductors. Nevertheless, since, as discussed in our previous work,5 FLSC nucleates at the domain walls, a magnetic origin of SC is an important ingredient. So spin and orbital degrees of freedom might be strongly coupled to each other. We speculate that both spin and orbital fluctuations might contribute to the pairing mechanism of unconventional superconductivity in iron-based superconductors.

Figure 3 depicts a composite plot of the phase diagram of the doping dependence of \( T_s, T_N, T_c \), and \( T_{fl} \). \( T_s \) and \( T_N \) nearly coincide at \( x = 0 \) but they are slightly different for the underdoped samples. Increasing doping suppresses the AFM order, gradually giving way to superconductivity. Bulk superconductivity (triangles, determined from magnetization measurements, data not shown) emerges at \( x \approx 0.03 \) and \( T_c \) attains a maximum value for \( x \approx 0.06 \), where the zero-field magnetic and SC transition temperatures are equal. Compared with the bulk \( T_c, T_{fl} \) (stars) is less doping dependent, FLSC is already present in the undoped, parent compound, and persists up to about \( x = 0.06 \), where long-range AFM order vanishes.
This latter fact suggests again that there is a close relationship between FLSC and AFM. In fact, previous experiments on CaFe$_2$As$_2$ showed that FLSC nucleates at the AFM domain walls,\cite{5} i.e., where the AFM exchange interaction is suppressed and the local AFM fluctuations are enhanced. When the AFM transition temperature \(T_{\text{AFM}}\) is close to optimum \(T_c\) is, indeed, a result of the suppression of \(T_c\) emerging where the AFM fluctuations \(T_{\text{FLSC}}\) sets in is close to the optimal bulk transition temperature \(T_c^0\), suggesting that FLSC is a precursor state to bulk SC. Since the suppression of \(T_{\text{FLSC}}\) in the vicinity of the onset of bulk superconductivity \((x \approx 0.03)\), in the doping range where a maximum in the orbital order emerges, we speculate that besides spin fluctuations, orbital fluctuations may also play an important role in the emergence of unconventional SC in iron-based superconductors.

IV. SUMMARY

In summary, we show that FLSC, which might have the same origin as the bulk SC, coexists with AFM as a result of competing SC and AFM orders. The temperature \(T_{\text{FLSC}}\) where FLSC sets in is close to the optimal bulk transition temperature \(T_c\), suggesting that FLSC is a precursor state to bulk SC. Since the suppression of \(T_{\text{FLSC}}\) is in the vicinity of the onset of bulk superconductivity \((x \approx 0.03)\), in the doping range where a maximum in the orbital order emerges, we speculate that besides spin fluctuations, orbital fluctuations may also play an important role in the emergence of unconventional SC in iron-based superconductors.

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FIG. 3. (Color online) Temperature \(T\) vs doping \(x\) phase diagram of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ single crystals. The phase boundaries are taken from Ref. \cite{16}. The open symbols represent data from this work: structural \(T_s\) (squares), antiferromagnetic \(T_N\) (circles), filamentary superconductivity \(T_{\text{FLSC}}\) (stars), and bulk superconductivity \(T_c\) (triangles) transition temperature determined from magnetization measurements (data not shown). The red and black solid lines denote first-order and second-order transitions, respectively (taken from Ref. \cite{24}). The pink solid circle is the magnetic tricritical point, \(x_{\text{tr}}^0 = 0.022\).\cite{24,25}

\[ T_{\text{FLSC}}(x) \approx T_{\text{bulk}}(x) \approx T_{\text{AFM}}(x) \]

\[ x_{\text{tr}}^0 \approx 0.022 \]

\[ T_{\text{FLSC}} \approx T_{\text{bulk}} \approx T_{\text{AFM}} \]

\[ x_{\text{tr}}^0 \approx 0.022 \]

\[ T_{\text{FLSC}} \approx T_{\text{bulk}} \approx T_{\text{AFM}} \]

\[ x_{\text{tr}}^0 \approx 0.022 \]


