

NMR evidence of pairing fluctuations above T_c and absence of spin magnetism in the time-reversal symmetry-breaking state of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$

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Zero-field muon-spin-rotation (μSR) experiments and the observation of a spontaneous Nernst effect indicate broken time-reversal symmetry (BTRS) at $T_c^{\mathbb{Z}_2}$, slightly above the superconducting transition temperature T_c in $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, with $x \sim 0.8$. This was interpreted as evidence of a distinct state of matter where BTRS above T_c arises from noncondensed Cooper pairs, which maintain long-range interband correlations. However, an inhomogeneous superconducting state competing with magnetism as the origin of the observed behavior cannot be completely ruled out to date. Here, we present detailed ^{75}As nuclear magnetic resonance (NMR) measurements of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, with $x = 0.766$, yielding $T_c^{\mathbb{Z}_2} > T_c$. The NMR data demonstrate that the superconductivity is homogeneous throughout the entire sample volume despite the occurrence of nanoclustering of Ba/K atoms. We observe a suppression of the spin-lattice relaxation rate $1/T_1T$ below the characteristic temperature T^* , well above T_c . The specific heat yields somewhat lower values of T^* , consistent with the microscopic theory of electron quadrupling. The observed monotonous suppression of T^* and $T_c^{\mathbb{Z}_2}$ in applied magnetic field, the overall temperature dependence of $1/T_1T$, the NMR spectral characteristics, and the μSR Knight shift rule out spin magnetism and the proximity to a magnetic instability. We conclude that our data indicate the appearance of pairing fluctuations below T^* and that the breaking of time-reversal symmetry represents an unconventional state which is unrelated to spin magnetism.

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I. INTRODUCTION

A number of experiments on $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ in a narrow doping range ($0.75 \lesssim x \lesssim 0.81$) revealed several unprecedented and unconventional effects. They include the observation of a broken time-reversal symmetry (BTRS) superconducting state [1,2] and vortices that carry a fraction of the magnetic flux quantum in the superconducting state [3,4]. Importantly, the fractionalization of vortex cores was

demonstrated for the surface of KFe_2As_2 , having an effective doping level $x \sim 0.75$ [5]. Particularly unusual observations were made above the superconducting transition at T_c which were interpreted as the formation of a new state of matter, an electron-quadrupling condensate that breaks time-reversal symmetry ($\mathbb{Z}_2$) [6–8].

The quadrupling condensate is distinguished by symmetry from the superconducting state below T_c and the normal state above the BTRS transition temperature $T_c^{\mathbb{Z}_2} > T_c$. In the quadrupling state, superconductivity is suppressed by phase fluctuations, resulting in $\langle \Delta_a \rangle = 0$. From symmetry arguments, a BTRS transition above T_c implies that the order parameter is fourth order in fermionic fields $\langle \Delta_a \Delta_b^* \rangle \neq 0$, where the gaps Δ_a and Δ_b belong to different bands. The important consequence of the quartic order parameter is that it results in the appearance of spontaneous real-space loop currents [6,9].

In support of this interpretation, two anomalies were observed in specific-heat measurements of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ [7].

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The anomaly at higher temperature, $T_c^{\mathbb{Z}_2}$, coincides with the appearance of a spontaneous Nernst signal, and the anomaly at lower temperature, T_c , corresponds to the appearance of zero resistance. BTRS superconductivity in this doping range was initially proposed based on muon-spin rotation (μ SR) measurements [1,2], which were later found to be in good agreement with other anomalies [6], also indicating that the BTRS state exists above T_c in a narrow potassium-doping range close to $x \sim 0.8$. It was concluded that BTRS superconductivity and the BTRS state between T_c and $T_c^{\mathbb{Z}_2}$ is preceded by pairing correlations, which must set in at the temperature T^* , which is above $T_c^{\mathbb{Z}_2}$. In support of this interpretation, the electric and thermoelectric transport data indicated the existence of a crossover temperature T^* at this doping range [6].

The evidence of the existence of the quadrupling state in $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ relies on a coherent interpretation of various experimental signatures [6,7]. In particular, it is essential that the observed BTRS state near the superconducting phase transition is unrelated to magnetic order.

In this study, we pursue two aims: (1) testing a scenario in which the BTRS state is related to spin magnetism instead of being a result of real-space spontaneous loop currents as claimed before and (2) discriminating between inhomogeneous superconductivity and superconducting or, more generally, pairing fluctuations as the origin of the anomalous behavior above T_c at $x \sim 0.8$. Our main findings are as follows: (1) The absence of a dynamic correlation peak in the NMR $1/T_1T$ rate, as well as of any spectroscopic signatures of static magnetism in the transverse-field (TF) μ SR and NMR data, excludes the possibility that spin magnetism is responsible for the BTRS state. (2) The superconductivity evolves homogeneously from fluctuating Cooper pairs in the entire sample volume, despite the nanoclustering of Ba and K atoms.

