Neutron scattering study of underdoped Ba_{1−x}K_{x}Fe_{2}As_{2} (x=0.09 and 0.17) self-flux-grown single crystals and the universality of the tricritical point

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A neutron scattering study of the under-doped $Ba_{1-x}K_xFe_2As_2$, x=0.09 and 0.17 self-flux grown single crystals and the universality of the tricritical point

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We present a combination of elastic neutron scattering measurements in zero and 14.5 T and magnetization measurements in zero and 14 T on under-doped superconducting $Ba_{1-x}K_xFe_2As_2$ x=0.17, and the same measurements in zero field on a non-superconducting crystal with x=0.09. The data suggest that the under-doped materials may not be electronic phase separated but rather have slightly inhomogeneous potassium doping. The temperature dependence of the magnetic order parameter (OP) below the transition of the sample with x=0.09 is more gradual than that for the case of the un-doped $BaFe_2As_2$, suggesting that this doping may be in the vicinity of a tricritical point. We advance therefore the hypothesis that the tricritical point is a common feature of all superconducting 122s. For the x=0.17 sample, while T_c is suppressed from ≈17 K to ≈8 K by a magnetic field of 14 T, the intensity of the magnetic Bragg peaks (1 0 3) at 1.2K is enhanced by 10% showing competition of superconductivity (SC) and antiferromagnetism (AFM). The intensity of the magnetic Bragg peaks (1 0 3) in the (T_c, T_N) temperature interval remain practically unchanged in 14.5 T within a 10% statistical error. The present results are discussed in the context of the existing literature.

I. INTRODUCTION

High temperature superconductivity (HTSc) in the iron pnictides, with a T_c as high as 55 K for the case of $SmFeAsO_{1-\delta}$ and $SmFeAsF_xO_{1-x}$[1,2], is one of the most perplexing discoveries of the decade in the field of condensed matter physics. The 122 series (AFe_2As_2, A = Ba, Sr, Ca, Eu) is of great interest since it is an oxygen-free HTSc. Superconductivity in the 122s can be induced by doping in any of the three atomic sites[3–12]. The hole-doping achievable through chemical substitution with either K[3], Na[4] or Cs[5] in the atomic site A can give a T_c as high as 39 K in the case of $Ba_{0.55}K_{0.45}Fe_2As_2$[3]. The antiferromagnetic (spin-density wave) and structural (tetragonal to orthorhombic) transitions that are near-coincident in the parent compounds[13,14] are concomitantly and gradually suppressed upon doping. Although in the electron-doped $BaFe_2As_2$ the two transitions separate with doping[15], it seems that there are examples pointing otherwise, as in the case of the isovalent ruthenium doped $BaFe_2As_2$[16,17] and the case of electron-doped $SrFe_{2(1-x)}Co_{2x}As_2$[18]. The last is surprising if we consider the result on Sn-flux grown $CaFe_21-xCo_{2x}As_2$ crystals[19] for which the two transitions are clearly separated. In the case of potassium (hole) doped $BaFe_2As_2$[20] the question of concomitant or separated transitions remains controversial. While powder neutron diffraction data argue for concomitant magnetic and structural transitions across the whole series[20,21], heat capacity on Sn-flux grown $Ba_{0.84}K_{0.16}Fe_2As_2$ single crystals shows two

distinctive peaks attributed by the authors to the magnetic and structural phase transitions, respectively[22]. In most cases the source of contradictory results appears to be connected to issues of sample quality. It has been pointed out that the proper flux to grow the 122s is FeAs[23–25], as other fluxes contaminate the sample with flux element inclusions, with a consequent impact on the physical properties. Neutron diffraction on powder $BaFe_2As_2$[14] determined a first-order structural and magnetic transition. Complementary high resolution X-ray diffraction and heat capacity measurements on high quality $BaFe_2As_2$ crystals revealed a 1st order magnetic transition preceded by a structural transition that starts as a 2nd order transition at a slightly higher temperature but with a first order jump in the orthorhombic distortion co-incident with the first order magnetic transition[26,27]. For the electron-doped $BaFe_{2(1-x)}Co_{2x}As_2$ it has been shown recently that the magnetic transition order changes upon doping from 1st to 2nd through a tricritical point[27,28], which is believed to be relevant to the superconductivity phenomenon itself[29]. For this series the structural transition is 2nd order. This seems to be different for the case of polycrystalline hole-doped $Ba_{1-x}K_xFe_2As_2$ of C. Avci *et al.*[20] for which both the magnetic and structural transitions are 1st order over the entire doping range. An early report on Sn-flux grown K-doped $BaFe_2As_2$ revealed an electronic phase separated material[30]. A more recent atom probe tomography study on self-flux grown underdoped $Ba_{0.72}K_{0.28}Fe_2As_2$ provides evidence for a mixed scenario of phase coexistence and phase sep-

