Observation of anomalous admixture of superconducting and magnetic fractions in BaFe$_{2-x}$Co$_x$As$_2$ single crystals

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Abstract

Increasing doping concentration ($x$) in pnictide superconductors leads to a loss of long range magnetic order and the emergence of superconductivity. In these doped compounds the role of magnetic fluctuations and their effect on superconductivity is not well understood. Below $T_c$, in optimally doped BaFe$_{2-x}$Co$_x$As$_2$ single crystals at low applied magnetic fields ($H$), we observe a coexistence of regions with both positive and negative (diamagnetic) magnetization response. With increasing $H$ the positive magnetization response weakens and the diamagnetic signal associated with superconductivity enhances. Above $T_c$, resistivity and magnetization measurements in the normal state of the material reveal the presence of a weak diamagnetic response admixed with the magnetic response of a magnetized state. Magneto-optical imaging studies in the optimally doped sample shows a spatially inhomogeneous local magnetic response in the normal state with local regions having diamagnetic like response existing alongside magnetized regions with short range magnetic order. Above $T_c$, the estimated volume fraction with diamagnetic fluctuations is found to be maximum in the optimally doped sample. We present a doping dependent $H$–$T$ diagram identifying different fluctuation regimes. We propose our results suggest the admixture of magnetic and superconducting fractions in BaFe$_{2-x}$Co$_x$As$_2$ and a close correlation between them.

1. Introduction

The doping phase diagram in Pnictide superconductors has magneto-structural transformation boundaries interrupted by a superconducting dome [1–8]. Unraveling the intricate relationship between superconductivity, magnetism and structural transformation in pnictides is a topic of current interest. In general it has been found that with increasing doping, long range magnetic order gets suppressed and superconductivity emerges in these compounds. The bulk superconducting transition temperature ($T_c$) is maximum at optimal doping concentration ($x$). MuSR studies on BaFe$_{1-x}$Co$_x$As$_2$ have shown that while Co doping results in loss of long range magnetic order, short range order may still be present in the system [9–12]. Iron isotope replacement studies in doped 1111 and 122 pnictides show that not only the bulk superconducting transition temperature ($T_c$) but the magnetic ordering temperature ($T_N$) also gets affected, suggesting a close interplay between magnetism and superconductivity [13] in these compounds. Studies on underdoped pnictides show the coexistence of superconductivity (SC) along with magnetic fluctuations [14–20] below $T_c$. Theories have also suggested that magnetism plays an important role in mediating strong superconducting pairing correlations in pnictides [21–29]. It is plausible that magnetism of some form is important for sustaining superconducting correlations in these doped compounds. ARPES studies in a BaFe$_2$(As$_{1-x}$P$_x$)$_2$ suggest the presence of local superconducting correlations present above $T_c$ [30]. Recent NMR studies suggest evidence for a pseudogap like state in CaFe$_{1-x}$Co$_x$As$_2$ compound [31]. However STM study in BaFe$_{1-x}$Co$_x$As$_2$ did not find evidence of superconducting gap like features above $T_c$ [32]. Another study in FeSe crystals suggested the presence of
preformed pairs above $T_c$ [33]. From the above it also appears that evidence for the presence of superconducting fluctuations above $T_c$ in pnictides is not a well resolved issue. In this article we present our results on the behavior of local and bulk magnetization response measured in, under, optimally and over, doped, superconducting BaFe$_2$–$x$Co$_x$As$_2$ single crystals. At low applied magnetic field ($H$), for $T < T_c$, local magnetization measurements show the presence of weak positive magnetization response in the optimally doped sample. The local positive magnetic response transforms into a diamagnetic response with increasing $H$. Just above $T_c$, in all the samples the bulk magnetization response becomes positive and exhibits a maxima, with a long tail extending up to high $T$. Analysis of magnetization and electrical conductivity in the normal state (above $T_c$) reveals the presence of a weak diamagnetic response buried within the response of a magnetic background which has short range magnetic order. Sensitive magneto-optical imaging studies performed at $T > T_c$ reveals a spatially inhomogeneous magnetic response present in the normal state of the sample. The inhomogeneous response is associated with local regions exhibiting weak diamagnetic response existing alongside regions with magnetic fluctuations which exhibit positive magnetic response. We observe that the sample with optimum doping concentration exhibits the strongest effect of magnetic fluctuations and the same sample also has the highest volume fraction with diamagnetic response above $T_c$. Using the data gathered for each doping concentration, we construct the $H$–$T$ diagrams in which different fluctuation regimes are identified. We believe our results suggest a coexistence of both magnetic and superconducting fluctuations above and below $T_c$ and a close correlation exists between them in mediating superconductivity in the BaFe$_2$–$x$Co$_x$As$_2$ system.