II. EXPERIMENT

A. Samples

We performed the NMR measurements on a platelike $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ single crystal with a mass of 0.33 mg and a doping level of $x = 0.766(1)$. We show sample-characterization data using various probes, as well as additional results of the NMR and μ SR analysis, in the Supplemental Material (SM) [10].

The K doping level x of the single crystal used for the NMR measurements was determined using the known relation between the c -axis lattice parameter and the K doping determined in previous studies: $x = -14.574 + 1.1234c$ (Å) and $R = 0.99852$ [11]. To confirm that the sample is single phase, we performed 2Θ x-ray measurements from both sides of the crystal (see Fig. 1 in the SM [10]). We obtained the same c -axis parameter within 0.01%, indicating a homogeneous distribution of the K doping in the NMR sample. For the μ SR measurements, we selected about 30 crystals based on the T_c obtained in low-field magnetization measurements using Quantum Design magnetic property measurement system and physical property measurement system (PPMS) devices. The resulting real part of the AC susceptibility in zero static (DC) magnetic field is shown in Fig. 3 in the SM [10]. The

μ SR sample has a slightly higher T_c and a broader transition width compared to the NMR sample. Using the T_c value and observed transition width, we estimated the doping level of the μ SR sample, $x = 0.75(2)$ K, using the previously obtained phase diagram from Ref. [2].

The low-temperature specific heat at different magnetic fields was measured in a Quantum Design PPMS using the thermal-relaxation method.

B. NMR

We performed the NMR measurements in a ^4He flow cryostat mounted in an 8 T superconducting magnet with a spatial field homogeneity of about 1 ppm over the sample volume. The ^{75}As NMR data were recorded using a commercial phase-coherent spectrometer. We used standard Hahn spin-echo pulse sequences to measure the spectra, with carefully adjusted pulse parameters in order to avoid radio-frequency (RF) heating effects. We measured the nuclear spin-lattice relaxation time T_1 by using a saturation-recovery method and described the data using the stretched-exponential function for the central transition ($I_z = -1/2 \leftrightarrow +1/2$; see the SM for more details [10]).

C. μ SR

We performed high-TF μ SR measurements at the HAL-9500 spectrometer (PSI, Villigen) on a stack of selected crystals with a total mass of about 13 mg. The muon polarization was in the ab plane, and the magnetic field was applied along the c axis. The μ SR data were analyzed using the MUSRFIT software package [12].

III. RESULTS

A. NMR measurements

In Fig. 1(a), we show examples of ^{75}As NMR spectra at different temperatures. For all temperatures, the spectra can be well described by two Gaussian contributions, a broader peak A at lower frequencies and a sharper peak B at higher frequencies. We attribute these contributions to the inhomogeneous distribution of Ba/K atoms, resulting in potassium-poor and potassium-rich nanoclusters in the sample volume (compare scanning tunneling microscope (STM) measurements in Fig. SF13 in the SM [10]). Despite these nanoscale inhomogeneities, the individual intensities of peaks A and B [Fig. 1(b)] yield the same temperature dependence. Moreover, we observe the same temperature dependence for the spin-lattice relaxation rates of peaks A and B, presented in Fig. 2(b). These observations therefore indicate a rather homogeneous superconducting state throughout the sample volume, despite the inhomogeneous Ba/K distribution.

Below T_c , we observe a sharp decrease in the normalized intensity [Fig. 1(b)], which reflects the increased RF shielding in the superconducting state. Upon closer inspection, the intensity is already slightly reduced at $T^* > T_c$. We can interpret this as RF shielding, stemming from the appearance of uncondensed Cooper pairs, which also results in consistent behavior of various physical quantities which are presented below.

