Incommensurate Spin-Density Wave Order in Electron-Doped BaFe2As2 Superconductors

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Abstract
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Neutron diffraction studies of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ reveal that commensurate antiferromagnetic order gives way to incommensurate magnetic order for Co compositions between 0.056 < x < 0.06. The incommensurability has the form of a small transverse splitting (0, ±ε, 0) from the commensurate antiferromagnetic propagation vector $Q_{\text{AFM}} = (1, 0, 1)$ (in orthorhombic notation) where $ε ≈ 0.02–0.03$ and is composition dependent. The results are consistent with the formation of a spin-density wave driven by Fermi surface nesting of electron and hole pockets and confirm the itinerant nature of magnetism in the iron arsenide superconductors.

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Unconventional superconductivity is often associated with the pairing of electrons via spin fluctuations that appear close to a magnetic ordering instability. In this respect, the nature and origin of the magnetic instability itself is an important ingredient of any theory of superconductivity. In the iron arsenide compounds, the magnetism has been discussed from two limits; an itinerant and a local moment picture that may become relevant in the parent Fe$_2$As$_2$ system. Detailed magnetic diffraction studies have observed only C-AFM order with a propagation vector $Q_{\text{AFM}}$ in several Fe$_2$As$_2$ systems including the parent compounds [1–3] and doped compounds: Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [4–6], Ba(Fe$_{1-x}$Ni$_x$)$_2$As$_2$ [7], Ba(Fe$_{1-x}$Rh$_x$)$_2$As$_2$ [8], Ba(Fe$_{1-x}$Ru$_x$)$_2$As$_2$ [9], Ba$_{1-x}$K$_x$Fe$_2$As$_2$ [19]. Incommensurability has been claimed in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ by local probes such as $^{75}$As nuclear magnetic resonance [20], $^{57}$Fe Mössbauer [21], and muon spin resonance [22] measurements. However, detailed measurements of the AFM ordering by both neutron and high resolution x-ray resonant magnetic diffraction have found no incommensurability in Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ up to x = 0.047 [6].

In this Letter, neutron diffraction data demonstrate that IC magnetic order does indeed develop near optimally doped compositions of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ with x ≈ 0.056, just before long-range magnetic ordering is completely suppressed at x = 0.06. The IC propagation vector $τ = Q_{\text{AFM}} + (0, ε, 0)$ corresponds to a transverse splitting ($ε ≈ 0.02–0.03$) whose value depends on composition. The direction and magnitude of the observed IC splitting is consistent with calculations of the generalized susceptibility determined by density functional theory, allowing us to conclude that static magnetism and the spin fluctuations for superconducting compositions are tied to an itinerant SDW instability.

Single crystals of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ were prepared using a solution growth technique described elsewhere [23] with the following compositions (and masses); x = 0.054 (124 mg), 0.056 (248 mg), 0.057 (73 mg), 0.059 (136 mg), and 0.062 (106 mg). The sample compositions were determined through a series of characterization measurements including resistivity, magnetization, and wavelength dispersive spectroscopy (WDS) [23]. All samples are orthorhombic below the tetragonal-orthorhombic transition temperature ($T_N$) and the data are discussed in terms of the orthorhombic indexing $Q = (2πd/a, 2πd/b, 2πc/c)$ where $a = b = 5.6$ Å and $c = 13$ Å. The characteristic phase

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transition temperatures $T_S$, $T_N$ (antiferromagnetic ordering temperature), and $T_c$ (superconducting transition temperature) were determined using similar procedures outlined previously [5,24]. $T_N$ was determined by the break in slope of the integrated neutron intensity (order parameter) at $Q_{AFM}$ separating Bragg scattering from a "tail" of critical scattering or magnetic short-range order that can persist above $T_N$. This method can lead to a small uncertainty in the true value of $T_N$ for the weak magnetic signals encountered in this study. However, all reported transition temperatures ($T_S$, $T_N$, and $T_c$) are consistent with our current understanding of the phase diagram of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [25]. The sharpness of the superconduring transition, predictable evolution of $T_S$ and $T_N$ with relatively small changes in composition, and uniformity of the WDS signal at multiple locations on the crystals confirm good chemical homogeneity with compositional spread $\Delta x < 5\%$ [23].

Neutron diffraction measurements were performed at Oak Ridge National Laboratory using the High Flux Isotope Reactor’s HB1A triple axis spectrometer using a horizontal collimation $48' - 48' - 40' - 68'$ and $E_f = 14.7$ meV. All samples have resolution-limited mosaic full-widths of $<0.4$ degrees and were mounted in a closed-cycle refrigerator. Samples were studied in the vicinity of $Q_{AFM} = (1, 0, 1)$ and $(1, 0, 3)$ in two scattering planes; in the $(H, 0, L)$ plane, allowing the search for IC splitting along the orthorhombic $a$ axis ($[H, 0, 0]$ is referred to as the longitudinal direction), and in the $(\zeta, K, 3\zeta)$ or $(\zeta, K, \zeta)$ planes, allowing the search for incommensurability along the $b$ axis ($[0, K, 0]$, transverse direction). [See Fig. 1(a)]

Typical transverse $[0, K, 0]$ and longitudinal $[H, 0, 0]$ neutron diffraction scans are shown in Figs. 1(b) and 1(c) for the $x = 0.059$ sample at $T \approx T_c < T_N$, where magnetic Bragg intensity is a maximum. The observation of a pair of Bragg peaks located symmetrically at positions $(0, \pm \epsilon, 0)$ around $Q_{AFM}$ in the transverse scan clearly indicates IC magnetic order for this composition. No longitudinal splitting is observed, therefore IC magnetic order is present with propagation vector $\tau = (1, \epsilon, 1) = Q_{AFM} + (0, \epsilon, 0)$, as illustrated in Fig. 1(a).

