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# Competition between orthorhombic and re-entrant tetragonal phases in underdoped $Ba_{1-x}K_xFe_2As_2$ probed by the response to controlled disorder

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# Competition between orthorhombic and re-entrant tetragonal phases in underdoped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ probed by the response to controlled disorder

## Abstract

Low-temperature (22 K) irradiation with 2.5-MeV electrons, creating point defects affecting elastic scattering, was used to study the competition between stripe C-2 and tetragonal C-4 antiferromagnetic phases which exist in a narrow doping range around  $x = 0.25$  in hole-doped  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ . In nearby compositions outside of this range, at  $x = 0.22$  and  $x = 0.19$ , the temperatures of both the concomitant orthorhombic/stripe antiferromagnetic transition  $T_{\text{C}2}$  and the superconducting transition  $T_{\text{c}}$  are monotonically suppressed by added disorder at similar rates of about 0.1 K/ $\mu\Omega$  cm, as revealed through using resistivity variation as an intrinsic measure of scattering rate. In a stark contrast, a rapid suppression of the C-4 phase at the rate of 0.24 K/ $\mu\Omega$  cm is found at  $x = 0.25$ . Moreover, this suppression of the C-4 phase is accompanied by unusual disorder-induced stabilization of the C-2 phase, determined by resistivity and specific heat measurements. The rate of the C-4 phase suppression is notably higher than the suppression rate of the spin-vortex phase in the Ni-doped  $\text{CaKFe}_4\text{As}_4$  (0.16 K/ $\mu\Omega$  cm).

## Disciplines

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## Competition between orthorhombic and re-entrant tetragonal phases in underdoped $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ probed by the response to controlled disorder

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Low-temperature (22 K) irradiation with 2.5-MeV electrons, creating point defects affecting elastic scattering, was used to study the competition between stripe  $C_2$  and tetragonal  $C_4$  antiferromagnetic phases which exist in a narrow doping range around  $x = 0.25$  in hole-doped  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ . In nearby compositions outside of this range, at  $x = 0.22$  and  $x = 0.19$ , the temperatures of both the concomitant orthorhombic/stripe antiferromagnetic transition  $T_{C_2}$  and the superconducting transition  $T_c$  are monotonically suppressed by added disorder at similar rates of about  $0.1 \text{ K}/\mu\Omega \text{ cm}$ , as revealed through using resistivity variation as an intrinsic measure of scattering rate. In a stark contrast, a rapid suppression of the  $C_4$  phase at the rate of  $0.24 \text{ K}/\mu\Omega \text{ cm}$  is found at  $x = 0.25$ . Moreover, this suppression of the  $C_4$  phase is accompanied by unusual disorder-induced stabilization of the  $C_2$  phase, determined by resistivity and specific heat measurements. The rate of the  $C_4$  phase suppression is notably higher than the suppression rate of the spin-vortex phase in the Ni-doped  $\text{CaKFe}_4\text{As}_4$  ( $0.16 \text{ K}/\mu\Omega \text{ cm}$ ).

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Cooper pair binding mediated by magnetic fluctuations [1] is actively discussed as a possible mechanism of superconductivity in several classes of unconventional superconductors including heavy fermions [2], high- $T_c$  cuprates [3], and, more recently, iron-based superconductors [4]. A fingerprint of this model is the observation of the highest superconducting transition temperature,  $T_c$ , coinciding with a quantum critical point (QCP) where the temperature of the magnetic transition,  $T_N$ , goes to zero at a point in a  $T$ - $x$  phase diagram, with  $x$  being a nonthermal control parameter such as doping, pressure, magnetic field, or disorder [1,5,7]. Strong magnetic fluctuations at the QCP lead to non-Fermi liquid behavior of all electronic properties, for example, logarithmic divergence of the heat capacity and  $T$ -linear electrical resistivity [5,6].