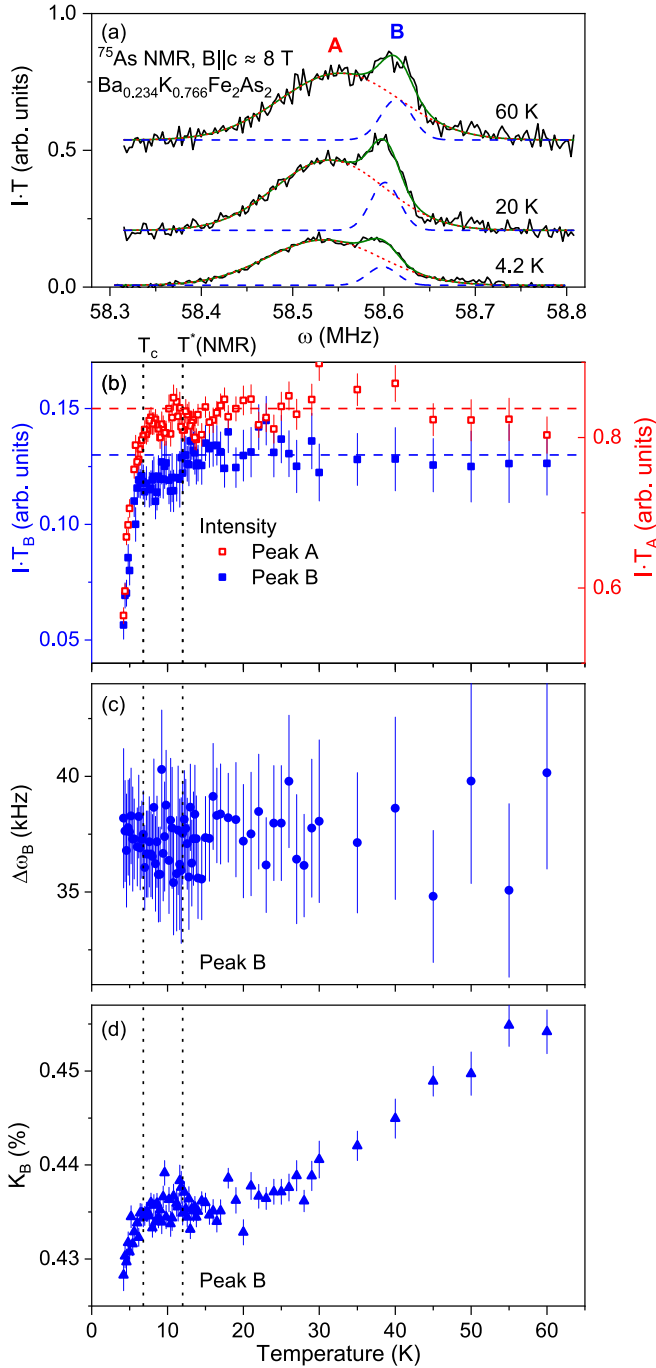


FIG. 1. (a) ^{75}As NMR spectra (black lines) compared to a fit with a double-Gaussian function (green lines) consisting of two peaks, A and B (red dotted and blue dashed lines, respectively), at 4.2, 20, and 60 K (the reference frequency is $\omega_0 = 58.35$ MHz). (b) Temperature dependence of the normalized intensity (red open squares and blue solid squares for peaks A and B, respectively). The values are normalized to the maximum intensity of the total line at 60 K. The dashed horizontal lines are guides to the eye, indicating the expected metallic behavior. Temperature dependence of (c) the spectral linewidth and (d) the Knight shift of peak B. The dotted vertical lines in (b)–(d) indicate T_c and $T^*(\text{NMR})$.

The spectrum further provides evidence against static magnetic order since the line shape remains unchanged across T^* and $T_c^{Z_2}$. This absence of quasi-static magnetism is reflected

