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Expansion of the tetragonal magnetic phase with pressure in the iron arsenide superconductor $Ba_{1-x}K_xFe_2As_2$

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In the temperature-concentration phase diagram of most iron-based superconductors, antiferromagnetic order is gradually suppressed to zero at a critical point, and a dome of superconductivity forms around that point. The nature of the magnetic phase and its fluctuations is of fundamental importance for elucidating the pairing mechanism. In $Ba_{1-x}K_xFe_2As_2$ and $Ba_{1-x}Na_xFe_2As_2$, it has recently become clear that the usual stripelike magnetic phase, of orthorhombic symmetry, gives way to a second magnetic phase, of tetragonal symmetry, near the critical point, in the range from $x = 0.24$ to $x = 0.28$ for $Ba_{1-x}K_xFe_2As_2$. In a prior study, an unidentified phase was discovered for $x < 0.24$ but under applied pressure, whose onset was detected as a sharp anomaly in the resistivity. Here we report measurements of the electrical resistivity of $Ba_{1-x}K_xFe_2As_2$ under applied hydrostatic pressures up to 2.75 GPa, for $x = 0.22$, 0.24, and 0.28. The critical pressure above which the unidentified phase appears is seen to decrease with increasing x and vanish at $x = 0.24$, thereby linking the pressure-induced phase to the tetragonal magnetic phase observed at ambient pressure. In the temperature-concentration phase diagram of $Ba_{1-x}K_xFe_2As_2$, we find that pressure greatly expands the tetragonal magnetic phase, while the stripelike phase shrinks. This reveals that pressure may be a powerful tuning parameter with which to explore the interplay between magnetism and superconductivity in this material.

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I. INTRODUCTION

The phase diagram of iron-based superconductors of the $BaFe_2As_2$ family is characterized by competing antiferromagnetic (AF) order and superconductivity. Usually, the AF order decreases with concentration (doping) and a dome of superconductivity surrounds the critical point [1]. The AF order is a stripelike spin-density wave, with a wave vector $\mathbf{Q} = (\pi,0)$ and the magnetic moments lying in the plane. At the magnetic transition temperature, or slightly above it, the lattice changes from tetragonal at high temperature to orthorhombic at low temperature [2,3].

In $Ba_{1-x}X_xFe_2As_2$, where X = K or Na, the phase diagram was recently found to be richer than this simple picture. Resistivity measurements under pressure revealed the existence of an internal transition inside the AF phase of $Ba_{1-x}K_xFe_2As_2$ [4]. As the onset temperature T_N of the orthorhombic AF phase (o-AF) is suppressed with hydrostatic pressure P, an additional phase transition to a "new phase" appears below a transition temperature $T_0 < T_N$, for $0.16 < x < 0.21$, when $P > 0.9$ GPa [4]. A tetragonal magnetic phase (t-AF) was then discovered in the closely related compound $Ba_{1-x}Na_xFe_2As_2$, by neutron and x-ray diffraction on powder samples [5]. Subsequent neutron scattering on single crystals showed that in this t-AF phase the spins are aligned parallel to the c axis [6]. A similar phase of tetragonal symmetry was then found in $Ba_{1-x}K_xFe_2As_2$ at ambient pressure, for $0.24 < x < 0.28$ [7]. The magnetic moments in the t-AF phase of $Ba_{1-x}K_xFe_2As_2$ are also oriented along the c axis [8,9]. Infrared spectroscopy showed that the t-AF phase has a double-Q magnetic structure [8], as opposed to the single-Q structure of the o-AF phase. A pressure study of a $Ba_{1-x}K_xFe_2As_2$ sample with $x = 0.15$ by specific heat, transport, and the Nernst effect confirms the bulk nature of the sequence of phase transitions previously detected only in resistivity [10]. Additionally, the authors show that the pressure-induced "new phase" suppresses the large Nernst signal of the o-AF phase, indicating the suppression of the nematicity as in the t-AF phase at ambient pressure. Several theoretical studies have investigated the properties of the tetragonal magnetic phase in iron-based superconductors [5,11–20].