2. Experimental details

The single crystals of BaFe$_2$–$x$Co$_x$As$_2$ with $x = 0.10, 0.14,$ and $0.20$ were prepared using self-flux method [34]. The bulk $T_c$ was identified from the onset of diamagnetism in a $T$ dependent magnetization $M(T)$ measurement at low $H$ and also from the sharp drop in zero field resistance, $R(T)$, data ($T_c$’s are indicated by arrows in figures 1 (a)–(c)). The $T_c$’s for the three samples with different doping concentrations are tabulated in table 1. The $T_c$’s determined from $M(T)$ and $R(T)$ differ slightly as they were measured in different cryogenic systems. Based on the variation of $T_c$ with Co doping concentration ($x$) (see table 1), we label the sample with $x = 0.14$ as optimally doped (with the highest $T_c(0) = 26.80$ K), $x = 0.10$ as underdoped and $x = 0.20$ as overdoped sample. In table 1 a summary of the electron probe micro analysis (EPMA) investigation indicates a small error in $x_{\text{EPMA}}$ value of Co concentration, suggesting negligible local variation in the concentration of Co across the samples. The $T_c(x)$ in table 1 is consistent with earlier reports [1] (note in [1], $x$ is defined as Ba(Fe$_{1−x}$Co$_x$)$_2$As$_2$ while we define $x$ as BaFe$_{2−x}$Co$_x$As$_2$). The RRR for $x = 0.10, x = 0.14$ and $x = 0.20$ samples were measured to be 1.92, 2.45 and 2.71, respectively, and resistivity at 30 K was about 100 $\Omega$ cm. The local magnetic field distribution $B_z(x, y)$ (where the $x, y, z$-coordinates along the sample surface are $(x, y, z)$ and $z$ is along the vertical direction) at different $H$ and $T$ was measured using high sensitivity magneto-optical (MO) imaging [35–37] technique. In all our measurements, $H$ was applied parallel to crystallographic $c$-axis of the crystals ($H \parallel c$). The details of our MOI set up have been presented elsewhere [37]. Briefly, in the MOI technique (performed in the reflection mode), linearly polarized light is reflected from the sample surface on which a thin layer of Faraday active material (YIG) with high Verdet’s constant is placed. The reflected light is Faraday rotated by an angle proportional to the local $B_z$ on the sample. By sensitively imaging and calibrating the reflected Faraday rotated light intensity measured across the sample ($I(x, y)$), one infers the local $B_z(x, y)$ across the sample surface, as $I(x, y) \propto |B_z(x, y)|^2$ [35].

We also employ a differential magneto-optical (DMO) imaging technique [38–40] to image the differential changes in $B_z$ produced in the sample in response to modulation of the externally applied field, $H$. In the DMO technique, which is a variant of the MOI technique, we obtain DMO images in the MOI setup by periodically modulating $H$ by an amount $\delta H = 1$ Oe and capturing $k$ nos of MO images at $H$ and $H + \delta H$, namely, $I_i(H)$ and $I_i(H + \delta H)$, where $i$ is the index of the image ($I$) captured at $H$ or $H + \delta H$. The differential image, is

$$\delta I(x, y) = \frac{\sum_{i=1}^{k} I_i(H + \delta H)}{k} - \frac{\sum_{i=1}^{k} I_i(H)}{k} (k = 20 \text{ is used}).$$

It can be shown [38–40] that in response to external $H$ modulation, as $B_z$ changes by $\delta B_z$, $\delta I(x, y) \propto B_z \delta B_z$ for small rotation angles and hence $\delta I / I \sim \delta B_z / B_z$ for moderately low $B_z$ values.
3. Results and discussion

3.1. Zero magnetic field transport measurement

Inset of figure 1(a) shows the normalized $R(T)$ (normalized with $R$ at 300 K) behavior for all three samples at $H = 0$ Oe. The normalized $R(T)$ for $x = 0.10$ (underdoped) sample shows an anomalous upturn in $R$ at 67 K, which is associated with the magnetico-structural transition found in underdoped pnictides [1]. For the optimally doped and overdoped samples, the absence of any such anomaly in $R(T)$ suggests the suppression of long range magnetic order with increasing Co doping concentration.

3.2. Bulk magnetization response in the superconducting state

Figures 1(a)–(c) show the bulk zero field cooled (ZFC), $4\pi M$ versus $T$ at different $H$ for $x = 0.10, x = 0.14$ and $x = 0.20$ samples respectively. At $T \ll T_c$ and low $H$, the $4\pi M/H$ values saturate to a value close to $-1$, indicating a strong bulk Meissner response in all the samples. In figures 1(a)–(c) the diamagnetic $4\pi M(T)/H$...
value decreases from \(-1\) with increasing \(H\), due to the penetration of vortices into the bulk of the superconductor. The \(M(H)\) hysteresis observed at 5 K (<\(T_c\)) for underdoped (red), optimally doped (green) and overdoped (blue) superconductor in figure 1(d) confirms the presence of bulk pinning in these samples. The above features in \(M(T)\) and \(M(H)\) in figures 1(a)–(d) confirm bulk type II superconductivity in all the three samples. By determining the maximum width, \(\Delta M(H)\) of the \(M(H)\) hysteresis loop for the optimally doped (figure 1(d)), the maximum critical current density, \(J_\text{c} = 20\Delta M/a(1 - a/3b)\) [41] (for a rectangular plate like crystal geometry, with in–plane sample dimensions \(a, b(b > a)\)) was estimated to be about \(8 \times 10^7\) Amp cm\(^{-2}\), which is comparable with similar values reported in BaFe\(_{2.4}\)Co\(_{0.6}\)As\(_2\) samples [42].