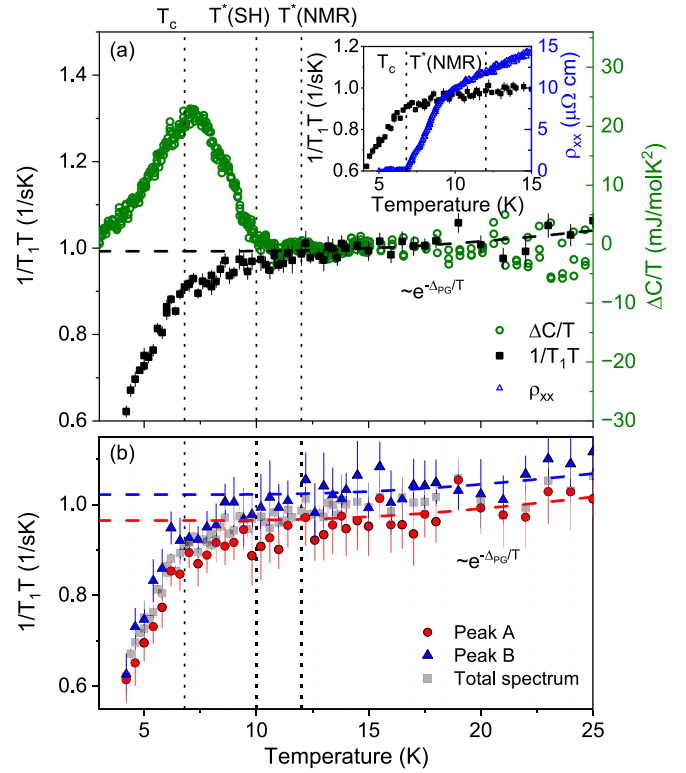


FIG. 2. (a) Temperature dependence of the ^{75}As nuclear spin-lattice relaxation rate $1/T_1T$ (black rectangles) and the specific heat $\Delta C/T$ with the subtracted normal-state behavior (green circles) at 8 T. The dotted vertical lines mark the critical temperatures T_c , $T^*(\text{SH})$, and $T^*(\text{NMR})$, where $T_c^{Z_2}$ is expected to be slightly above T_c . The inset shows the temperature dependence of $1/T_1T$ compared with the resistivity (blue triangles) at 8 T. (b) Temperature dependence of $1/T_1T$ of the individual contributions A (red circles) and B (blue triangles) in the ^{75}As NMR spectrum compared to $1/T_1T$ of the total spectrum (squares). The dashed lines in (a) and (b) are fits to the data above 12 K to a high-temperature pseudogap behavior [$1/T_1T = A \exp(-\Delta_{PG}/T) + C$], with $\Delta_{PG} \approx 70$ K.

more clearly in the temperature-dependent linewidth $\Delta\omega$ of the individual spectral contributions [Fig. 1(c)], where any change in the underlying distribution of hyperfine fields would be reflected in a change in $\Delta\omega$. In Figs. 1(c) and 1(d), we show the results for the sharper peak B, while the data for the broader peak A are presented in the SM in Fig. SF7 [10]. This absence of line deformations is in stark contrast to signatures of magnetic order, such as spin-density-wave order at low Co- and K-doping levels of the BaFe_2As_2 system, where a line broadening of the order of several megahertz was reported [13–15].

In Fig. 1(d), we show the temperature dependence of the NMR Knight shift for peak B. At high temperatures, the Knight shift changes monotonously, contrary to expectations for a simple metal, which would follow the Korringa relation ($K_s^2 T_1 T = \text{const}$) [16]. Grafe *et al.* reported similar behavior for another iron-based superconductor, $\text{LaFeAsO}_{0.9}\text{F}_{0.1}$, and associated it with gapped behavior, comparable to that observed in cuprates [17]. Below T_c , the Knight shift drops, consistent with the expectations for a singlet-pairing state. Between T^* and T_c , the Knight shift shows a slight slope change,

suggesting a reduction of the density of states (DOS) due to the emergence of prepairing fluctuations. However, the broad NMR linewidth prevents precise resolution of this feature. To gain further insight into the Knight shift in this temperature regime, we employ TF- μ SR, which we present below.

We show the temperature-dependent ^{75}As nuclear spin-lattice relaxation rate of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, with $x = 0.766(1)$, at 8 T in Fig. 2(a) and compare it to the temperature dependence of the specific heat and electrical resistivity [inset of Fig. 2(a)] measured at the same field. Above 12 K, $1/T_1T$ has a temperature dependence reminiscent of a pseudogap, which was also reported for other iron-based superconductors [18,19] (see Fig. SF9 in the SM [10] for data above 25 K). In contrast to these other materials, where $1/T_1T$ flattens out at temperatures approaching T_c , our measurements show a deviation from such a behavior, and instead, we observe a decrease below $T^*(\text{NMR}) \approx 12$ K, which is far above T_c . This trend continues down to $T_c = 6.8$ K, where the resistance becomes zero [see the inset in Fig. 2(a)], and we observe a sharp drop in $1/T_1T$ without the appearance of a Hebel-Slichter peak, which is a commonly observed behavior for iron-based superconductors [20–22].

At temperatures below $T^*(\text{NMR})$, there are no signs of a spin-fluctuation-driven enhancement of $1/T_1T$, and instead, the data point towards a further suppression of the density of states, which is consistent with the appearance of pairing fluctuations below $T^*(\text{NMR})$. We exclude inhomogeneous nucleation of superconductivity as the origin of the decrease below $T^*(\text{NMR})$, following the arguments about the spectral intensity above and our theoretical analysis of an inhomogeneous $s_{\pm}$ superconductor below. Further, we analyzed the relaxation of components A and B separately and found that they show qualitatively the same temperature dependence [Fig. 2(b)]. $1/T_1T$ of both peaks shows a sharp decrease below T_c as well as a deviation from the high-temperature pseudogap behavior below $T^*(\text{NMR})$. Component B has a faster relaxation than component A, which is consistent with the higher frequency, and is a result of the stronger corresponding hyperfine coupling. The appearance of pairing fluctuations above T_c is also consistent with the temperature dependence of the specific heat, which starts to increase at $T^*(\text{SH})$, between T_c and $T^*(\text{NMR})$, with an onset temperature about 2 K below $T^*(\text{NMR})$.