In this article, we extend our prior study of $Ba_{1-x}K_xFe_2As_2$ under pressure, performed up to $x = 0.21$ [4], by studying three further samples, with $x = 0.22$, 0.24, and 0.28. We are able to connect the additional phase induced by pressure with the tetragonal phase seen at ambient pressure. Pressure is seen to cause a dramatic expansion of the tetragonal magnetic phase, on the backdrop of a shrinking orthorhombic phase.

II. METHODS

Single crystals of $Ba_{1-x}K_xFe_2As_2$ were grown from self-flux [21]. Three underdoped samples were measured, with a superconducting transition temperature $T_c = 20.8 \pm 0.5$, 25.4 ± 0.5, and 30.1 ± 0.5 K, respectively. Using the relation between T_c and the nominal K concentration x reported in

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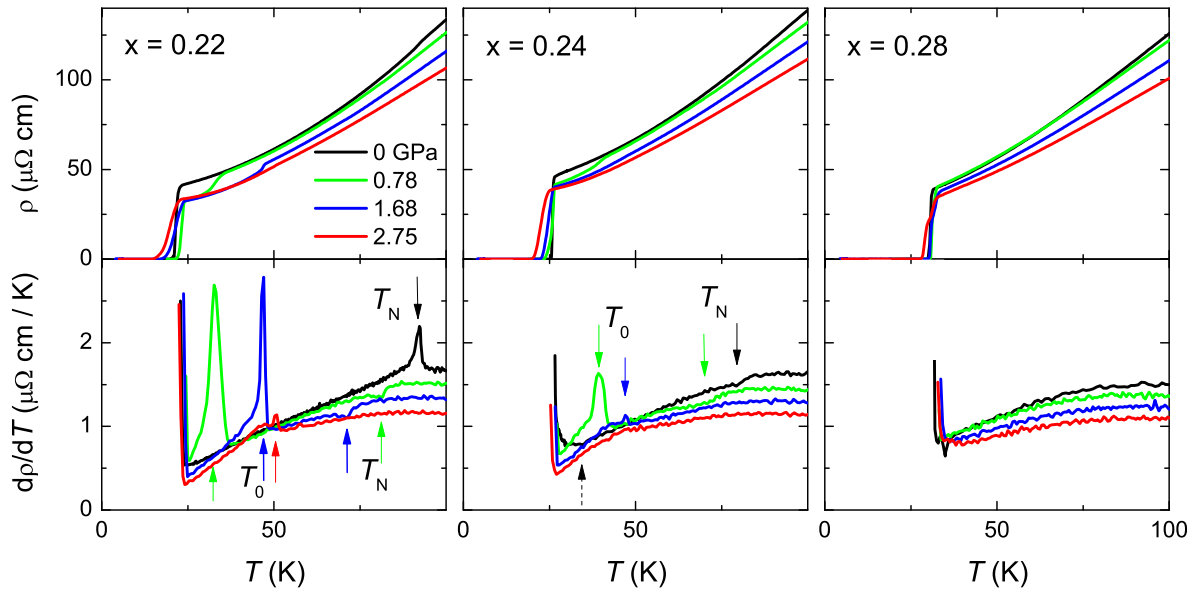

FIG. 1. Top: In-plane electrical resistivity of $Ba_{1-x}K_xFe_2As_2$ for $x = 0.22$, 0.24, and 0.28 (different columns) for four different pressures, as indicated. Bottom: Temperature derivative of the data in the top panels. The peak (dip) between 60 and 100 K signals the onset of stripelike antiferromagnetic order at T_N (arrows). The peak at lower temperature signals the onset of the tetragonal magnetic phase at T_0 (arrows). In the lower middle panel ($x = 0.24$), the dashed arrow marks the foot of the peak at T_0 in the black curve at ambient pressure.