We now turn to ab initio density functional calculations of the magnetic susceptibility to show that the observed IC-AFM order can be understood as a SDW driven by Fermi surface nesting. Previous calculations show maxima in the generalized spin susceptibility away from $Q_{AFM}$ in doped $AFe_2As_2$ compounds and therefore point to a tendency for IC-SDW order [17,18]. To gain insight into potential incommensurability at doping levels where we observe static IC-AFM order, we performed calculations of the generalized bare susceptibility employing the full-potential linearized augmented plane wave (FPLAPW) method [26], with a local density functional [27]. We used $R_{MT}K_{\text{max}} = 8.0$ and $R_{MT} = 2.4, 2.2$ and 2.2 for Ba, Fe, and As, respectively. To obtain self-consistency

![FIG. 1 (color online).](image)

(a) Reciprocal space plane with $L = \text{odd}$ indicating commensurate (C, empty circles) and incommensurate (IC, filled circles) magnetic Bragg peak positions at $Q_{AFM} = (1, 0, L = \text{odd})$ and $\tau = (1, \pm \epsilon, L = \text{odd})$, respectively, in orthorhombic notation. The size of the incommensurability parameter is exaggerated for clarity. Shaded points labeled $C^*$ and $IC^*$ show the location of magnetic Bragg peaks that are present due to orthorhombic twinning. Dashed arrows illustrate the direction of longitudinal $[H, 0, 0]$ and transverse $[0, K, 0]$ neutron diffraction scans. Raw (b) transverse and (c) longitudinal scans for Ba(Fe$_{0.94}$Co$_{0.05}$)$_2$As$_2$ at $T = 23 \text{K} \approx T_c$. The lines are Gaussian fits to the data. Ab initio calculations of the generalized susceptibility in the (d) transverse and (e) longitudinal directions through $Q_{AFM}$. we chose 550 $k$-points in the irreducible Brillouin zone and used 0.01 mRy/cell as the total energy convergence criteria. The virtual crystal approximation was used to consider Co-doping effects and the whole reciprocal unit cell is divided into $80 \times 80 \times 80$ parallelipipeds, corresponding to 34061 irreducible $k$ points. Our calculations of the generalized susceptibility for electron doping with $x = 0.05$ show splitting in the transverse direction and a single peak in the longitudinal direction in Figs. 1(d) and 1(e), respectively, consistent with other doping dependen
calculations \[17,18\]. The \textit{ab initio} calculations, therefore, show a tendency for IC-SDW order with propagation vector \( \tau = Q_{\text{AFM}} + (0, \epsilon, 0) \) in agreement with experimental observations.

Figure 2 shows the transverse \([0, K, 0]\) scans through \((1, 0, 3)\) for other compositions and temperatures. The scans performed at temperatures above \(T_S\) serve as an estimate of the background. The propagation vector, peak widths, and integrated intensities were determined by Gaussian fits to the scans shown in Fig. 2. The transverse \([0, K, 0]\) scans show only a single resolution-limited peak for the \(x = 0.054\) sample and, combined with x-ray resonant magnetic diffraction results from M. G. Kim et al. \[6\], establish stripe-type C-AFM order at \(Q_{\text{AFM}}\) for all Co compositions below approximately 0.054. Broad peaks split in the transverse direction are observed in \([0, K, 0]\) scans for \(x = 0.056, 0.057,\) and 0.059, clearly establishing the transition to an IC magnetic phase with propagation vector \(\tau\). For the \(x = 0.056\) sample, both C and IC peaks are observed, suggesting that the transition is first-order in its dependence on Co concentration with the phase boundary close to \(x = 0.056\). The coexistence of C and IC phases could arise from a small spread in the Co concentration across the sample, as noted above. Figure 2(b) shows that the line shapes at \((1, 0, 3)\) and \((1, 0, 1)\) positions are equivalent with an integrated intensity ratio of 0.36(9), close to that expected for collinear C-AFM order with the magnetic moment pointing along the \(a\) axis. Therefore, the IC-AFM structure is most likely also collinear, and not helical or cycloidal. No signatures of higher harmonics have been observed, indicating a sinusoidal modulation of the moment size along the \(b\) direction.

We now discuss the temperature dependence of transverse scans shown in Fig. 2. For \(x = 0.054\) [Fig. 2(a)], the suppression of the integrated intensity (magnetic order parameter) below \(T_c\) indicates the competition of C-AFM with superconductivity, as reported previously \[5,24,28\]. The magnetic intensity in Figs. 2(b)–2(d) has allowed us to outline regions of C and IC magnetic order in the phase diagram. Figure 3(a) shows the experimental phase diagram of \(\text{Ba(Fe}_{1-x}\text{Co})_2\text{As}_2\) delineating regions of magnetic order, superconductivity, and structural phases as based on previous studies \[9–11,23–25\]. This work, summarized in Figs. 3(b)–3(d), has allowed us to outline regions of C and IC magnetic order in the phase diagram. Figure 3(b) shows the evolution from C (at \(x = 0.054\)) to IC-AFM order (from \(x = 0.056-0.059\)) in transverse scans performed at \(T > T_c\). The \(x = 0.062\) sample has no
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