B. μ SR measurements

Recent TF- μ SR measurements of another unconventional superconductor, Sr_2RuO_4 , showed that the μ SR and NMR Knight shift show consistent behavior, but μ SR offers higher sensitivity to changes in the magnetic susceptibility [23]. Therefore, to obtain additional insight into the static susceptibility of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ above T_c , we performed TF- μ SR Knight-shift measurements at about 8 T. Furthermore, the different form factors of the μ SR and NMR measurements help exclude a hypothetical cancellation of local internal fields, which would reduce the experimental sensitivity to a magnetic transition.

We note that the previously performed zero-field (ZF) μ SR measurements [2] are quite distinct from the TF- μ SR measurements since without an applied magnetic field, the

ZF signal is insensitive to anomalies that do not break time-reversal symmetry. Therefore, the TF data provide additional information that was missing in Refs. [1,2].

We performed TF- μ SR measurements at 8 T on a stack of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ single crystals with $x = 0.75(2)$. Note that the μ SR sample has a lower doping level than the single crystal used for the NMR measurements. The choice of the sample is related to a technical difficulty in growing enough crystals for μ SR measurements with doping close to $x \sim 0.8$. Therefore, the μ SR and NMR data cannot be compared directly, but $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ at $x \approx 0.75$ is still in the quadrupling state with reduced splitting between T_c and $T_c^{\mathbb{Z}_2}$ and a narrower temperature range with pairing fluctuations [7].

Several examples of fast-Fourier-transformed high-field μ SR spectra, measured at different temperatures, are shown in Fig. 3(a). The spectra consist of three peaks. Like in previous observations for FeSe [24], there are two muon-stopping sites, I and II, with I being a more intense peak. In addition, peak II overlaps with the Ag-holder background, which makes the analysis of its temperature dependence less reliable. Therefore, we focused on the analysis of peak I relative to the Ag peak, which, within the experimental error bars, exhibits a temperature-independent Knight shift [24].

To obtain the Knight shift, we described the spectra using three $A_i \cos(\omega_i t + \phi) \exp(-\lambda_i t)$ components, where A_i corresponds to the muon fractions stopping in the Ag holder and at sites I and II in the sample. We determined the A_i values at the base temperature, where the splitting between the Ag and II peaks is the most prominent, and fixed them for all other temperatures. Further details of the analysis can be found in Ref. [24]. The resulting temperature dependence of the Knight shift, defined as $K_\mu = (B_I - B_{\text{Ag}})/B_{\text{Ag}}$, is plotted in the inset of Fig. 3(b), where B_I is the temperature-dependent position of peak I and B_{Ag} is the position of the Ag peak. The Knight shift was not corrected for the demagnetizing effect, which should be only marginal at this field.

In the normal state, the Knight shift is negative, similar to that measured for FeSe [24]. The overall temperature dependence is consistent with the normal-state susceptibility, showing a broad minimum at around 100 K for this doping level [2]. Figure 3(b) shows the μ SR Knight shift at temperatures approaching the superconducting transition. Here, the negative Knight shift reduces linearly towards T_c , without any sign of Curie-Weiss behavior, as would be expected in the case of local magnetic moments. The Knight shift deviates from the linear, normal-state behavior at $T^*(\mu\text{SR}) \sim 10$ K, which is about 2 K above $T_c \sim 8$ K, measured by AC susceptibility at 8 T. The Knight shift shows a second kink at T_c , similar to the behavior found for the NMR Knight shift [Fig. 1(d)]. The monotonous reduction of the Knight shift at low temperatures consistently indicates the absence of local magnetic moments and magnetic phase transitions in $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ at K doping $x \sim 0.8$, in accord with the NMR data.