Ref. [3] and wavelength-dispersive x-ray spectroscopy [22], we obtain $x = 0.22$, 0.24, and 0.28, respectively. These x values are also consistent with the measured antiferromagnetic ordering temperature T_N (which coincides with the structural transition from tetragonal to orthorhombic) [3], equal to 91 ± 2 and 79 ± 5 K, respectively for the two lower dopings. The sample with $x = 0.28$ shows no magnetic or structural transition. The resistivity at room temperature of all samples lies between 250 and 350 $\mu\Omega$ cm, in agreement with previous studies [23]. As before [4], we have normalized the resistivity at $T = 300$ K to 300 $\mu\Omega$ cm. Hydrostatic pressures up to 2.75 GPa were applied with a hybrid piston-cylinder cell [24], using a 50:50 mixture of *n*-pentane:isopentane. This pressure transmitting medium has been shown to present the best hydrostatic conditions, i.e., the smallest uniaxial pressure component, in the pressure range up to 3 GPa [25]. The pressure was measured via the superconducting transition of a lead wire inside the pressure cell. The electrical resistivity ρ was measured for a current in the basal plane of the orthorhombic crystal structure, with a standard four-point technique using a Lakeshore ac-resistance bridge. The transition temperatures are defined as follows: T_c is where $\rho = 0$; T_N and T_0 are detected as extrema in the derivative $d\rho/dT$.

III. RESISTIVITY

Figure 1 shows the in-plane resistivity (top panels) and its temperature derivative (bottom panels) of each sample, for a selection of pressures. T_N is detected as a peak in the derivative for the first sample at ambient pressure, and then as a dip for higher pressures or doping. The transition at T_0 shows up as a sharp peak, below T_N. For those concentrations and applied pressures where both T_N and T_0 are detected, the resistivity curves and their temperature derivatives resemble those of a sample with $x = 0.25$ at ambient pressure, where the t-AF phase is present (see the Supplemental Material of Ref. [7].) In that publication, resistivity is identified as a good probe of T_0 via a comparison with thermodynamic probes such as the thermal expansion or specific heat. In Fig. 2, the full set of derivative curves is displayed for $x = 0.22$ and 0.24, allowing one to track the anomalies at T_N and T_0 as a function of pressure.

As previously reported for samples with lower doping [4], T_N decreases linearly with pressure. For $x = 0.22$, the peak in the derivative at T_N evolves into a dip at 0.48 GPa. We are able to follow this dip up to $P = 2.0$ GPa, above which it disappears. The evolution of the peak at T_0 is different. At 0.48 GPa, the peak at T_0 appears. T_0 goes up with pressure until it stays almost constant above 2.3 GPa. The height of the sharp peak at T_0 increases slightly at first, and then decreases above $P \simeq 1.5$ GPa. The behavior for $x = 0.24$ is similar, but shifted to lower pressures. T_N can be followed only up to 0.94 GPa. The transition at T_0 appears as a peak as soon as we apply pressure. In fact, a slight upturn of the derivative with decreasing T, indicative of an onset of the transition at T_0, can be seen even at ambient pressure. The onset is marked by an up-pointing dashed arrow in the lower middle panel of Fig. 1. We see that the new phase is present in this sample at $P = 0$. This provides a direct link between what was initially called the "new phase" and what is now known to be the t-AF phase. (In our previous study, a similar situation was found for $x = 0.19$ at $P = 1.08$ GPa. At zero magnetic field, a slight onset of the transition at T_0 was seen above T_c, which was completely uncovered by a magnetic field of $H = 15$ T shifting the T_c far below T_0, which is itself unaffected by the field [4].) This