In iron-based superconductors, this phenomenology is clearly observed in isovalent P-substituted  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  ( $\text{Ba122}$ ) [8–10]; however it fails in hole-doped  $\text{Ba}_{1-x}\text{A}_x\text{Fe}_2\text{As}_2$  ( $A = \text{Na}$  and  $\text{K}$ ) compositions which have the highest  $T_c$ . Here, the suppression of the transition temperature  $T_{C_2}$  of the orthorhombic antiferromagnetic phase with a stripe pattern of in-plane moments ( $C_2$  phase) [11,12] does not proceed monotonically to zero, but rather is interrupted by the emergence of a new tetragonal  $C_4$  magnetic phase below temperature  $T_{C_4}$  [13–18]. Being in very close proximity to the highest  $T_c$  doping range, this phase may play an important, yet not understood, role in the superconducting pairing [19].

The  $C_4$  phase is also observed in other hole-doped 122-type compounds, such as  $\text{Ca}_{1-x}\text{Na}_x\text{Fe}_2\text{As}_2$  [20],  $\text{Sr}_{1-x}\text{Na}_x\text{Fe}_2\text{As}_2$  [21], and  $\text{Ba}(\text{Fe}_{1-x}\text{Mn}_x)_2\text{As}_2$  [22]. The  $C_4$  phase in  $\text{Sr}_{1-x}\text{Na}_x\text{Fe}_2\text{As}_2$  was shown to be a double- $Q$  spin-charge density wave, with a moment equal to zero on every second iron atom [23]. A similar  $C_4$  phase but with a different type of magnetic order was found in electron-doped  $\text{CaK}(\text{Fe}_{1-x}\text{TM}_x)_2\text{As}_2$ , with  $\text{TM} = \text{Co}$  and  $\text{Ni}$  [24]. Theoretically, the origin of this phase has been attributed to itinerant magnetism [25,26], magnetic moments with effects of frustration [27], or the effects of spin-orbit coupling [28,29].

It was recently suggested that disorder can lead to a stabilization of the spin-charge density wave  $C_4$  phase as compared to the  $C_4$  spin-vortex state and the  $C_2$  phase in the phase diagram of the hole-doped compositions [30]. Motivated by this theoretical prediction, we report here a study on the effect of electron irradiation in hole-doped  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ , with  $x = 0.25$ , revealing clear signatures of the  $C_4$  phase in temperature-dependent resistivity and heat capacity measurements. For reference, we also study the effect of electron irradiation on nearby compositions with  $x = 0.19$  and  $x = 0.22$  outside the  $C_4$  phase doping range. We find that disorder suppresses the  $C_4$  phase at a rate which is significantly higher than the suppression rate of the  $C_2$  phase in nearby compositions and in the spin-vortex phase of  $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$  [31]. It also leads to an unusual slight increase of  $T_{C_2}$ , suggesting its stabilization with disorder. Our results clearly show competition between these two types of magnetic orders.

Single crystals of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  were grown as described in detail in Ref. [32]. Large, above  $5 \times 5 \text{ mm}^2$  surface area, crystals were cleaved on both sides to a thickness of typically

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0.1 mm to minimize the variation of the K content with thickness. The cleaved slabs were characterized by electron-probe microanalysis with wavelength dispersive spectroscopy (WDS). The crystals from three different batches were used with WDS compositions determined as  $x = 0.19, 0.22,$  and  $0.25$ . The large slabs were cleaved into bars for four-probe resistivity measurements so that all samples were originating from the same slab of the crystal. Samples typically had a size of  $2 \times 0.5 \times 0.1 \text{ mm}^3$  and long and short sides corresponding to the crystallographic  $a$  axis and  $c$  axis, respectively. Low-resistance contacts to the samples were made by soldering Ag wires with tin [33,34]. The contacts were found to be both mechanically and electronically stable under electron irradiation. Four-probe resistivity measurements were performed in a Quantum Design PPMS. Specific heat was measured in a helium cryostat by using an ac calorimeter built on SiN membrane chips at frequencies in the 1-Hz range as described in Refs. [35,36].

For our study we selected samples with the sharpest features in the temperature-dependent resistivity  $\rho(T)$  at concomitant tetragonal/orthorhombic and paramagnetic/ $C_2$  antiferromagnetic transitions in samples  $x = 0.19$  and  $0.22$ . The largest problem however is finding samples with sharp features at the  $C_2$  to  $C_4$  transition for  $x = 0.25$ , which is extremely sensitive to sample-to-sample variation without detectable composition variations with  $\Delta x \sim 0.003$ . We therefore did all precharacterization of the samples with resistivity and only performed specific heat measurement on selected samples.