### 3.3. Bulk magnetization response in the normal state

For any conventional superconductor, the diamagnetic \(M(T)\) response is expected to monotonically approach zero as \(T \rightarrow T_c(H)\). Instead, figures 2(a)–(c) show that at \(T > T_c(H)\) the \(M(T)\) crosses zero and becomes positive above \(T_c(H)\). The positive \(M(T)\) behavior has a maxima at \(T_p\) with a peak height \(M_{\text{pzc}}\). Beyond \(T_p\), there is a long tail in \(M(T)\) extending up to \(T \gg T_c(H)\). At first glance, the shape of the positive \(M(T)\) behavior can be mistaken for the Wohlleben or the Paramagnetic Meissner Effect (PME) [43, 44]. The PME is a positive \(M(T)\) response with a peak like feature found in superconductors which have been field cooled (FC) in a small \(H\). Also in PME, the (positive) peak value of \(M\) is known to decrease in height with increasing \(H\) [43, 44]. Non-uniform field cooling of a superconductor or surface superconductivity effects generate magnetic flux compression leading to the positive PME peak [43, 44]. However, we argue that the features in figures 2(a)–(c) are not associated with PME because, (i) the peak at \(T_p\) is found in a ZFC-\(M(T)\) measurement rather than in an FC measurement and (ii) the \(M_{\text{pzc}}\) increases with increasing \(H\) rather than decreasing in increasing \(H\). Infact our observation that \(M_{\text{pzc}}\) increases with \(H\) suggests a magnetic origin of this positive \(M(T)\) response found above \(T_c\). Figures 2(a)–(c) show that for similar \(H\) (for e.g. 200 Oe) the \(M_{\text{pzc}}\) values are almost comparable for \(x = 0.10\) and \(x = 0.14\) samples while it is smaller for the \(x = 0.20\) sample. At same \(H\) the \(M_{\text{pzc}}\) value is the smallest for \(x = 0.20\) sample despite the fact that this sample is expected to have the highest Co doping concentration. The above observations suggest that the \(M_{\text{pzc}}\) feature is not directly linked to the concentration of Co.

### 3.4. Imaging local magnetic field distribution across the \(x = 0.14\) sample below and above \(T_c\)

For any conventional superconductor, the local magnetic field inside the sample should be zero at \(H = 0\) Oe, i.e., no MO contrast difference should exist between the sample and outside it as both regions have \(B = 0\) G. However, the MO image of the optimally doped sample in figure 3(a) obtained at 11 K and \(H = 0\) Oe, shows an unconventional feature: the presence of a bright MO image contrast inside the sample for \(H = 0\) Oe. The bright contrast corresponds to a non-zero mean \(B_x \sim 35\) G (see figure 3(e)), which is higher than the value in the regions outside the sample. Furthermore in a conventional superconductor due to vortex penetration, the MO contrast should change from dark \((B_x \approx 0\) G\) to bright \((B_x = 0\) G\) with increasing \(H\). However, figures 3(a)–(d) show the MO contrast inside the sample changes from bright to dark as \(H\) is increased. The observed unusual darkening of the MO contrast (figures 3(a)–(d)) suggests an increase in diamagnetic shielding response with increase in \(H\).

The \(B_x(r)\) profile measured at different \(H\) along representative horizontal solid and dashed lines (see figure 3(a)), are plotted in figures 3(e) and (f) respectively, where \(r\) is the distance measured along the line. From figure 3(e), it is evident that the \(B_x(r)\) profile inside the sample at \(H = 0\) Oe shows non–Meissner like features \((B_x = 0\) G\) with \(B_x \sim 35\) G. Outside the sample, far away from its edges (the edges identified by vertical dashed lines in figures 3(e) and (f)) \(B_x \approx H\). Near the sample edges, \(B_r > H\) due to presence of circulating Meissner screening currents. In figures 3(a)–(d), the MO contrast changing from bright to dark with increasing \(H\) corresponds to the \(z\)-component of the magnetization (estimated using \(B_x(r)–H\)) of the sample changing sign from positive to negative (see figure 3(f)). In figure 3(e) one observes that deep inside the sample \(B_x–H\) is positive \((B_x(100 \mu m) \sim 35\) G\) for low fields \((0 < H \leq 40\) Oe\). As \(H\) is increased up to 705 Oe (see figure 3(f)), \(B_x–H\) turns negative (note that a negative \(B_x–H\) is a characteristic signature of diamagnetic shielding response in a superconductor). Thus at low \(H\) (in the range of few tens of Gauss) the local magnetization response is positive corresponding to positively magnetized regions in the sample. At higher \(H\) the magnetic response is dominated by the diamagnetic response of the superconductor.