aration originating from variation of the dopant atom distributions[31]. In the present article we report complementary zero and 14.5 T elastic neutron scattering and zero and 14 T magnetization measurements on underdoped non-SC x=0.09 and SC x=0.17 ($T_c{\approx}17$ K). For the non-SC x=0.09 sample the AFM transition is sharp (width within 1 K), consistent with a weakly 1st order transition, and the temparature dependence of the magnetic order parameter (OP) squared is more gradual than that for the case of the parent $BaFe_2As_2$. This possibly indicates proximity to a tricritical point. For the higher doping sample x=0.17, the transition presents a distribution of T_Ns due to a slight variation of the potassium dopant, leading to a rounding of the transition of about 6 K. This rounding makes it difficult to differentiate between first and second order behavior of the transition. Our neutron data show that, although the SC under-doped x=0.17 sample has a SC volume fraction of ≈40%, the downturn in the AFM order parameter below T_c and its enhancement in magnetic field provide possible evidence for microscopic coexistence of AFM and SC, similar to the case of the electron-doped 122s[32].

II. EXPERIMENTAL PROCEDURE

Single crystals of $Ba_{1-x}K_xFe_2As_2$ with potassium dopings x of 0.09, 0.17, 0.41, and 0.45[33] were synthesized by a self-flux method with details given in an earlier report[34]. Samples with x=0.41 and 0.45 show no sign of AFM ordering, and are fully superconducting with T_c=38 K[34] and 39 K, ΔT_c=2 K, respectively. The potassium doping has been extrapolated by comparing the c lattice determined from room temperature neutron data with c vs. x (K doping) data on crystals of H. Lou $et\ al.$[35] (Fig. 1). This data agree with c vs. x of the polycrystalline samples[36]. Crystals of $SrFe_{2-x}Ni_xAs_2$, x=0.155 were grown by self-flux method as well[37] and the precise Ni doping value was determined by inductively coupled plasma analysis.
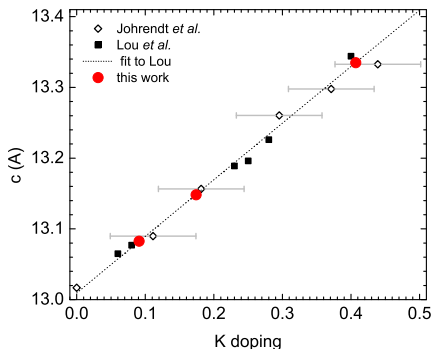

FIG. 1: c lattice versus x potassium doping, literature and present study data.

Magnetization measurements on the K-doped samples were carried out using a Magnetic Property Measurement System (MPMS) and a Physical Property Measurement System (PPMS) from Quantum Design®. Resistivity measurements on the Ni-doped $SrFe_2As_2$ were performed in the PPMS. Zero field neutron diffraction measurements were performed at Oak Ridge National Laboratory (ORNL) with the High Flux Isotope Reactor's HB-1A triple axis spectrometer, using a horizontal collimation 40' – 40' - $sample$ – 40' – 68' and fixed energy E_i=14.6 meV. The samples studied had K concentrations of x=0.09, 0.17, and 0.41 with masses of 19.5 mg, 45 mg, and 71.5 mg, respectively. In order to assess further sample quality, rocking cuvres of the (008) Bragg peak were recorded. For the x=0.09 sample the rocking curve showed two peaks that were separated by approximately 0.7°. Fitting these peaks to a Lorentizan squared profile gave FWHMs of 0.60° and 0.70°. For the x=0.17 sample, the rocking curve of the same peak gave one main peak with a FWHM of 0.77°.

Neutron diffraction measurements of the x=0.17 sample of 50 mg in zero field and 14.5 T were performed at the Helmholtz-Zentrum Berlin (HZB) with a configuration of 60' – 20' - $sample$ – 20' and fixed E_f=5.0 meV. The rocking curve of the (002) Bragg peak showed two peaks that were separated by 0.86°, both peaks had a FWHM of 0.47°.

For both ORNL and HZB neutron scattering experiments, the samples were mounted in a closed-cycle refrigerator and studied in the vicinity of the magnetic Bragg position Q_{AFM}=(1 0 3). For all magnetization and neutron scattering measurements, the magnetic field was parallel with the (a b) crystallographic plane.