The samples for resistivity measurements during and after electron irradiation were mounted on a thin mica plate in a hollow Kyocera chip, so that they could be moved between the irradiation chamber and the resistivity setup (in a different  $^4\text{He}$  cryostat) without disturbing the contacts. The low-temperature 2.5-MeV electron irradiation was performed at the SIRIUS Pelletron linear accelerator operated by the Laboratoire des Solides Irradiés (LSI) at the Ecole Polytechnique in Palaiseau, France [37]. The Kyocera chip was mounted inside the irradiation chamber and was cooled by a flow of liquid hydrogen to  $T \approx 22 \text{ K}$  in order to remove excess heat produced by relativistic electrons upon collision with the ions. The flux of electrons amounted to about  $2.7 \mu\text{A}$  of electric current through a 5-mm-diameter diaphragm. This current was measured with the Faraday cup placed behind a hole in the sample stage, so that only transmitted electrons were counted. The irradiation rate was about  $5 \times 10^{-6} \text{ C}/(\text{cm}^2 \text{ s})$  and large doses were accumulated over the course of several irradiation runs. Throughout the manuscript we use “pristine” and “unirradiated” interchangeably to describe samples that were not exposed to electron radiation.

Three samples selected, A, B, and C, had sharp maximums in temperature-dependent resistivity derivatives at  $T_{C4}$  equal to 33 K (A) and 35 K (B and C) and had minimums at  $T_{C2}$  equal to 60 K (A) and 56.5 K (B and C), respectively. The selected sample A of  $x = 0.25$  composition was irradiated multiple times, adding doses in small steps and tracking the fine evolution of its temperature-dependent resistivity to determine  $T_{C2}$ ,  $T_{C4}$  and the superconducting  $T_c$ . The sample was extracted from the irradiation chamber following each irradiation dose

step and its temperature-dependent resistivity was measured *ex situ* after annealing at room temperature. This annealing, however, did not remove residual disorder, so that the sample resistance gradually increased in successive runs. A second sample B with slightly higher  $T_{C4} \sim 35 \text{ K}$ , suggesting somewhat higher K content was mounted on the same chip and underwent the same irradiation procedure; however, it was not measured in the intermediate steps. After an accumulation of a significant dose and the ensuing characterization by resistivity which produced results that were qualitatively consistent with sample A (we found a slight increase in  $T_{C2}$  by about 2 K), a small piece ( $100 \mu\text{m} \times 160 \mu\text{m}$ ) was cut from the area between potential contacts to be used for microcalorimetric measurements. Another pristine sample C, having identical  $T_{C4}$  and  $T_{C2}$  with sample B, was measured as a reference sample in the specific heat apparatus. The samples of other compositions  $x = 0.19$  and  $x = 0.22$  were irradiated without intermediate measurements, receiving the maximum dose in one run.