In figures 3(g)–(i), for \(H = 0\) Oe and \(T > T_c\), we observe a bright magneto–optical intensity \((B_x > 0\) G\) uniformly present across the sample surface albeit its intensity decreases as \(T \sim 220\) K (figure 3(i)). Figure 3(e) inset shows \(B_y(r) \approx 0\) G (as measured across the line in the MO image in figure 3(i)) at 220 K and 0 Oe, indicating that the non-zero \(B_x\) present across the sample above \(T_c\) weakens and disappears by 220 K. We associate the positive \(B_x\) value observed above \(T_c\) at 0 Oe (figures 3(g)–(i)) with the presence of short range magnetic order in the optimally doped sample. Neutron diffraction and \(\mu\)SR studies on Co doped BaFe\(_{2.4}\)As\(_2\) system had already suggested that while doping destroys long range magnetic correlations between Fe moments, short range
magnetic order may still be preserved locally [9–11]. In our local and bulk magnetization measurements, we believe that the weak positive magnetic response found above \( T_c \) is associated with short range magnetic order due to the following observations: (i) the \( M \) value increases with \( H \) (see figures 2(a)–(c)), (ii) presence of local magnetization hysteresis above \( T_c \) (as seen in figure 3(i)) along with weak hysteretic behavior suggests, that above \( T_c \), the local magnetic response is unlike a conventional paramagnetic response (section I of supplementary information shows the \( T \)-dependence of the local magnetization hysteresis loops above \( T_c \)). From the behavior of the hysteresis loops especially at high \( T \), we argue the presence of short range magnetic order present in the system above \( T_c \).

3.5. Identification of characteristic temperatures

Based on the above observations in figures 2 and 3, we attribute the positive magnetization response found above \( T_c \) with the presence of short range magnetic order at \( T < T_{\text{mf}} \) (mf: magnetic fluctuations). From figures 2(d)–(f), we identify \( T_{\text{mf}} \) as the temperature below which one observes a paramagnetic Curie like \( 1/T \) dependence of \( M \). However, the observed local hysteresis and nonlinear \( M(H) \) dependence in \( x = 0.14 \) sample, as discussed in
suggests that it is not possible to attribute the Curie like behavior below $T_{mf}$ to a conventional paramagnetic response. We believe that the regions possessing short range magnetic correlations fluctuate due to thermal fluctuations at finite $T$ resulting a Curie like $1/T$ dependence in $M(T)$ above $T_c$, as seen in figures 2(d)–(f). Such a feature of a system with short range magnetic order exhibiting paramagnetic like features at a finite temperature is not uncommon. In the context of nanomagnetic systems one observes the superparamagnetic effect where above a temperature called the blocking temperature, the magnetic orientation in these systems is not stable due to thermal fluctuations being large compared to the magnetic anisotropy energy. In such low dimensional systems the magnetic response appears like a paramagnetic system above the blocking temperature while the shape of the magnetization versus field curve is unlike a paramagnet [45]). Despite the paramagnetic like behavior, the inherent presence of short-range magnetic correlations is responsible for the observed Curie like behavior. Therefore, figure 3(j) and in section I of supplementary information, suggests that it is not possible to attribute the Curie like behavior below $T_{mf}$ to a conventional paramagnetic response. We believe that the regions possessing short range magnetic correlations fluctuate due to thermal fluctuations at finite $T$ resulting a Curie like $1/T$ dependence in $M(T)$ above $T_c$, as seen in figures 2(d)–(f). Such a feature of a system with short range magnetic order exhibiting paramagnetic like features at a finite temperature is not uncommon. In the context of nanomagnetic systems one observes the superparamagnetic effect where above a temperature called the blocking temperature, the magnetic orientation in these systems is not stable due to thermal fluctuations being large compared to the magnetic anisotropy energy. In such low dimensional systems the magnetic response appears like a paramagnetic system above the blocking temperature while the shape of the magnetization versus field curve is unlike a paramagnet [45]).
range magnetic correlation in the system leads to the observation of local hysteresis at lower \( T \) (see supplementary information section 1) and the nonlinear \( M(H) \) dependence as seen in figure 3(j). Therefore \( T_{\text{onset}}^{i}(H) \) identifies the onset of magnetization response due to stabilization of short range magnetic correlations in the samples. Figure 3(a)–(c) show the \( T_{\text{onset}}^{i}(H) \) line for underdoped, optimally doped and overdoped samples respectively. In figures 2(d)–(f), beyond \( T_{\text{onset}}^{i} \) we observe a large deviation of \( M \) from the linear Curie behavior. We propose this large deviation in \( M \) below \( T_{c}^{i} \) is due to the onset of a weak diamagnetic response above \( T_{c} \) which suppresses the paramagnetic like response in this temperature regime. In subsequent sections 3.7 and 3.8, we will present further evidence (via local magnetization and bulk resistivity measurements) for the presence of diamagnetic fluctuations above \( T_{c} \) in the optimally doped sample.

