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Unusual Sensitivity of Superconductivity to Strain in Iron-Based 122 Superconductors

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Abstract: Co-doped BaFe₂As₂ has been previously shown to have an unusually significant improvement of T_c (up to 2 K, or almost 10%) with annealing 1-2 weeks at 700 or 800 °C, where such annealing conditions are insufficient to allow significant atomic diffusion. While confirming similar behavior in optimally Co-doped SrFe₂As₂ samples, the influence on T_c of strain induced by grinding to ~50 μ sized particles, followed by pressing the powder into a pellet using 10 kbar pressure, was found to increase the annealed transition width of 1.5 K by approximately a factor of ten. Also, the bulk discontinuity in the specific heat at T_c , ΔC , on the same pellet sample was completely suppressed by grinding. This evidence for a strong sensitivity of superconductivity to strain was used to optimize single crystal growth of Co-doped BaFe₂As₂. This strong dependence (both positive via annealing and negative via grinding) of superconductivity on strain in these two iron based 122 structure superconductors is compared to the unconventional heavy Fermion superconductor UPt₃, where grinding is known to completely suppress superconductivity, and to recent reports of strong sensitivity of T_c to damage induced by electron-irradiationinduced point defects in other 122 structure iron-based superconductors, Ba(Fe_{0.76}Ru_{0.24})₂As₂ and Ba_{1-x}K_xFe₂As₂. Both the electron irradiation and the introduction of strain by grinding are believed to only introduce non-magnetic defects, and argue for unconventional superconducting pairing.

I. Introduction

Since the discovery¹ of iron-based superconductivity (for reviews, see refs. 2-5), a number of unusual properties in these fascinating materials have been discovered. Sefat et al. discovered⁶ superconductivity in $Ba(Fe_{1-x}Co_x)_2As_2$, with the peak of the superconducting transition temperature, T_c , dome vs composition at x=0.1 and the maximum T_c equal to 22 K in as-prepared single crystals.

One of the properties of these materials which aroused interest was the substantial increase in T_c in Ba(Fe_{1-x}Co_x)₂As₂ with annealing. Gofryk et al.⁷, in the first annealing experiments, reported that crystals of Ba(Fe_{1-x}Co_x)₂As₂ attained T_c =25 K and a decrease in the transition width ΔT_c of 25% after 2 weeks at 800 °C. Kim et al.⁸ further investigated T_c vs annealing in Ba(Fe_{1-x}Co_x)₂As₂. They found in their optimized self-flux grown samples that as-prepared crystals with T_c =25 K reached T_c values as high as 26.6 K via annealing for 1 week at 700 °C. With further optimization using finer gradations (in all, 17 different compositions between x=0.05 and 0.30) in Co concentration, Tam et al.⁹ found as-prepared Ba(Fe_{1-x}Co_x)₂As₂ crystals with T_c =25.5 K, with comparable annealing ¹⁰ resulting in T_c onset=27.2 K.

Such annealing, for 1 week at less ¹¹ than 60% of T_{melt} (i. e. without appreciable atomic diffusion) of pure $BaFe_2As_2$, resulting in such rapid increases in T_c (~1.7 K or ~7% of T_c), seemed more effective than in other superconductors. (For example, annealing elements like Cd or Zn to narrow ΔT_c is done ¹² at 95% of T_{melt} . The 14.4 K T_c superconductor YNi_2B_2C , prepared by melting together the constituents, when annealed at 1200 °C for 5 days (72% of T_{melt}), shows ¹⁴ no change either in T_c or ΔT_c .) The possibility that this unusual response of T_c with annealing is a clue to the unusual superconductivity in

iron-based superconductors motivated us in the present work to further investigate annealing in a second 122 structure system, $Sr(Fe_{1-x}Co_x)_2As_2$.

The results of this investigation as detailed below suggest a possible answer to the puzzle first posed by Gofryk et al. of why superconductivity in Ba(Fe_{1-x}Co_x)₂As₂ is improved so rapidly with relatively short annealing at only $\approx 60\%$ of T_{melt}.