In Fig. 1 we show the temperature-dependent resistivity of selected samples with  $x = 0.19, 0.22,$  and  $0.25$  in the pristine state before irradiation. The room-temperature resistivity of the samples was set to  $300 \mu\Omega \text{ cm}$ , the statistically significant value as determined on a big array of crystals [32]. The actually measured values for the individual samples were within the 10% uncertainty of the geometric factor determination. The  $\rho(T)$  curves show the typical behavior of hole-doped  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  [32,38], with a broad crossover at around 200 K. Samples with  $x = 0.19$  and  $x = 0.22$  show a small acceleration of resistivity decrease upon cooling through  $T_{C2}$  and a rather sharp superconducting transition at  $T_c$ . The  $T_{C2}$  feature is most clearly seen as a sharp feature in the temperature derivative of the resistivity,  $d\rho/dT$  [Fig. 1(b)]. The  $\rho(T)$  of the sample with  $x = 0.25$  shows a slight step up at  $T_{C2}$ , leading to a sharp minimum in resistivity derivative. The resistivity of the samples just above  $T_c$  decreases monotonically with  $x$  from about  $40 \mu\Omega \text{ cm}$  in  $x = 0.19$  to  $30 \mu\Omega \text{ cm}$  in  $x = 0.25$  and the residual resistivity ratios increase from about 7 to 10, respectively. The  $T_{C2}$  feature is shifting down in temperature with increasing  $x$  reaching  $T_{C2} = 60 \text{ K}$  for  $x = 0.25$  [the same feature in samples B and C is observed at 56.5 K in resistivity and at 57.4 K in heat capacity (sample C), indicating its bulk nature, see Fig. 3(a) below]. In Fig. 1(c) we plot the characteristic temperatures as determined from resistivity measurements (circles represent  $T_{C2}$  and open up-triangles represent  $T_c$  as determined from offset criterion) as a function of  $x$  in comparison with the phase diagram by Böhmer *et al.* [15] (lines in the figure). The position of the  $x = 0.25$  sample in this phase diagram does not follow the  $T_{C2}$  line. However, if we allow for a small variation of  $x$  for our  $x = 0.25$  WDS sample to match  $T_{C2}$  with the value reported by Böhmer *et al.* [15], we simultaneously match the  $T_{C4}$  feature (red solid square) as well. The composition difference amounts to approximately 1%, which is presumably coming from the difference in calibrations in the composition analysis between WDS (our case) and energy dispersive X-ray spectroscopy (as used by Böhmer *et al.* [15]). The onset of the resistive transition to the superconducting phase in samples A and B (not shown) occurs at 30 K with no indication of the  $T_c$  depression reported in Ref. [15].

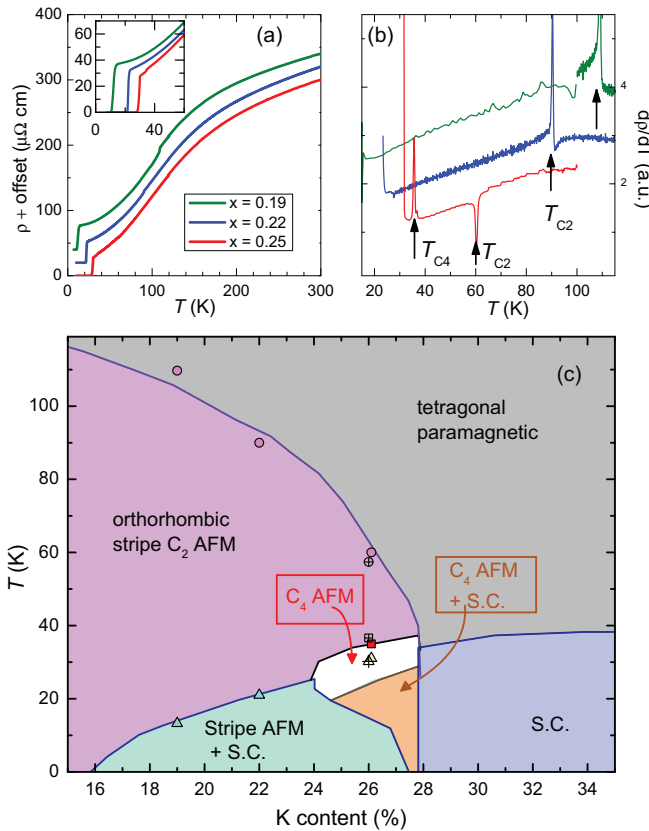


FIG. 1. (a) Temperature-dependent resistivity of selected samples of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ ,  $x = 0.19$  (green),  $0.22$  (blue), and  $0.25$  (red). The curves are offset vertically. Inset: Low-temperature region showing differences in the superconducting transition temperatures and resistivity values at  $T_c$ . (b) Resistivity derivative, revealing a sharp feature at the structural transitions at  $T_{C2}$  and  $T_{C4}$ . (c) Doping phase diagram of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  in the range of  $C_4$  phase formation, as proposed by Böhmer *et al.* [15] (lines). The positions of  $C_2$  (circles),  $C_4$  (square), and superconducting (triangle) transitions for samples with  $x = 0.19$  and  $x = 0.22$  are matching well with the diagram, but the position of  $x = 0.25$  sample was adjusted to  $0.264$  to match the  $C_2$  line with the concomitant match of the  $C_4$  line. Symbols with crosses show the positions of the features in the heat capacity measurements (see Fig. 3 below).