### 3.6. Local positive magnetization response not associated with impurity effects

The EPMA data \( (\text{XEPMA}) \) in Table 1 shows the average Co concentration sensitively measured across 12–13 regions (each region of size \( 10 \, \mu m \times 10 \, \mu m \)) distributed across the sample. The small error in \( \text{XEPMA} \) values shows that across the samples the fluctuations in, Co concentration and the overall sample stoichiometry (data not shown) are minimal. Furthermore, to rule out the possibility that the origin of positive magnetization response in the sample is predominantly associated with ferromagnetic inclusions in the sample (due to impurity effects), we measured the local iso-field magnetization response in ZFC state at various \( T \)'s at 200 Oe in optimally doped sample. Figure 4(a) shows the \( \langle B_{r} \rangle - H \) versus \( T \), where the average value \( \langle \cdot \rangle \) is obtained by spatially averaging \( B_{r}(x, y) - H \) value over the dot shaped (red) location (an area of \( 25 \, \mu m^{2} \) marked in the MO image in figure 4(a) inset). Figure 4(a) shows a peak like feature in \( \langle B_{r}(T) - H \rangle \) with a long decreasing magnetization tail (identical to the corresponding bulk \( 4\pi M(T) \) data at 200 Oe, see figure 2(b)) above \( T_{c} \) which turns strongly diamagnetic for \( T < T_{c} \). We argue that, if the positive magnetization feature observed above \( T_{c} \) at the dot shaped region were to be associated with local ferromagnetic impurities then the superconducting response in this region should have been significantly suppressed, as ferromagnetism is known to strongly suppress the superconducting order parameter [46]. However this dot shaped region shows the onset of a significant diamagnetic response below \( T_{c}(H) \), which is not expected if this region had ferromagnetic impurities. We also observe that the bulk and local (at the dot location) \( T_{c}'s \) are similar, further suggesting uniformity of \( T_{c} \) across the sample. These suggest that local ferromagnetic impurities do not play any significant role in determining the magnetization response of our samples. In fact the following two observations suggest that the positive magnetization response coexists along with superconductivity uniformly across the sample rather than suppress it (we shall also present more evidences for this in the next section); (1) above \( T_{c} \) the positive magnetization response is present uniformly over the entire sample rather than in any specific locations of the sample (see figures 3(g)–(i)), (2) below \( T_{c} \), the same regions which exhibit positive magnetization response also show a robust bulk superconducting magnetization response (see figures 1(a)–(d)). We suggest that the results in the paper cannot be attributed to effects of local magnetic impurities present in the samples. Further evidence pertaining to this issue is presented in section II of the supplementary information.

### 3.7. Imaging the spatially inhomogeneous state with diamagnetic and paramagnetic response above \( T_{c} \) in \( x = 0.14 \) sample using DMO

Figure 4(b) shows the distribution of \( \delta B_{z}/B_{r}(H) \) values, namely, \( P(\delta B_{z}/B_{r}) \) (measured across a dashed rectangular region on the sample shown in figure 4(a) inset) at 11, 35 and 70 K with \( H = 50 \, \text{Oe} \) for \( x = 0.14 \) sample. The distribution shows that at 11 K (\( T < T_{c} \)) the mean value of \( \delta B_{z}/B_{r}(H) \sim 0 \). The strong diamagnetism of superconducting regions shield the small external field modulation of \( \delta H = 1 \, \text{Oe} \), therefore \( B_{z}(H + \delta H) \sim B_{z}(H) \) and hence mean \( \delta B_{z} \sim 0 \) G in the superconducting regions. Thus the mean value of \( \delta B_{z}/B_{r}(H) \sim 0 \) is taken as the response corresponding to superconducting regions in the sample. At \( T = 35 \, \text{K} (T > T_{c}) \), the mean \( \delta B_{z}/B_{r}(H) \) value becomes positive (see arrow locations in figure 4(b)) and the distributions get narrower. At 70 K the mean value of \( \delta B_{z}/B_{r}(H) \) shifts further, becoming more positive compared to that at 35 K. We expect that regions which are magnetic are expected to respond to the modulation of the external magnetic field of \( \delta H = 1 \, \text{Oe} \) thus the \( B_{z}(H + \delta H) > B_{z}(H) \) and hence the mean \( \delta B_{z} \) values would get predominantly positive in the magnetic regions. Thus, below \( T_{c} \) at 11 K the \( P(\delta B_{z}/B_{r}) \) distribution suggests a predominant superconducting phase while well above \( T_{c} \) there is a magnetic phase.