II. Experimental

Single crystal samples of nominal composition $Sr(Fe_{0.86}Co_{0.14})_2As_2$ (near optimal doping) were prepared using self-flux growth techniques as in refs. 8-9. The crystals nucleate out of self-flux (FeAs) during a slow cool (3 °C/hr) between 1200 and 900 °C, followed by a more rapid cooling (75 °C/hr) to room temperature. Crystals are then separated from the self-flux mechanically. A single crystal of mass 18.3 mg was chosen for a series of measurements on the <u>same</u> sample: measurement of magnetic susceptibility, χ , and specific heat, C, on the unannealed crystal, annealing (700 °C for 2 weeks) of this crystal in an outgassed alumina crucible sealed via arc melting into a niobium cylinder containing an As vapor source⁸⁻⁹, then measurement of χ and C on the annealed crystal.

Following the comparison of χ and C for the same single crystal, unannealed and annealed, the annealed crystal was ground in an inert atmosphere glove box in an agate mortar for 2-3 minutes for x-ray characterization. Before being x-rayed, the powder was pressed into a pellet (at 150,000 psi (10 kbar) to avoid poor thermal contact between the grains) and the susceptibility of the pressed powder pellet was measured. When these data showed severe degradation of the superconductivity, the specific heat on the pressed

powder pellet was also measured. All of these measurements were on the same 18.3 mg single crystal, or the 12.9 mg pressed pellet from the powder therefrom.

The size of the powder making up the pellet (and of the powder made from a second crystal discussed below) was roughly determined by breaking it up as gently as possible with the blunt end of a wooden Q-tip and passing the powder through successively-sized sieves. Approximately half of the powder passed through a 270 mesh sieve (hole size 53 μ) and none of the powder passed through a 325 mesh sieve (hole size 45 μ).

In order to obtain an x-ray pattern on annealed powder to compare line widths and therefore strain between ground and annealed samples, a separate¹⁵ crystal of mass 43 mg was ground in the inert atmosphere glove box. One part of the powder was x-rayed and measured via magnetic susceptibility and a second part annealed (without an As vapor source) for 2 weeks at 700 °C, and then measured by x-ray diffraction as well as by magnetic susceptibility. In addition, x-ray diffraction was measured on the unannealed single crystal to obtain the line widths of 00L reflections as discussed below.

III. Results

It was expected from the previous annealing work⁸⁻⁹ on Ba(Fe_{1-x}Co_x)₂As₂ crystals that T_c^{onset} and the bulk transition width of the specific heat of the single crystal of Sr(Fe_{0.86}Co_{0.14})₂As₂ would increase and narrow respectively upon annealing for 2 weeks at 700 °C. The susceptibility (Fig. 1) and specific heat (Fig. 2) data of the unannealed and annealed 18.3 mg single crystal of Sr(Fe_{0.86}Co_{0.14})₂As₂ confirm this expectation. T_c^{onset} increases with annealing as measured by the susceptibility/bulk specific heat by ~1.5 K/0.9

K and the transition in the specific heat at T_c , ΔC , sharpens considerably. These results are indeed comparable to those⁷⁻⁹ in optimally doped Ba(Fe_{1-x}Co_x)₂As₂.

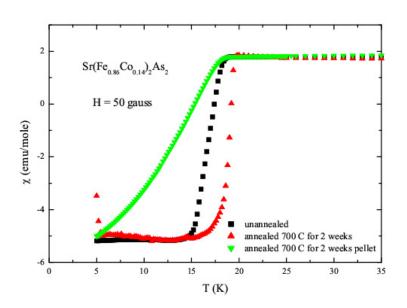


Fig. 1 (color online) Magnetic susceptibility, χ , vs temperature of an 18.3 mg single crystal of $Sr(Fe_{0.86}Co_{0.14})_2As_2$ in three conditions: unannealed crystal (black squares), crystal after annealing at 700 °C for 2 weeks (red triangles), and ground powder pressed into a pellet (green inverted triangles), >45 μ diameter, made from the annealed single crystal. T_c^{onset} increases ~ 1.5 K with annealing, while the transition width, ΔT_c , decreases from 2.4 to 1.5 K.