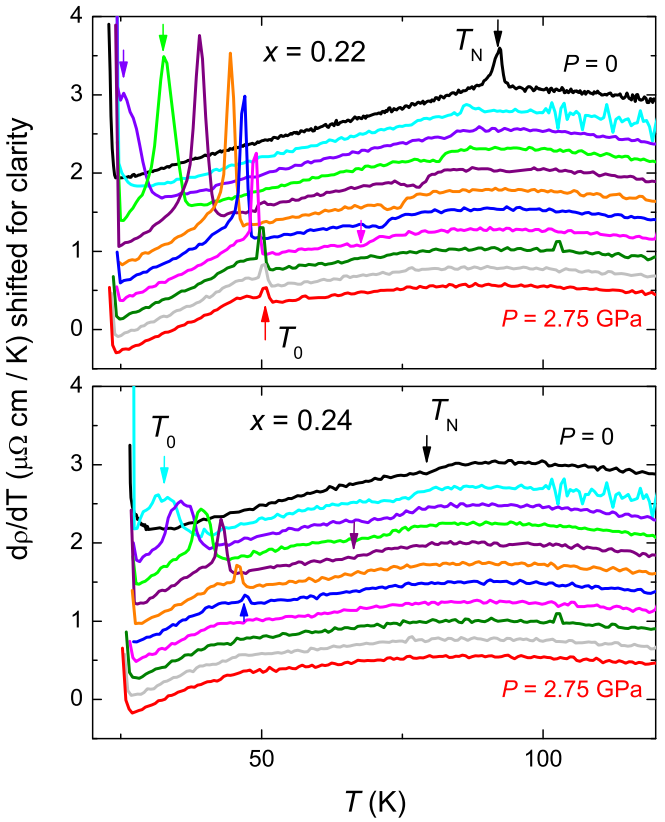

FIG. 2. Top: Temperature derivative of the resistivity of $Ba_{1-x}K_xFe_2As_2$ with $x = 0.22$, for 11 different pressures, from ambient pressure ($P = 0$) at the top (black) to $P = 2.75$ GPa at the bottom (red), with the following intermediate values: $P = 0.28, 0.48$, 0.78, 0.94, 1.37, 1.68, 2.0, 2.31, and 2.4 GPa. The curves are shifted for clarity. The black down-pointing arrow marks T_N at $P = 0$. The next down-pointing arrow marks T_N at the highest pressure where it can still be detected. T_0 shows up as a peak at low temperature (e.g. down-pointing arrows below 50 K). The up-pointing arrow marks T_0 at the highest pressure where the peak can still be detected. Bottom: The same for $x = 0.24$.

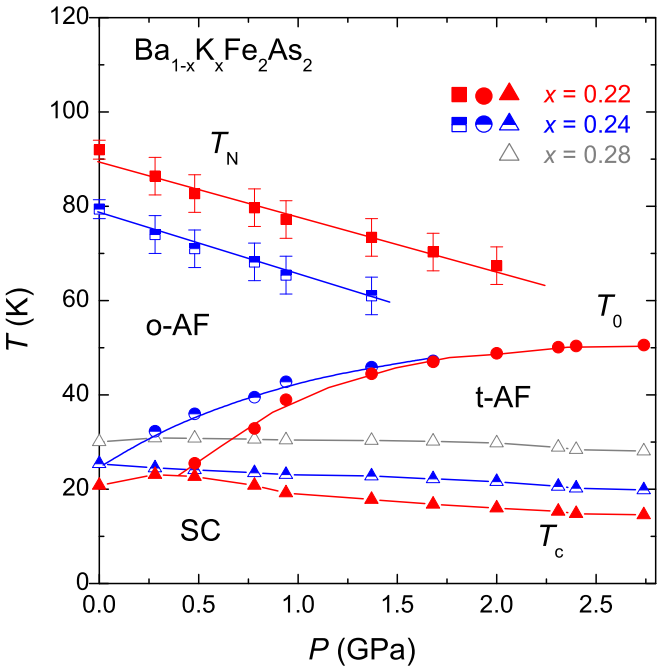

FIG. 3. Temperature-pressure phase diagram of $Ba_{1-x}K_xFe_2As_2$, for $x = 0.22$, 0.24, and 0.28 (solid, half-solid, and open symbols, respectively), showing the orthorhombic antiferromagnetic (o-AF) transition temperature T_N (squares), the superconducting (SC) transition temperature T_c (triangles), and the tetragonal antiferromagnetic (t-AF) transition temperature T_0 (circles).

$x = 0.24$ sample is apparently right at the border of the t-AF phase, as a very tiny amount of either pressure or additional K content is enough to clearly induce the t-AF phase. The peak at T_0 stays sharp but its height decreases above $P \simeq 1$ GPa, and the last pressure where it is observed is 1.68 GPa. The curve at this pressure looks very much as the one at the highest pressure in the $x = 0.22$ sample.