In Fig. 2 we show the evolution of the temperature-dependent resistivity  $\rho(T)$  with electron irradiation. The irradiation increases the resistivity of the samples, with the increase being nearly temperature independent above  $T_{C2}$ , but strongly temperature dependent below  $T_{C2}$ . This difference in response to controlled disorder above and below  $T_{C2}$  is found in other  $\text{BaFe}_2\text{As}_2$ -based materials, P-doped [39], Ru-doped [40,41], and K-doped [42,43]. Since the resistivity above  $T_{C2}$  roughly obeys the Matthiessen rule, we used the postirradiation increase of resistivity at the set temperature  $T = 95$  K (dashed lines in the left-hand panels in Fig. 2) as an intrinsic measure of disorder. The electron dose dependence of the resistivity for samples  $x = 0.22$  (blue circles) and  $x = 0.25$  (green squares) is shown in the inset in Fig. 2(a).

Irradiation suppresses  $T_{C2}$  in samples with  $x = 0.19$  (not shown) and  $x = 0.22$  [Fig. 2(b)]. This is similar to the results of previous studies for all types of substitutions in  $\text{BaFe}_2\text{As}_2$

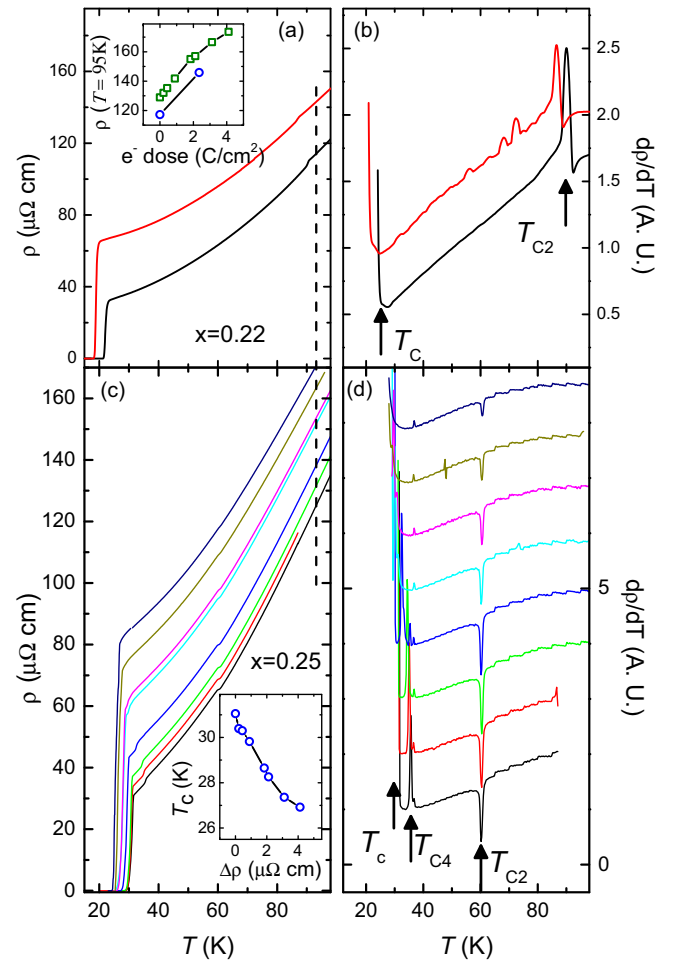


FIG. 2. Temperature-dependent resistivity (left panels) and resistivity derivative (right panels) of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  with  $x = 0.22$  (top panels) and  $x = 0.25$  (bottom panels). Black curves in panels (a) and (b) are for the pristine  $x = 0.22$  sample, and red curves are for the sample after electron irradiation with  $2.35$   $\text{C}/\text{cm}^2$ . Panels (c) and (d) show systematics of the evolution of the temperature-dependent electrical resistivity in the sample with  $x = 0.25$  with irradiation, bottom to top: pristine sample (black),  $0.212$  C (red),  $0.438$  C (green),  $0.893$  C (blue),  $1.835$  C (cyan),  $2.115$  C (magenta),  $3.115$  C (dark yellow), and  $4.115$  C (navy). The inset in panel (a) shows resistivity at  $95$  K as a function of electron irradiation dose for the samples with  $x = 0.22$  (blue) and  $x = 0.25$  (green). Inset in panel (c) shows the evolution of the superconducting transition temperature in the sample with  $x = 0.25$  as a function of the change of resistivity at  $95$  K, above  $T_{C2}$ .