We use the \( \delta B_{z}/B_{r}(H) \) values to color the DMO images at 11, 35 and 70 K (\( H = 50 \, \text{Oe} \)). The DMO image in figure 4(c) (11 K and 50 Oe) shows bright and dark stripes where the contrast variations represent changes in \( B_{z} \) (\( \delta B_{z} \)) produced in the sample in response to modulation in the external \( H \) by \( \delta H = 1 \, \text{Oe} \). From the calibrated DMO images, we calculate the \( \delta B_{z}/B_{r}(H) \) values at each pixel in the image and these values are used to color the original gray images (see figures 4(d)–(f)). The correspondence between \( \delta B_{z}/B_{r}(H) \) values (in percentage terms) and the color palette is represented beside each image in figures 4(d)–(f). In figure 4(d) at 11 K, the maximum negative value of \( \delta B_{z}/B_{r}(H) \sim -2.60\% \) has the darkest blue shade and the green shade represents maximum
positive value of $\delta B_z/B_z(H) \sim 2.50\%$. A common feature in all the three colored images in figures 4(d)–(f), is that the location of the blue and green shades of color is not randomly distributed over the sample. It appears that the green regions, where $\delta B_z/B_z(H)$ is significantly positive, are bunched up over stripe-like regions (see figure 4(d)) which are in close proximity to superconducting regions (blue regions). The positive $\delta B_z/B_z(H)$ values (green-yellowish shade) in figure 4(d) are regions with short range magnetic order (regions with magnetic fluctuations). Figure 4(d) thus displays the inhomogeneous nature of the superconducting state at $T < T_c$ where the blue shaded (superconducting) regions are interspersed with greenish (magnetic) regions. As $T$ is increased, the color contrast in the image changes from one which mainly has a bluish shade (superconducting) at 11 K (figure 4(d)) to one which predominantly has greenish shade (magnetic) (figure 4(e)) above $T_c$ at 35 K. From figures 4(e) and (f) it is interesting to observe that above $T_c$ although most of the regions are positively magnetized (green-yellowish contrast), they co-exist along with regions with weak superconducting diamagnetic response (dark blue contrast regions with negative $\delta B_z/B_z(H)$). With increasing $T$, figures 4(d)–(f) elucidates that by 70 K (close to $T^{\text{onset}}_c$ in figure 2(e)) the density of the bluish (superconducting) regions have significantly reduced although a small fraction still survives. It may be worthwhile mentioning that similar to the stripe-like features we observe in figure 4, an earlier study [47] had found that below $T_c$ stripe-like regions with enhanced diamagnetic shielding response are located over twin boundaries in underdoped BaFe$_2$As$_2$ samples. It is possible that the stripe-like regions with local diamagnetism we observe above $T_c$ are located around extended defects in these samples. In our optimally doped sample, the regions with weak diamagnetic (negative $\delta B_z/B_z(H)$) response seen in figures 4(e) and (f) at 35 and 70 K suggest the presence of an spatially inhomogeneous magnetic state between $T_c$ and $T^{\text{onset}}_c$ (see figure 2(e)).
3.8. Behavior of normal state conductivity, evidence of weak diamagnetism in the normal state

Figure 5(a) shows resistance (R) versus T/Tc(0) plot of x = 0.14 sample at 0 and 12 T measured up to T/Tc(0) ~ 4. Using this R(T) data, we plot the conductivity (σ) difference between the 0 T and 12 T data, σ0T−12T versus T/Tc(0). The shaded region (above Tc(0)) in figure 5(b) uncovers the excess conductivity (σex > σn) at 0 T. The origin of the excess conductivity above Tc at 0 T is due to the presence of superconducting fluctuations which get suppressed by a large field of 12 T. In figure 5(b) one observes that the excess conductivity regime exists up to T/Tc(0) ~ 2.5, which is close to Tc(0) ~ 60 K for x = 0.14 sample. We analyze the 0 T conductivity data within the Azlamov–Larkin (AL) [48] theory, which proposes the presence of fluctuating Cooper pairs (namely, superconducting fluctuations) above Tc in zero magnetic field. The existence of superconducting fluctuations causes the conductivity to be higher than that in the normal state of a superconductor without any superconducting fluctuation. In three dimensions, the temperature dependence above Tc of the zero magnetic field excess conductivity also called Paraconductivity is given by the AL expression [49]

\[ \Delta \sigma_{\text{AL}}^{(3D)} = \frac{e^2}{32\hbar\xi(0)} \varepsilon^{-1/2}, \]  

(2)

where \( \varepsilon = \ln(T/Tc(0)) \) and ξ(0) is the coherence length of the superconductor. (To calculate \( \Delta \sigma \) : the \( \sigma_{0T}(T) \) data above 95 K is fitted (not shown here). This fit represents the normal state temperature dependence of \( \sigma_{0T} \) in the absence of superconducting fluctuations. Using the fitting parameters we calculate the expected normal state conductivity (in the absence of any fluctuations) over the entire temperature range of our measurement, i.e., \( \sigma_n(T) \). The excess conductivity due to the presence of diamagnetic fluctuations then is \( \Delta \sigma = \sigma_{0T}(T) - \sigma_n(T) \). In figure 5(c) we plot \( \Delta \sigma \) versus \( \varepsilon \) along with the \( \Delta \sigma_{\text{AL}}^{(3D)}(\varepsilon) \) behavior given by equation (2) (red dashed line) using ξ(0) ~ 2.5 nm [30]. Figure 5(c) shows that \( \Delta \sigma \) obeys equation (2) for small values of \( \varepsilon \) suggesting the presence of diamagnetic fluctuations above Tc. However for 0.1 < \( \varepsilon \) < 1, the \( \Delta \sigma \) becoming larger than the theoretically estimated value, \( \Delta \sigma_{\text{AL}}^{(3D)} \), indicating a weaker temperature dependence of \( \Delta \sigma \) in x = 0.14 sample compared to equation (2). This suggests the presence of significant diamagnetic fluctuations sustained in the sample up to T well above Tc, and may also indicate the pairing interactions could be strong leading to weak T dependence above Tc. In figure 5(c) we see that above the arrow locating Tc(0), the \( \Delta \sigma(\varepsilon) \) drops rapidly as diamagnetic response disappears. Thus the magnetic response above Tc in the x = 0.14 sample has an admixture of magnetic response associated with magnetic fluctuations and diamagnetic response (related to superconducting fluctuation). The above observation concurs well with the observation of the presence of stripes with diamagnetic shielding response present above Tc at 35 and 70 K in figures 4(e) and (f).