IV. TEMPERATURE-PRESSURE PHASE DIAGRAM

Figure 3 presents the temperature-pressure phase diagram for the three samples. T_N decreases linearly with P, with a slightly steeper slope at $x = 0.24$. By contrast, T_0 rises rapidly, at least initially. At $x = 0.22$, T_0 saturates above $P = 2.3$ GPa. At $x = 0.24$, we can no longer detect T_0 above $P = 1.68$ GPa (Fig. 2), the pressure at which it merges with the T_0 line at $x = 0.22$ (Fig. 3).

At $x = 0.24$, the phase diagram is such that if the T_0 line (blue) saturates at high pressure, as it does in the case of $x = 0.22$ (red T_0 line), then a linear extension of the T_N line (blue) will hit that T_0 line, implying that the t-AF phase would persist to pressures beyond the end of the o-AF phase.

As for superconductivity, note that T_c decreases as soon as the tetragonal phase appears (Fig. 3), as found in prior studies of $Ba_{1-x}K_xFe_2As_2$ [4,26] and $Ba_{1-x}Na_xFe_2As_2$ [26,27], in agreement with the negative dT_c/dP expected from the Ehrenfest relation applied to the thermodynamic data [7].

V. TEMPERATURE-CONCENTRATION PHASE DIAGRAM

Combining our present results with those of our previous study [4], we plot the temperature-concentration phase diagram of $Ba_{1-x}K_xFe_2As_2$ in Fig. 4. For comparison, we also reproduce the phase diagram at zero pressure reported in Ref. [7]; the agreement with our own ambient-pressure data is excellent. We see that the T_N line moves down with pressure, in parallel fashion. This suggests that the critical concentration x_N where T_N goes to zero shifts down with pressure.

On the backdrop of this shrinking o-AF phase, the tetragonal magnetic phase undergoes a major expansion with pressure (Fig. 4). While the t-AF phase occupies a small area below T_N at ambient pressure, its area grows by an order of magnitude at $P = 2.4$ GPa. In other words, at high pressure the tetragonal phase becomes the dominant magnetic phase in the temperature-concentration phase diagram of $Ba_{1-x}K_xFe_2As_2$. A recent study of thermal expansion and specific heat revealed a complex phase diagram in $Ba_{1-x}Na_xFe_2As_2$ with an expanded tetragonal phase [28]. There, in agreement with our results, chemical pressure might lead to the expansion of the