[39–43]. The response to irradiation in the  $x = 0.25$  sample is qualitatively different [Fig. 2(c)]. While the superconducting transition temperature is monotonically suppressed with increasing resistivity, the  $T_{C4}$  feature moves to lower temperatures significantly faster than  $T_c$  and eventually becomes indistinguishable from the superconducting transition. Furthermore, the  $T_{C2}$  feature is not suppressed with increasing scattering, but, in fact, a slight increase of  $T_{C2}$  with irradiation is found in heat capacity measurements.

The findings in resistivity measurements are well matched by the heat capacity measurements. In the pristine state [Fig. 3(a)], clear changes of slope are seen in the  $C/T$  vs

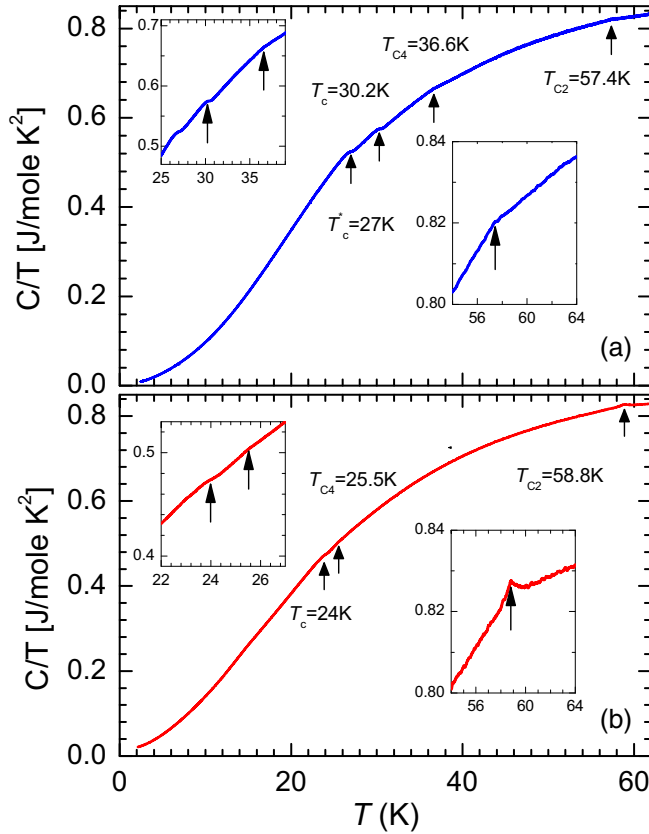


FIG. 3. Temperature-dependent heat capacity,  $C/T$ , of the  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$   $x = 0.25$  sample before (a) and after (b) electron irradiation with  $5.045 \text{ C/cm}^2$ . The right-hand insets zoom in on the  $T_{C2}$  phase transition, and the left-hand insets zoom in on the low-temperature transitions.

$T$  plot at  $T_{C2} = 57.4 \text{ K}$  and  $T_{C4} = 36.6 \text{ K}$  as well as two low-temperature features corresponding to the superconducting transition and possibly the reentrant  $C_2$  phase. These features are shown with crossed symbols in Fig. 1 above.  $T_{C4}$  is strongly suppressed after irradiation, faster than the superconducting transition, while the  $C_2$  transition becomes sharper and moves slightly up in temperature.