3.9. Comparison of local magnetization in x = 0.14 and x = 0.20 samples

In figure 6, we compare the local Bz distribution in x = 0.14 and x = 0.20 samples measured at T = 11 K (< Tc) and H = 0 Oe. In figure 6(a) the x = 0.14 sample exhibits a larger positive Bz response than x = 0.20 sample. Although small and much weaker than x = 0.14 sample, the Bz distribution deep inside the overdoped (x = 0.20) sample is not zero but finite with Bz ~ 2 G (see inset figure 6(a)). This is consistent with the observation in figures 3(a)–(c) that the positive M features in bulk M(T) are also present in x = 0.20 sample although they are much weaker as compared to x = 0.14 sample. Figure 6(b) shows the Bz(t) measured across a line shown in inset of figure 6(b) for x = 0.20 sample at 11 K and different H. The Bz(t) profiles for x = 0.20
We now estimate the superconducting volume fraction of diamagnetic signal enhances the yellow shaded region, the response from magnetic images such as those in figures 2(a)–(f). A schematic representation of δM is shown in figure 2(d). The denominator in equation (3) represents the saturated diamagnetic signal of the Meissner fraction of the superconductor at H = 50 Oe and T < Tc (see figures 1(a)–(c)). Using equation (3), the estimated v_d to be 0.094% for x = 0.10 (underdoped), 0.346% for x = 0.14 (optimally doped), and 0.008% for x = 0.20 (overdoped) sample. Hence, the largest superconducting volume fraction above Tc is found for optimally doped sample, which also has significantly high value of 4πM^0_M, i.e., the response due to magnetic fluctuations (in fact the optimally doped sample has the highest saturated value of 4πM^0_M as shown in figure 3 (supplementary information). We believe the comparatively larger v_d in x = 0.14 (optimally doped) sample helps in establishing long range superconducting phase coherence in the small temperature window between Tc and Tp. We would like to mention here that MO images such as those in figures 4(c)–(f) represent the response measured on the surface of the sample and therefore such images are not suitable for estimating v_d.

3.11. Identifying regions with admixed magnetic and superconducting fractions in H–T phase space

Figures 7(a)–(c) show the H–T phase diagram constructed using the characteristic temperatures (section 3.5) identified from features in bulk magnetization measurements (see figures 2(d)–(f)) for the three samples. The bluish region below T^scf_c in figures 7(a)–(c) represents the H–T regime where only magnetic fluctuation response is found in the samples and the positive magnetization response across this regime increases with decrease in T. The yellow shaded region represents the H–T regime where the magnetic state becomes inhomogeneous due to the appearance of regions with diamagnetic shielding response at T^scf_c (above Tc) coexisting alongside regions with short range magnetic order (magnetic fluctuations). In between Tp and T^scf_c the appearance of regions with superconducting fluctuations causes the magnetization behavior to bend away from the Curie like behavior (see figures 2(d)–(f)). The paraconductivity behavior in figure 5(c) also shows the appearance of significant superconducting fluctuations below T^scf_c. Similarly in figures 4(e) and (f), an inhomogeneous magnetic state with diamagnetic fluctuations appears below 70 K which is near T^scf_c. Within the yellow shaded region, the response from magnetic fluctuations dominates over the diamagnetic response (for example, refer to figures 4(e) and (f) for sample x = 0.14 and the magnetization hysteresis loops above Tc in figure 3(j) and section I of supplementary information). As we decrease the temperature towards Tc, the strength of diamagnetic signal enhances (due to growing superconducting correlations in the sample), resulting the onset of significant deviation of the net M from the Curie like behavior below T^scf_c (see figures 2(d)–(f)). The peak like
feature in the $M(T)$ curves of figures 2(a)–(c) occurs because with reducing $T$ the diamagnetic response enhances at a rate faster than the increase in magnetization due to the presence of regions with magnetic fluctuations. In figures 7(a)–(c) across the orange shaded region (between $T_p$ and $T_c$), the diamagnetic response dominates over magnetic fluctuations. Here the $M$ changes sharply from positive values at $T_p$ towards negative values (diamagnetic) below $T_c$. Finally below $T_c(H)$ bulk superconductivity sets in (see red shaded region in figures 7(a)–(c)) with weak magnetic fluctuations surviving in the superconducting state (see figure 4(d)). In this regime at low $H (\leq 40 \text{ Oe})$ we observe the positive $B_p-H$ (see figure 3(e)) due to magnetic fluctuations surviving along with a diamagnetic bulk $M(T)$ response (figure 1) below $T_c$. From figures 7(a)–(c) one observes that the orange region (between $T_p$ and $T_c$ lines) is the narrowest for the optimally doped sample, i.e., the transition (from positive $M_{SC}(H)$) into the bulk superconducting state (diamagnetic $M$) is the sharpest for $x = 0.14$ sample. The different lines in figures 7(a)–(c) are weakly field dependent in the moderate field range. Note that the $T_c$ and $T_p$ lines are far more sensitive to $x$ variation compared to $T_{CFL}$ which shifts a little only for $x = 0.20$ and $T_{CFL}$ is almost independent of $x$.