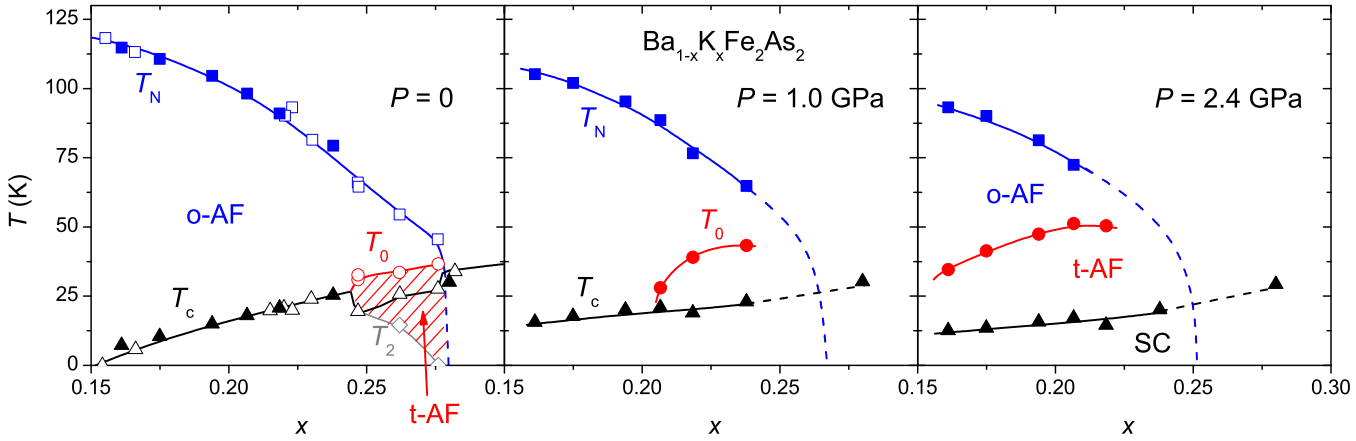

FIG. 4. Temperature-concentration phase diagram of $Ba_{1-x}K_xFe_2As_2$, showing T_N (blue squares), T_0 (red circles), and T_c (black triangles), for three different values of the applied pressure: $P = 0$ (left panel), 1.0 GPa (middle panel), and 2.4 GPa (right panel). This includes data from our previous study [4]. Ambient-pressure data from Ref. [7] are also shown in the left panel (open symbols), including a transition back to the o-AF phase, below T_2 (diamonds). All lines are a guide to the eye. The evolution from left to right, with increasing pressure, reveals a major expansion of the tetragonal magnetic phase (t-AF), on the backdrop of a shrinking stripe phase (o-AF). Extrapolating to higher pressure, we expect the former to become the dominant magnetic phase coexisting with superconductivity in $Ba_{1-x}K_xFe_2As_2$.

tetragonal phase [28]. In the context of recent calculations, it may be that pressure favors the t-AF phase because it changes the ellipticity of the electron pockets in the Fermi surface of $Ba_{1-x}K_xFe_2As_2$ [16].

VI. SUMMARY

In summary, we have shown that the new phase discovered in $Ba_{1-x}K_xFe_2As_2$ from sharp signatures in the resistivity under pressure [4] is the tetragonal antiferromagnetic phase observed and identified subsequently by various probes in both $Ba_{1-x}Na_xFe_2As_2$ [5,6] and $Ba_{1-x}K_xFe_2As_2$ [7–9]. Under pressure, this t-AF phase expands enormously, by an order of magnitude for 2.4 GPa in terms of the area it occupies in the temperature-concentration phase diagram, relative to the orthorhombic stripelike AF phase that dominates at ambient pressure. As a result, at high pressure, superconductivity exists on the border of a dominant tetragonal magnetic phase. It is then likely that fluctuations of that double-Q phase play a role in the pairing. Recent calculations suggest that such fluctuations could actually enhance T_c [19].

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[1] P. C. Canfield and S. L. Budk'o, Annu. Rev. Condens. Matter Phys. **1**, 27 (2010).

[2] D. K. Pratt, W. Tian, A. Kreyssig, J. L. Zarestky, S. Nandi, N. Ni, S. L. Budâko, P. C. Canfield, A. I. Goldman, and R. J. McQueeney, Phys. Rev. Lett. **103**, 087001 (2009).

[3] S. Avci, O. Chmaissem, D. Y. Chung, S. Rosenkranz, E. A. Goremychkin, J. P. Castellan, I. S. Todorov, J. A. Schlueter, H. Claus, A. Daoud-Aladine, D. D. Khalyavin, M. G. Kanatzidis, and R. Osborn, Phys. Rev. B **85**, 184507 (2012).

[4] E. Hassinger, G. Gredat, F. Valade, S. R. de Cotret, A. Juneau-Fecteau, J.-Ph. Reid, H. Kim, M. A. Tanatar, R. Prozorov, B. Shen, H.-H. Wen, N. Doiron-Leyraud, and L. Taillefer, Phys. Rev. B **86**, 140502(R) (2012).

[5] S. Avci, O. Chmaissem, J. M. Allred, S. Rosenkranz, I. Eremin, A. V. Chubukov, D. E. Bugaris, D. Y. Chung, M. G. Kanatzidis, J.-P Castellan, J. A. Schlueter, H. Claus, D. D. Khalyavin, P. Manuel, A. Daoud-Aladine, and R. Osborn, Nat. Commun. **5**, 3845 (2014).

[6] F. Waßer, A. Schneidewind, Y. Sidis, S. Wurmehl, S. Aswartham, B. Büchner, and M. Braden, Phys. Rev. B **91**, 060505(R) (2015).