III. RESULTS AND DISCUSSION

The thermal evolution of the integrated intensity of the (1 0 3) magnetic Bragg peak in the non-SC x=0.09 sample is shown in Fig. 2, upper panel. In addition, the scattering from the same magnetic Bragg peak of the undoped parent compound $BaFe_2As_2$ is also shown (data is taken from Ref. 38). The peak intensity scales like the magnetic OP squared. For the sample x=0.09 the Néel temperature is 136 K. The figure shows that the magnetic OP squared in the x=0.09 sample evolves in a much more gradual manner than Wilson $et\ al.$'s data on x=0[38]. Even so, there is a clear and sharp jump (within 1 K) of magnetic OP squared directly below the Néel temperature. This clearly shows that magnetic phase transition in this sample is still first order, albeit one that is weaker than the parent compound's. In critical phenomena language this corresponds to a slight increase in the effective critical exponent describing the temperature dependence of the OP below the first order transition. We speculate that x=0.09 may be close to a tricritical point, similar to the one found in Co-doped $BaFe_2As_2$[27,28]. It has been shown for the electron Co-doped $BaFe_2As_2$[28]
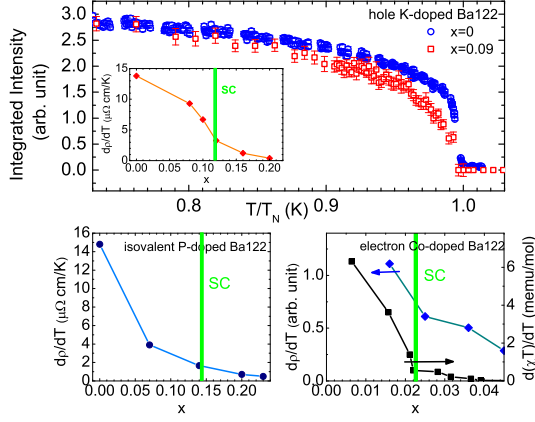
FIG. 2: The upper panel shows the integrated intensity of the (1 0 3) magnetic Bragg peak versus reduced temperature of the non-SC K-doped sample x=0.09 ($\square$) plotted against the same data for the un-doped x=0 ($\circ$) (from Ref. 38). The inset shows the magnitude of the dρ/dT peaks vs. x potassium doping of the series extracted from Ref. 39. Similar dρ/dT vs. x data is plotted for the isovalent phosporus-doped BaFe$_2$As$_2$ in the lower left panel, and for the electron cobalt-doped in the lower right panel. The thick vertical lines indicate the doping corresponding to emergence of sueprconductivity in the series.

that, around the tricritical point, the heat capacity C and d(χT)/dT vs. doping present a change from a more abrupt variation (characteristic of a 1st order transition), to a monotonic and much slower variation (characteristic of a 2nd order transition). For the electron Co-doped BaFe$_2$As$_2$ in the lower right panel of Fig. 2, the magnitude of the peaks d(χT)/dT vs. doping reproduced from Ref. 28 is drawn in comparison with the magnitude of dρ/dT[40] peaks as extracted from Ref. 40. In the inset of the upper panel of Fig. 2 we plot the magnitude of the dρ/dT peaks vs. x of the same series, extracted from Ref. 39. The existence of an inflection point in the dρ/dT vs. x data may indicate a tricritical point at around x $\approx$ 0.12 for the hole K-doped system. As found for the case of the Co(electron)-doped BaFe$_2$As$_2$[28], this tricritical point in K(hole)-doped BaFe$_2$As$_2$ is in the near proximity of emergence of superconductivity 0.125 $\leq$ x $\leq$ 0.133[20]. Finally, the lower left panel shows magnitude of dρ/dT peaks vs. phosphorus doping as extracted from Ref. 41[41].