3.12. Discussion

The close relationship between magnetic fluctuations and superconductivity can be inferred from the following observation: as magnetic fluctuation effects get stronger as in $x = 0.14$ sample (see section II of supplementary information), simultaneously the superconducting volume fraction above $T_c (v_{SC} = 0.346\%)$ and the superconducting transition temperature also increases. With weakening of magnetic fluctuations as in $x = 0.20$ sample (see section II of supplementary information), $v_{SC} (=0.008\%)$ and $T_c$ decreases. To explain these features, we speculate that magnetic fluctuations perhaps play an important role in mediating superconducting pairing above $T_c$ in this compound. As seen from Neutron diffraction and muSR measurements [9–11], Co doping destroys long range SDW magnetic order in the parent compound however the doping generates local short range magnetic order between Fe moments present uniformly throughout the sample. Above $T_{CFL}$, the presence of this short range magnetic order (magnetic fluctuations) perhaps mediates superconducting pairing [21–28] resulting in our observation of diamagnetic response sustained up to temperatures well above $T_{CFL}$ (see figures 2(d)–(f), 4(d)–(f), 5(b), (c)). Recent experimental studies have found evidence for the presence of magnetic fluctuations along with superconductivity below $T_c$ [51, 52]. Studies in FeSe samples have shown evidence for BCS-BEC crossover in a Fermi system with strong spin imbalance, where the presence of preformed pairs in the crossover regime [33] was reported, namely, the possibility of superconducting fluctuations present above $T_c$. The work suggested the possibility that in such systems with spin imbalance, the superconducting state may be exotic with spin triplet pairing. While we do observe the presence of diamagnetic fluctuations above $T_c$ coexisting along with magnetic fluctuations, however in our present measurements we do not find evidence of exotic spin triplet pairing in BaFe$_{2-x}$Co$_x$As$_2$. We believe that in our BaFe$_{2-x}$Co$_x$As$_2$ samples below $T_c$, the extent of free energy reduction due to onset of superconductivity is higher than an increase in energy due to Zeeman contributions from any magnetized state surviving below $T_c$ (in the presence of $H$). Therefore, energetically the lower energy superconducting state gets favoured in the presence of a field $H$ as compared to the magnetic state (see figure 3(f)) and we find diamagnetic signal in the sample enhances with increasing $H$ below $T_c$. In BaFe$_{2-x}$Co$_x$As$_2$ compounds, as the same set of electrons need to choose between participating in pairing or contributing to magnetism near $T_c$, perhaps the above energy considerations favor a magnetic fluctuation mediated pairing [13, 53, 54], leading to the superconducting state being preferred over a magnetic state. While
these considerations could partially explain the observation in figure 3(f) of the diamagnetic response strengthening as $H$ is increased within a moderate field range, however we do not fully understand the origin of this phenomenon. Below $T_c$ the magnetic phase may remain as a microscopic phase coexisting along with the superconducting phase.

4. Summary and conclusions

We have presented evidence for the presence of a two component magnetic response in single crystals of BaFe$_2$-xCo$_x$As$_2$ with different Co-doping concentration. While one component of the response is diamagnetic, the other component is magnetic with the relative strengths of these two signals varying depending on whether the temperature is below $T_c$, or above it. Presence of excess conductivity above $T_c$ in the optimally doped sample is related to diamagnetic fluctuations above $T_c$. The normal magnetized state in this system is spatially inhomogeneous. The local susceptibility response suggests the inhomogeneous regions is related to regions with superconducting fluctuations coexisting with regions having short range magnetic order. We find the optimally doped sample with highest $T_c$ exhibits the strongest effect of magnetic fluctuations and it also possesses the highest volume fraction with diamagnetic response above $T_c$. Below $T_c$ along with bulk superconductivity, weak magnetic fluctuations are sustained although the response from regions with magnetic fluctuations get suppressed with the application of a magnetic field. The study suggests that in BaFe$_2$-xCo$_x$As$_2$ systems magnetic fluctuations perhaps play an important role in generating local superconducting pairing correlations above $T_c$.

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