What was not known previously⁷⁻⁹ is the very strong influence of grinding, followed by pressing into a pellet, on the superconductivity. As shown in Fig. 1, grinding¹⁶ the annealed 18.3 mg crystal to a grain size of no smaller than 45 μ , followed by pressing into a pellet, results in a large increase (from ~1.5 K to over 14 K) in the transition width

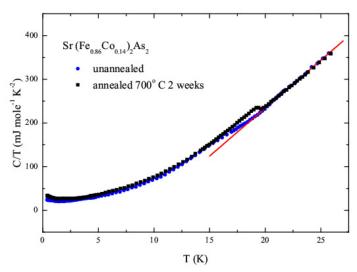


Fig. 2: (color online) Specific heat divided by temperature, C/T, vs temperature of an 18.3 mg single crystal of $Sr(Fe_{0.86}Co_{0.14})_2As_2$, unannealed (solid blue squares) and annealed at 700 °C for 2 weeks (solid black circles.) T_c^{onset} as measured by the bulk specific heat improves by \sim 0.9 K with annealing, comparable to work⁸ on $Ba(Fe_{1-x}Co_x)_2As_2$. The red line is an extrapolation of the normal state data to below T_c . The finite intercept of C/T ($T\rightarrow 0$), defined as $\gamma_{residual}$, in these samples is ~ 20 mJ/molK², which is larger than that

found8 in Co-doped BaFe2As2 and could indicate the presence of some normal material in

the crystal. However, the measured discontinuity in C at T_c =19.5 K, $\Delta C/T_c$ = 19 mJ/molK² in the annealed sample is within 25% of that⁸ for Co-doped BaFe₂As₂.

of the superconducting transition as measured by the magnetic susceptibility. Further, as shown in Fig. 3, specific heat of the pressed pellet of this same ground powder shows that the specific heat discontinuity at T_c , ΔC , present in both the annealed and unannealed crystal (Fig. 2) is totally smeared out in the ground material.

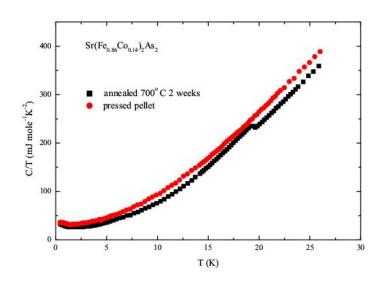


Fig. 3 (color online) Specific heat divided by temperature, C/T, versus temperature for annealed single crystal, 18.3 mg (solid black squares) and a pressed pellet of the ground powder, 12.9 mg (solid red circles), >45 µ diameter, from the same annealed crystal of $Sr(Fe_{0.86}Co_{0.14})_2As_2$. The absolute error bar for the pressed pellet data above 20 K is $\pm 5\%$ vs $\pm 3\%$ for the single crystal due to the larger addenda contribution (38 % vs 17 %). However, the relative precision $(\pm 1-2 \%)$ between the two measurements is sufficient to state

that the larger C/T for the pressed pellet sample is qualitatively correct.

In order to quantify the amount of strain introduced by the grinding, a separate 43 mg Co-doped SrFe₂As₂ crystal was ground¹⁵ in the same fashion, and some of the (homogenized) powder was annealed for 2 weeks at 700 °C. We then measured x-ray diffraction and susceptibility on a portion¹⁵ of the unannealed powder from this second crystal as well as on an annealed portion of the powder. The susceptibility of the unannealed starting crystal and on the ground powder are consistent with the results in

Fig. 1, while the susceptibility data of the annealed powder are consistent both in $T_c^{\,onset}$ and transition width with the annealed single crystal shown in Fig. 1, i. e. the annealed powder