In Fig. 4 we summarize our observations as plots of characteristic temperatures for  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  as a function of change of resistivity after irradiation. Figure 4(a) shows data for the sample with  $x = 0.19$ , Fig. 4(b) shows data for the sample with  $x = 0.22$ , and Fig. 4(c) shows data for the sample with  $x = 0.25$ . Note that the rates of the superconducting transition suppression with disorder,  $0.091$ ,  $0.118$ , and  $0.098 \text{ K}/\mu\Omega \text{ cm}$  for  $x = 0.19$ ,  $0.22$ , and  $0.25$ , respectively, are very close to each other and to the rate of the  $T_{C2}$  suppression,  $0.096$  and  $0.105$  for  $x = 0.19$  and  $x = 0.22$ . The rate of the  $C_4$  phase suppression in the  $x = 0.25$  sample,  $0.21 \text{ K}/\mu\Omega \text{ cm}$  in resistivity and  $0.24 \text{ K}/\mu\Omega \text{ cm}$  in heat capacity measurements, is about two times faster than that of the  $C_2$  phase suppression in the  $x = 0.19$  and  $x = 0.22$  samples. This rate is also significantly higher than the rate of  $C_4$  spin-vortex phase suppression in  $\text{CaK}(\text{Fe}_{1-x}\text{Ni}_x)_4\text{As}_4$ ,  $0.16 \text{ K}/\mu\Omega \text{ cm}$  [31]. A slight increase of  $T_{C2}$  in the  $x = 0.25$  sample is found after irradiation in heat capacity measurements.

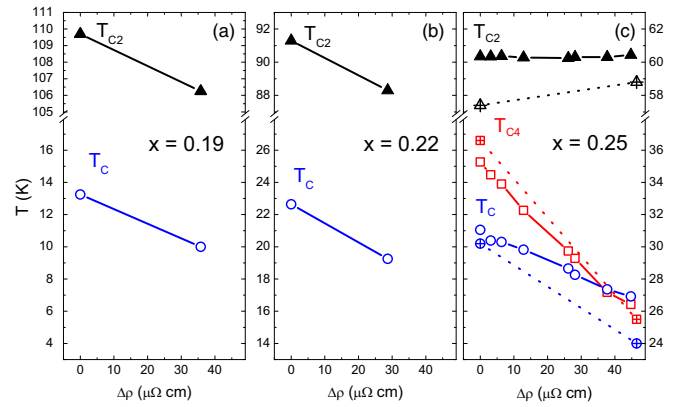


FIG. 4. Transition temperatures of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$   $x = 0.19$  (a),  $x = 0.22$  (b), and  $x = 0.25$  (c) samples as a function of scattering rate increase characterized with resistivity increase in the paramagnetic tetragonal phase above  $T_{C2}$ . Note the similar rates of superconducting  $T_c$  suppression in all compositions (blue open circles), the fast suppression of the  $C_2$  phase (black solid up-triangles) in  $x = 0.19$  and  $0.22$  samples, the two times faster suppression of the  $C_4$  phase in  $x = 0.25$  samples (open red squares from resistivity measurements, crossed squares from heat capacity measurements), and the slight increase of  $T_{C2}$  with irradiation in  $x = 0.25$  samples (solid up-triangles resistivity measurements, crossed triangles from heat capacity measurements). Blue crossed circles are  $T_c$  from heat capacity measurements.

It is most natural to explain our findings as evidence for competition between the  $C_2$  and the  $C_4$  phases, with a suppression of the  $C_4$  phase leading to a stabilization for a certain parameter range in the calculations of Hoyer *et al.* [30,44], though this paper considers the case of phase competition near the magnetic transition temperature as opposed to the case of the  $C_4$  phase existing deep in the domain of the  $C_2$  phase as found in our experiment.

In conclusion, we find that controlled disorder introduced by low-temperature irradiation with relativistic 2.5-MeV electrons rapidly suppresses the transition temperature between antiferromagnetic  $C_2$  and  $C_4$  phases and leads to the relative stabilization of the  $C_2$  phase. This behavior can be found for the parameter range characterized by weak nesting in the itinerant electron magnetism model by Hoyer *et al.* [30], though the phase stability relations were considered only at the transition temperature for magnetic ordering. Our findings suggest that further theoretical analysis that will consider possible first-order transition between two phases, and hence phase coexistence and possible separation, may be necessary.

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