IV. DISCUSSION

To distinguish whether possible prepairing fluctuations or a spatially inhomogeneous distribution of T_c are the cause of the observed behavior below T^* , we investigated the Ginzburg-Landau model of an inhomogeneous

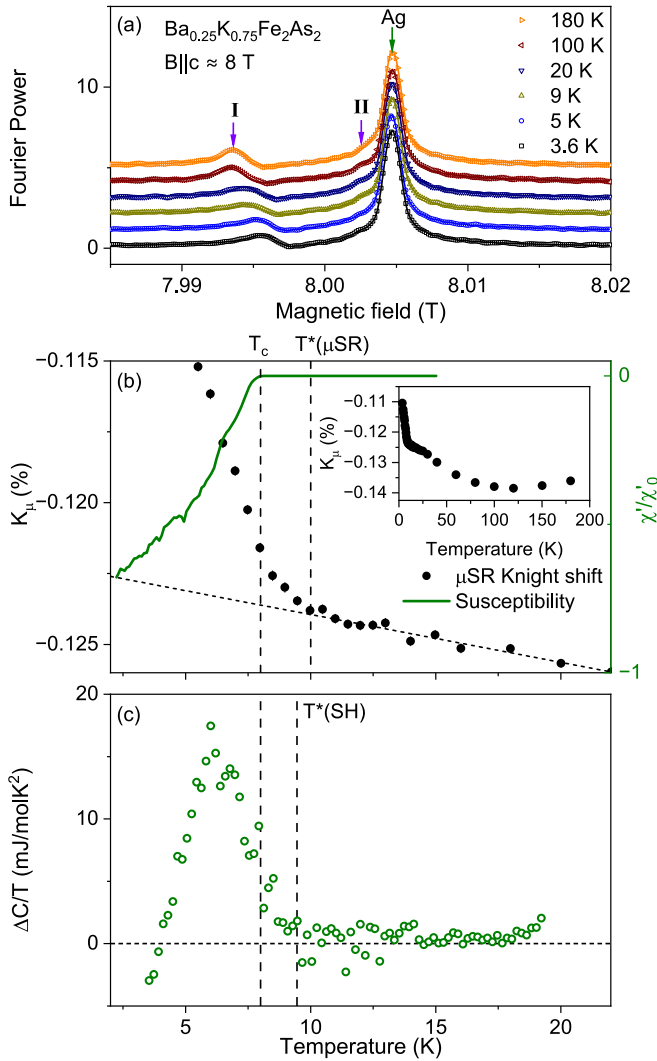


FIG. 3. (a) Fourier transform of TF-μSR time spectra, measured at about 8 T and selected temperatures for $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, with $x = 0.75(2)$. The arrows indicate different components of the signal: I and II correspond to muons stopping at two different sites in the sample, and Ag denotes the signal from muons stopping in the silver sample holder. (b) μSR Knight-shift data close to the superconducting transition measured at about 8 T, applied along the c axis. The negative Knight shift (left axis) already becomes less negative somewhat above T_c , indicating the formation of Cooper pairs above the superconducting transition. We determined T_c via AC-susceptibility measurements at 8 T using an AC field of $10 \mu\text{T}$ at a frequency of 417 Hz. The data are normalized by χ_0 , measured at 2 K and in zero field (right axis). The inset shows the μSR Knight shift at temperatures up to 200 K. (c) Temperature dependence of the specific heat of the μSR sample at 8 T. The onset of the specific heat at $T^*(\text{SH})$ is consistent with $T^*(\mu\text{SR})$.

$s_{\pm}$ superconductor. In Fig. 4, we show a comparison between our measurements for $x = 0.75$ and $0.766(1)$ and the Ginzburg-Landau calculations. This model has a non-BTRS ground state and is usually used to describe doping levels below 70% potassium doping. Further, this model was proposed to describe $1/T_1T$ in iron-based superconductors [25], which enables a direct comparison between

the predicted behavior of non-fluctuating inhomogeneous superconductivity and our experimental data. In order to maintain focus on effects stemming from a distribution of T_c and to avoid complexities associated with a larger parameter space, we refrained from using the $s + is$ model.

In the disorder model one can achieve a split between the onset of the specific heat (caused by local nucleation of pairing) and the magnetic susceptibility by choosing the magnetic field penetration length much larger than the size of nucleation regions. However, at the same time it is not able to reproduce the behavior of the spin-lattice relaxation rate and the μSR-Knight shift [Figs. 4(a) and 4(d) and 4(c) and 4(f)]. Figure 4(a) shows that the diamagnetic onset coincides with zero resistivity. In contrast, our numerical calculations show that, in the mean-field model with disorder, the diamagnetism already begins to develop when the resistivity is still finite and dropping [Fig. 4(d)]. The spin-lattice relaxation rate $1/T_1T$ [Figs. 4(b) and 4(e)] is also qualitatively different. Our experimental data show a change in the slope around the temperature where the resistivity drops to zero (extremum of the specific heat). However, our calculations based on the mean-field disorder model, predict a smooth change, which begins near the temperature where the specific heat starts to deviate from its normal-state behavior.