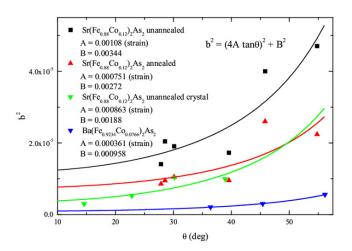


Fig. 4 (color online) Analysis of the line width, b (radians), vs angle of the xray reflections (hkl) to determine strain of unannealed (black squares) and annealed (red triangles) powder, as well as – using (00L) reflections - of an asgrown single crystal (green inverted triangles) of Sr(Fe_{0.88}Co_{0.12})₂As₂ and of a cooled-to-600 °C crystal (i. e. quasiannealed) of Ba(Fe_{0.9234}Co_{0.0766})₂As₂, blue inverted triangles. For example, for the (0010) line the full width (when plotted vs Θ) at half maximum in units of

 10^{-3} radians for the four different samples is 4.15, 3.09, 3.15, and 1.45 respectively. 'B' is the instrumentally caused line broadening. Errors bars for strain are ± 0.00005 except for the Co-doped Ba 122 sample, where the error bar is only ± 0.00002 . (Note that a term $(\{0.9\lambda/Dcos\Theta\}^2)$ in the equation for the line width that involves the particle size, D, is omitted since, with $D\sim 50~\mu$, the term is negligible.)

 T_c and ΔT_c are improved vis-à-vis the unannealed single crystal. Analysis ¹⁷ (Fig. 4) of the x-ray line widths of various (hkl) reflections of the annealed and unannealed powders between 55 and 110 degrees 2Θ results in a strain ϵ for the annealed powder of 0.0008 \pm 0.0001 and for the unannealed powder of 0.0011 \pm 0.0001, a small but - as evident from Fig. 4 – easily measurable ¹⁸ difference. Thus, although the annealing does cause a change in the amount of strain in the material, the small amount of the difference indicates a high sensitivity of the superconductivity to strain. Consistent with the susceptibility result (not shown) just discussed (that T_c (unannealed crystal) $< T_c$ (annealed powder)), the analyzed strain in Fig. 4 in the unannealed single crystal of $Sr(Fe_{0.88}Co_{0.12})_2As_2$ is indeed slightly larger ¹⁹ than that of the annealed powder, 0.000863 vs 0.000751.

These results make it evident that the superconductivity in Co-doped SrFe₂As₂ is a.) very sensitive to strain and b.) the strain present in ground powder, with its very broadened ΔT_c and $\Delta C \rightarrow 0$, vs that in annealed material differs by a relatively small amount (~30%). Thus, the question arises: what is the minimal amount of annealing necessary to improve the superconductivity in unannealed single crystals? Phrased another way, what minimal further heat treatment on the as-grown self-flux crystals (with the slow (3 °C/hr) cooling halted at 900 °C) is necessary to remove the strain which the present work implies is introduced by cooling from 900 °C to room temperature at 75 °C/hr? Ref. 8 states that annealing at 600 °C has essentially no effect on T_c^{onset} or ΔT_c in Co-doped BaFe₂As₂, therefore presumably removing this small amount of residual strain in the as-grown single crystals cooled slowly to 900 °C requires thermal treatment above 600 °C.

In order to make a first attempt at answering this question, and in a different 122 structure iron based superconductor in order to broaden the applicability of these results, we undertook the following. To verify indeed that the strain involved is produced by cooling at 75 °C/hr the as-grown crystals from 900 °C to somewhere above 600 °C (based on the ref. 8 result), we have reproduced/altered the growth procedure in our previous thorough study of annealing in Co-doped BaFe₂As₂ (refs. 8 and 9) for x=0.0766 (a composition slightly below that of optimal doping) as follows. Two batches of Ba(Fe_{0.9234}Co_{0.0766})₂As₂ crystals were grown in self flux, one heated to 1200 °C, cooled at 3 °C/hr until 900 °C, followed by cooling at 75 °C/hr to room temperature (the original procedure, followed also herein for Co-doped SrFe₂As₂.) The second batch was identical in every respect except it was cooled from 1200 °C down to 600 °C at 3 °C/hr, and then at 75