Figure 3 shows the integrated intensity of the (1 0 3) magnetic Bragg peak versus temperature of the sample x=0.17. The downturn of the intensity below T$_c$ provides evidence for the microscopic coexistence of AFM and SC. In the inset is shown the sum of counts versus temperature near T$_c$. The downturn of the magnetic OP is less pronounced than for the case of homologous superconducting electron doped BaFe$_{2(1-x)}$Co$_{2x}$As$_2$[32] because of the low superconducting volume fraction. The "rounded" Néel transition (over a $\approx$ 6 K temperature range) is due
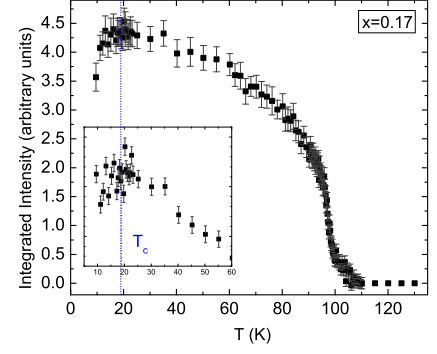

FIG. 3: Integrated intensity of the magnetic OP versus temperature for the sample x=0.17. The downturn of the OP below T$_c$ shows coexistence of AFM and SC. In the inset it is the sum of counts versus temperature near T$_c$. The vertical interrupted line is a guide to the eye for T$_c$. The rounded transition at T$_N$ is due to the slight K-doping inhomogeneity.

in part to a slight distribution in the potassium doping, and therefore this will give an averaged $\langle$T$_N\rangle$. If we assume that the effect is due solely to a spread in doping, then the $\approx$ 6 K wide transition corresponds on the phase diagram[36] to a variation on potassium doping x of about 2.5%. The presence in the sample of small fractions of material with slightly smaller values of potassium doping will result in a non-zero magnetic order parameter above $\langle$T$_N\rangle$ and SC critical temperatures below $\langle$T$_c\rangle$ (untraceable by means of resistivity and magnetization measurements).
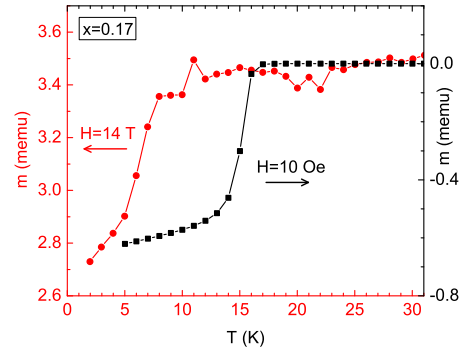

FIG. 4: Magnetization of the x=0.17 sample in H=10 Oe showing the onset of the diamagnetism at $\approx$17 K (right axis). In 14 T (left axis) T$_c$ decreases to $\approx$8 K.

In order to investigate the effects of magnetic field on superconductivity, we have measured the magnetization of an x=0.17 sample. The onset of the diamagnetism as measured in 10 Oe is at $\approx$17 K (Fig. 4, right axis). The superconducting volume fraction is about 40%. This
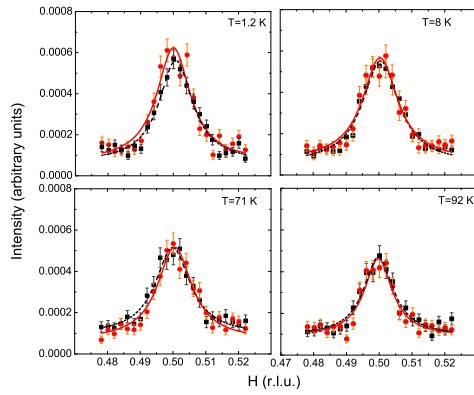
FIG. 5: Elastic neutron-scattering scans of the (1 0 3) magnetic Bragg peak at different temperatures: T=1.2, 8, 71, and 92 K of the sample x=0.17. For each temperature the zero field data are indicated with filled squares and data in 14.5 T with filled circles, and the curves are Lorentzian fittings (dashed line is for zero field).

FIG. 6: Resistivity vs. temperature of the $SrFe_{2-x}Ni_xAs_2$, x=0.155 normalized at its value at 320 K, in zero field and in 14 T$//c$. Before the measurement, the sample was annealed in low argon pressure for 24 hours at 700° C.

value is considerably higher than the 23 % reported for a higher potassium doped Sn-flux grown sample[30], and contrasts with the 98% value found by R. R. Urbano *et al.*[22] in their Sn-flux grown x=0.16. In a magnetic field of 14 T (Fig. 4, left axis) T_c decreases to ≈8 K. This is expected since under-doped superconducting samples have a lower critical field H_{c2} than those that are optimally doped, where the critical field was estimated to be above 75 T[42].