Our experimental and calculated μSR-Knight-shift results [Figs. 4(c) and 4(f)] also differ from each other. According to our calculations based on the mean-field disorder model, the μSR Knight shift should have the lowest sensitivity to the superconducting transition. However, our experimental results [Fig. 4(c)] show approximately the same onset temperature for the μSR Knight shift and the specific heat. Additionally, the experimental μSR Knight shift is approximately linear with temperature below the superconducting transition and should be curved according to the calculations based on the mean-field disorder model.

The comparison of these calculations with our NMR and μSR measurements, together with the specific-heat and transport data, shows that these experimental data cannot be explained by a disordered non-fluctuating superconductor. On the other hand, the appearance of a large temperature range above T_c where pairing fluctuations exist can favorably account for our experimental observations.

According to the microscopic theory of electron quadrupling [26], the BTRS quartic state can be interpreted as spontaneous symmetry breaking between the numbers of uncondensed Cooper pairs with $s + is$ symmetry versus the time-reversed counterpart with $s - is$. This implies that for this state to span a substantial temperature range, the system needs to have strong pair fluctuations prior to any symmetry-breaking phase transition: $T_c < T_c^{\mathbb{Z}_2} < T^*$.

In the model presented in Ref. [26], T^* can be defined to be approximately equal to T_c^{BCS} , which is the critical temperature in the BCS model that ignores fluctuations. A model that takes into account pairing fluctuations yields a lower T_c , while below $T^* = T_c^{\text{BCS}}$, fluctuation effects increase significantly. However, at T^* , no actual phase transition occurs. Therefore, since one deals with a crossover with no singularities at T^* , it cannot be defined exactly either in experiment or from theory. The crossover at T^* appears gradually, and hence, even above T^* , pairing fluctuations exist. Moreover,