°C/hr to room temperature. This extra temperature region of slow cooling did not result in larger crystals, since the crystals have already formed²⁰ by 900 °C, but it adds slow cooling (roughly equivalent to annealing for the same length of time at a fixed intermediate temperature) over a period of about 3 days from 900 down to 700 °C. The susceptibility of single crystals from both batches is shown in Fig. 5. Clearly, the strain removed by annealing at 700 °C for 2 weeks in the present work, or at 700 °C for 1 week as in refs. 8-9, can also be removed by merely cooling at 3 °C/hr further down in temperature, past the previous 900 °C changeover-in-cooling rate point, to 600 °C. In fact, as shown in the strain analysis graph, Fig. 4, the cooled-to-600 °C crystal of Ba(Fe_{0.9234}Co_{0.0766})₂As₂ shows a strain only half of that of the annealed for 2 weeks at 700 °C powdered sample – arguing for the effectiveness of the slow cooling procedure.

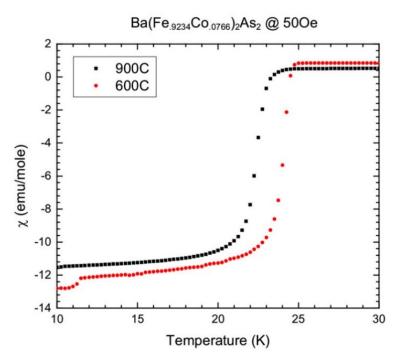


Fig. 5 (color online) Magnetic susceptibility, χ, vs temperature for single crystals with the nominal composition of Ba(Fe_{0.9234}Co_{0.0766})₂As₂ prepared either by cooling at 3 °C/hr from 1200 °C to 900 °C, followed by cooling at 75 °C/hr to room temperature (black points) or by cooling at 3 °C/hr down to 600 °C, followed by cooling at 75 °C/hr to room temperature (red points). The difference in T_c (either midpoint or onset) is approximately 1.7 K, the same as found⁹ after 1 week at 700 °C annealing as-grown crystals slow cooled down to 900 °C. Note the somewhat sharper onset to superconductivity upon cooling in the red points, whereas the black data is somewhat more rounded.

IV. Discussion and Conclusions

The first conclusion that can be reached is the solution to the puzzle raised by the previous annealing work $^{7-9}$ on Co-doped BaFe₂As₂: why does annealing 1-2 weeks at 700 $^{\circ}$ C, only 60% of T_{melt} have such an important effect on T_{c}^{onset} and the bulk specific heat transition width, ΔT_{c} ? Clearly, the superconductivity in both the Co-doped SrFe₂As₂ and BaFe₂As₂ 122 iron based superconductors - and presumably other 122's and 111's as well (although see the discussion below of CaFe₂As₂) - is extremely sensitive to strain as shown by the results presented above.

Before we discuss why T_c and ΔT_c are in Co-doped SrFe₂As₂ and BaFe₂As₂ are so sensitive to strain, we now make a digression in order to discuss whether the current results will apply to Co-doped CaFe₂As₂. First, it is important to note that results exist detailing how annealing affects the normal state properties in all three 122's. Annealing (at either 350 or 700 °C between 1 and 30 days) has been shown²¹ to have a relatively small (1-6 K) effect on the magnetic and structural transitions for BaFe2As2 and SrFe2As2 (at 135 K and 200 K respectively), and to change the c-axis lattice parameters at room temperature by less than 0.01 Å. This is in stark contrast to CaFe₂As₂ (quenched from 960 °C in order to decant the crystals from the FeAs self-flux), where annealing at 400 °C for 1 week changes²² the low temperature structure from a non-collapsed tetragonal phase below 100 K to an orthorhombic, antiferromagnetic state below 170 K, and increases²¹ the c-axis lattice parameter by 0.152 Å. As an explanation for the 'extreme case' of CaFe₂As₂, ref. 22 explains that the effect of annealing is to remove very fine, ~10 nm, precipitates whose average strain field mimics the effect of 0.4 GPa pressure, and to allow the formation of the necessary-for-superconductivity orthorhombic antiferromagnetic state. Since a.) refs. 8 and 9 found that annealing at temperature above 600 °C is necessary to improve T_c in