[7] A. E. Böhmer, F. Hardy, L. Wang, T. Wolf, P. Schweiss, and C. Meingast, Nat. Commun. **6**, 7911 (2015).

[8] B. P. P. Mallett, P. Marsik, M. Yazdi-Rizi, Th. Wolf, A. E. Böhmer, F. Hardy, C. Meingast, D. Munzar, and C. Bernhard, Phys. Rev. Lett. **115**, 027003 (2015).

[9] J. M. Allred, S. Avci, D. Y. Chung, H. Claus, D. D. Khalyavin, P. Manuel, K. M. Taddei, M. G. Kanatzidis, S. Rosenkranz, R. Osborn, and O. Chmaissem, Phys. Rev. B **92**, 094515 (2015).
[10] Y. Zheng, P. M. Tam, J. Hou, A. E. Böhmer, T. Wolf, C. Meingast, and R. Lortz, Phys. Rev. B **93**, 104516 (2016).
[11] J. Lorenzana, G. Seibold, C. Ortix, and M. Grilli, Phys. Rev. Lett. **101**, 186402 (2008).
[12] I. Eremin and A. V. Chubukov, Phys. Rev. B **81**, 024511 (2010).
[13] E. Berg, S. A. Kivelson, and D. J. Scalapino, Phys. Rev. B **81**, 172504 (2010).
[14] G. Giovannetti, C. Ortix, M. Marsman, M. Capone, J. van den Brink, and J. Lorenzana, Nat. Commun. **2**, 398 (2011).
[15] P. M. R. Brydon, J. Schmiedt, and C. Timm, Phys. Rev. B **84**, 214510 (2011).
[16] J. Kang, X. Wang, A. V. Chubukov, and R. M. Fernandes, Phys. Rev. B **91**, 121104(R) (2015).
[17] X. Wang, J. Kang, and R. M. Fernandes, Phys. Rev. B **91**, 024401 (2015).
[18] M. N. Gastiasoro and B. M. Andersen, Phys. Rev. B **92**, 140506 (2015).
[19] R. M. Fernandes, S. A. Kivelson, and E. Berg, Phys. Rev. B **93**, 014511 (2016).
[20] J. M. Allred, K. M. Taddei, D. E. Bugaris, M. J. Krogstad, S. H. Lapidus, D. Y. Chung, H. Claus, M. G. Kanatzidis, D. E. Brown, J. Kang, R. M. Fernandes, I. Eremin, S. Rosenkranz, O. Chmaissem, and R. Osborn, arXiv:1505.06175.
[21] H. Luo, Z. Wang, H. Yang, P. Cheng, X. Zhu, and H.-H. Wen, Supercond. Sci. Technol. **21**, 125014 (2008).
[22] M. A. Tanatar, W. E. Straszheim, H. Kim, J. Murphy, N. Spyrison, E. C. Blomberg, K. Cho, J.-Ph. Reid, B. Shen, L. Taillefer, H.-H. Wen, and R. Prozorov, Phys. Rev. B **89**, 144514 (2014)
[23] Y. Liu, M. A. Tanatar, W. E. Straszheim, B. Jensen, K. W. Dennis, R. W. McCallum, V. G. Kogan, R. Prozorov, and T. A. Lograsso, Phys. Rev. B **89**, 134504 (2014)
[24] I. R. Walker, Rev. Sci. Instrum. **70**, 3402 (1999).
[25] W. J. Duncan, O. P. Welzel, C. Harrison, X. F. Wang, X. H. Chen, F. M. Grosche, and P. G. Niklowitz, J. Phys.: Condens. Matter **22**, 052201 (2010).
[26] S. L. Bud'ko, M. Sturza, D. Y. Chung, M. G. Kanatzidis, and P. C. Canfield, Phys. Rev. B **87**, 100509(R) (2013).
[27] S. L. Bud'ko, D. Y. Chung, D. Bugaris, H. Claus, M. G. Kanatzidis, and P. C. Canfield, Phys. Rev. B **89**, 014510 (2014).
[28] L. Wang, F. Hardy, A. Böhmer, T. Wolf, P. Schweiss, and C. Meingast, Phys. Rev. B **93**, 014514 (2016).