Figure 5 shows elastic neutron-scattering scans of the (1 0 3) magnetic Bragg peak at different temperatures: T=1.2, 8, 71, and 92 K of the x=0.17 sample, in zero and 14.5 T. While for the 1.2 K the 14.5 T magnetic intensity is ≈10% higher than for the zero field, our data also show that a field of 14.5 T leaves the magnetic scattering practically unchanged within the errors for the (T_c, T_N) temperature range. This result certainly contrasts with the clear decrease of the magnetic intensity by ≈10% in 13.5 T reported on Sn-flux grown higher potassium doped $BaFe_2As_2$ (with T_c=32±1 K) of J. T. Park *et al.*[30]. Therefore our 14.5 T data on the hole K-doped $BaFe_2As_2$ is similar to the 10 T high resolution neutron data on the electron underdoped $BaFe_{1.92}Ni_{0.08}As_2$ (T_c=17 K)[43]. Here, bellow T_c the intensity of the magnetic (1 1 3) peak is enhanced with ≈10%, while above T_c the intensity remains almost unchanged. One experiment to test the interplay between AFM and SC would be to determine whether or not a high magnetic field induces AFM in an optimally doped sample (without any trace of static AFM in zero field). This is very difficult to apply to the case of the optimally K-doped $BaFe_2As_2$, as the critical field is over 75 T. Since for the case of the electron-doped 122s the critical field is much lower, we performed zero and in-field (14 T) resistivity measurements of optimally doped Ni-doped $SrFe_2As_2$.

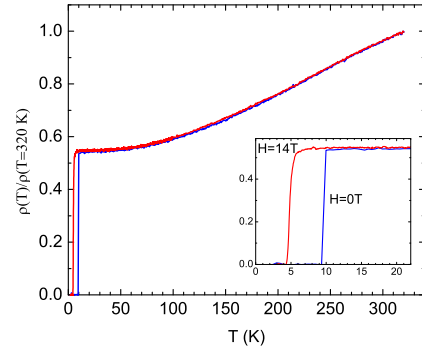Figure 6 shows resistivity vs. temperature of the

$SrFe_{2-x}Ni_xAs_2$, with x=0.155 normalized at its value at 320 K value, in zero and in 14 T$//c$ (it is known that for H$//c$ the critical field is lower than for H$//$(a b) configuration[44]). Before measurement, the sample was annealed in low argon gas pressure for 24 hours at 700° C[26,45]. Although T_c was suppressed from 10 to 5 K, there is no signature of any induced AFM in the 14 T$//c$ data. It is important to mention that Ni-doped samples with T_c of 5 K are well into the coexistence of Sc and AFM region on the phase diagram, therefore exhibiting robust AFM. Our high field resistivity data are in agreement therefore with neutron measurements in a 13.5 T$//c$ field on optimally electron-doped $BaFe_{1.9}Ni_{0.1}As_2$ that showed that the field did not induce static AFM order[46]. Therefore, part of the results of the hole- and electron-doped 122s seems to be consistent with a competing static AFM order and SC, similar to that for cuprate HTSc. The high field results reported here on the hole-doped $Ba_{0.83}K_{0.17}Fe_2As_2$ are similar to the case of cuprates for which the AFM order is strengthened with application of a magnetic field[47,48]. Despite the resemblance of the shape of the phase diagrams[50,51] for both iron pnictide and cuprate HTSc, the superconductivity in these materials appears to be of a different nature. We believe that these results will stimulate further exploration.

IV. SUMMARY

In summary, in the present article we report complementary elastic neutron scattering in zero and 14.5 T and magnetization measurements in zero and 14 T on underdoped SC x=0.17, and zero field on non-SC x=0.09. While for the non-SC x=0.09 sample the AFM transition is sharp consistent with a weakly 1st order transition, for higher doping x=0.17 the transition presents

a broad distribution on T_N due to a slight variation of the K dopant. For sample x=0.09 the temperature dependence of the magnetic OP is more gradual than for the case of parent $BaFe_2As_2$, indicative of proximity to a tricritical point. This tricritical point seems to be an universal feature among all superconducting 122s. The slight variation on the K dopant in the x=0.17 SC sample contributes to the fractional SC volume. Although the SC under-doped x=0.17 sample has a SC volume fraction of $\approx 40\%$, we were able to observe a downturn in the AFM order parameter below T_c, a clear sign of competition between AFM and SC, and similar to the one observed in the electron-doped 122s. As for the case of electron-doped 122s, a 14.5 T magnetic field enhances the AFM below T_c with $\approx 10\%$. This points, for the case at least of the 122s, toward a $s^{\pm}$ SC pairing symmetry[32] in the hole-doped material, similar to that in the electron-doped 122s. Finally we mention that while writing the current paper, we became aware of related work by E.

Wiesenmayer *et al.*[52]. Their combined X-ray and muon spin rotation on powder samples of the potassium underdoped materials show microscopical coexistence of AFM and SC.

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