Ba(Fe_{1-x}Co_x)₂As₂ (where the current work tells us that the annealing is causing the removal of strain harmful for superconductivity), and since b.) ref. 23 finds that annealing at 600 °C and above in Co-doped CaFe₂As₂ forms the non-collapsed tetragonal, non-magnetic, inimical-to-superconductivity phase, the removal of strain from grinding \Rightarrow improved T_c results presented here in Sr(Fe_{1-x}Co_x)₂As₂ likely cannot be used to optimize superconductivity (T_c~16 K) in Co-doped CaFe₂As₂. It would be interesting to measure the amount of strain present in the rather low T_c Co-doped CaFe₂As₂ samples – is the strain larger than seen in the higher T_c Sr(Fe_{1-x}Co_x)₂As₂ and Ba(Fe_{1-x}Co_x)₂As₂, or is the explanation the difference²³ in the order of the magnetic behavior in CaFe₂As₂ (strong first order) and the lack²³ of coexistence between magnetism and superconductivity anywhere in Co-doped CaFe₂As₂?

Returning now to our main question: why are T_c and ΔT_c in Co-doped SrFe₂As₂ and BaFe₂As₂ so sensitive to strain? Such a strong dependence of superconductivity on strain in Co-doped Ba and Sr 122 is indicative of an unconventional superconducting mechanism. One well known example²⁴ of a very strain sensitive superconductor is UPt₃, where grinding²⁴⁻²⁵ totally destroys superconductivity (from a T_c of ~ 0.5 K to below 0.05 K as measured by susceptibility.) The f-wave pairing symmetry in UPt₃ is expected to be very sensitive to damage and defects.²⁶

Theory⁵ suggests that the pairing mechanism, the so called s± scheme where the order parameter changes sign between different sheets of the Fermi surface, favored for the iron-based superconductors is also extremely sensitive to defects, not just magnetic defects as are known to degrade conventional superconductors but also including non-magnetic²⁷ defects introduced by grinding. Thus, the original intent of the present work –

to see if understanding the unusually rapid improvement of T_c^{onset} with annealing at only 60% of T_{melt} for just 1 week in $BaFe_{2-x}Co_xAs_2$ could shed light on the superconductivity in iron based superconductors – has produced evidence for extreme sensitivity of the superconductivity to defects introduced via grinding. This is reminiscent of the behavior of the known unconventional superconductor UPt_3 .

Another way to introduce non-magnetic defects in the lattice is via electron irradiation, which has been performed on 122 iron superconductors, UPt₃, as well as in the unconventional cuprate high temperature superconductors – thus allowing a quantitative intercomparison among all three. Electron irradiation, using 2.5 MeV electrons, by approximately 1.1 10^{19} e/cm² gives T_c reductions from the unirradiated T_{c0} (T_c/T_{c0}) of 0.84 for²⁸ UPt₃, 0.80 for²⁹ Ba(Fe_{0.76}Ru_{0.24})₂As₂, 0.87/0.66 for³⁰ Ba_{1-x}K_xFe₂As₂ (x=0.19/0.34) and 0.92 for³¹ YBa₂Cu₃O₇. These results are consistent with the extreme sensitivity of the superconductivity in Co-doped SrFe₂As₂ to grinding and the induced strain therefrom found in the present work. As well, the relief of a small amount of strain by replacing a 75 °C/hr cooling from 900 to 600 °C with 3 °C/hr cooling and the concomitant increase of T_c^{onset} by ~ 1.7 K in Co-doped BaFe₂As₂ is further consistent with the electron irradiation evidence²⁹⁻³⁰ for the extreme sensitivity of iron based superconductivity, with presumed²⁹⁻³⁰ s± pairing symmetry, to non-magnetic defects.

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- 16. Since the 122 iron based superconductors such as Co-doped BaFe₂As₂ and Co-doped SrFe₂As₂ are quite brittle, such grinding involves qualitatively (this will be quantified via xray line width analysis in later discussion) only a small application of force.
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which is consistent with increased magnetic behavior upon grinding. However, such behavior does not appear to be affecting T_c or ΔT_c .

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