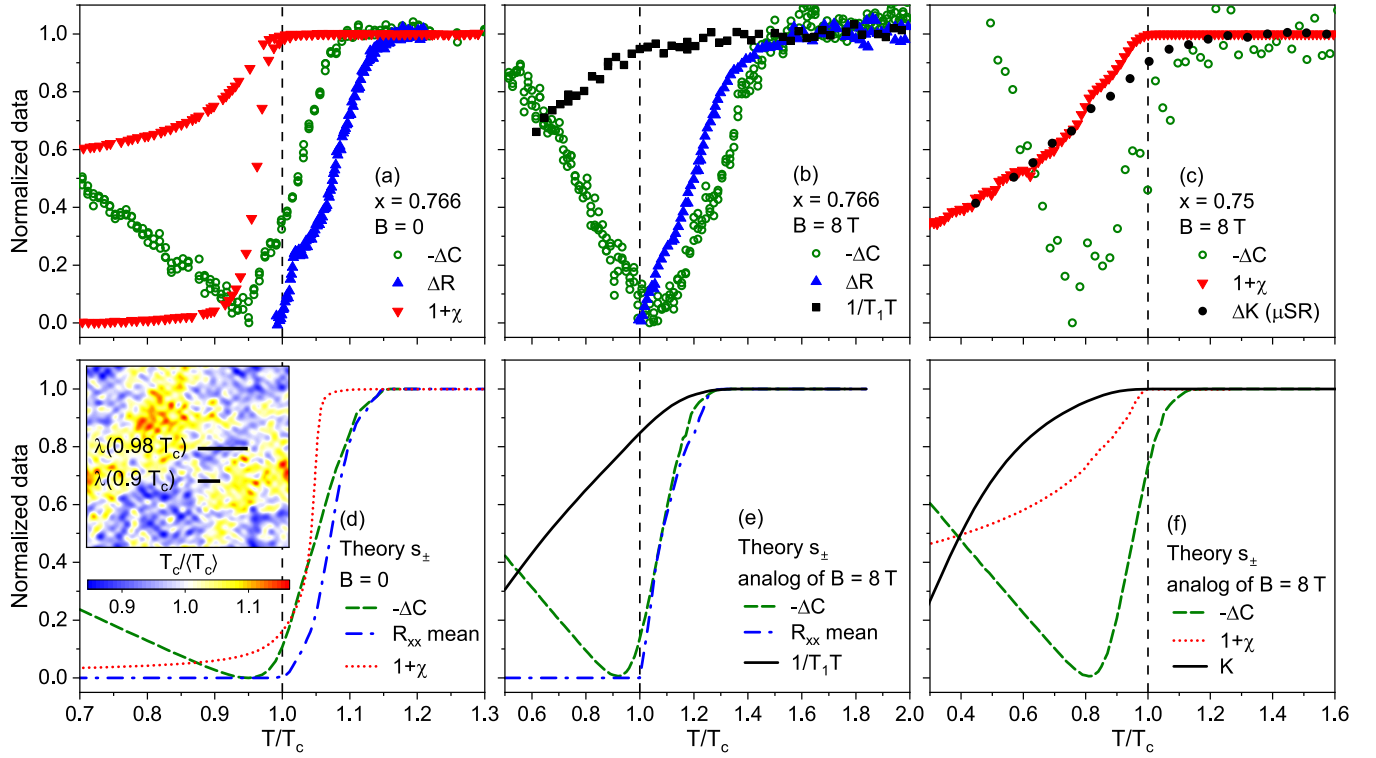


FIG. 4. Comparison of (a)–(c) normalized experimental data to (d)–(f) theoretical predictions of the two-component $s_{\pm}$ Ginzburg-Landau model. Shown are data for the negative specific-heat difference $-\Delta C$, background-subtracted resistivity ΔR , AC susceptibility $1 + \chi$, spin-lattice relaxation rate $1/T_1T$, and μ SR Knight shift K . The inset in (d) shows the spatial distribution of the local critical temperatures assumed in the Ginzburg-Landau calculations. The average magnetic field penetration depth λ is shown for scale in the inset. When the disorder size is much smaller than λ , all observables have identical critical temperatures. The disorder amplitude is chosen to result in a superconducting transition width in the specific heat according to our experimental data. The critical temperature T_c for the whole sample is defined by zero resistivity and by the onset of diamagnetism when resistivity data are not available [(c) and (f)]. For easier comparison of the experimental data to predictions from theory, we subtracted suitable backgrounds, scaled the data, and shifted them along the vertical axes such that for $T \rightarrow 0$ or for T around T_c the value is 0, while for $T \gg T_c$ all observables are 1. We note that subtracting the background is somewhat ambiguous and introduces additional errors, yet the qualitative behavior does not change.

different probes have different levels of sensitivity to pairing fluctuations. Hence, one should consistently interpret T^* as an approximate crossover temperature.

The Knight shift and the spin-lattice relaxation rate $1/T_1T$ probe the DOS and the integrated DOS squared, respectively, at an energy scale on the order of the temperature. When comparing data for the DOS and specific heat from our theoretical work [26], we find that the latter typically has a less pronounced tail above T^* . This leads to estimates of T^* from NMR and μ SR being somewhat higher than $T^*(\text{SH})$ (see Figs. 2 and 3). On the other hand, the resistivity measurements yield less statistical noise; however, corrections due to fluctuations are larger (compared to $1/T_1T$) due to additional leading contributions such as Aslamazov-Larkin terms [27,28]. Because of these factors, the tail above T^* is clearly visible, so $T^*(R)$, defined as the temperature at which a deviation from the background is visible, gives an overestimate of T^* .

We note that our $1/T_1T$ data for the $x = 0.766(1)$ sample fit very well into the systematic behavior of the NMR data reported for $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ with different K-doping levels [10,29]. However, pairing fluctuations at temperatures

approaching T_c have not been reported yet and could easily be overlooked due to insufficient data-point density, which shows the importance of detailed measurements. Also, our systematic transport data indicate that the pairing fluctuations are most pronounced at the K-doping levels for which the BTRS state was previously detected by means of ZF- μ SR experiments [2,6]. Specifically, we observed that $T^*(R)$ as determined by resistivity measurements (see Fig. 14 in the SM [10]) is significantly closer to T_c for reference samples without BTRS superconductivity [6]. This result is also in line with the smaller splitting between T_c and $T^*(\mu\text{SR})$ observed for the sample with $x = 0.75(2)$ [Fig. 3(b)].

V. CONCLUSION

In conclusion, our detailed ^{75}As NMR and μ SR study of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, with $x = 0.766(1)$ and $x = 0.75(2)$, respectively, rules out the presence of magnetic order, which could be associated with a BTRS state. Instead, we found a monotonous decrease of $1/T_1T$ below the crossover temperature T^* , well above $T_c^{\text{Z}_2}$ and T_c . The comparison of our

NMR data with specific-heat, transport, and μ SR Knight-shift data indicates the presence of pairing fluctuations well above T_c , in accordance with the microscopic theory of electron quadrupling [26].

Further, our theoretical analysis of disorder in a non-fluctuating two-band $s_{\pm}$ superconductor cannot simultaneously account for a split of T^* and T_c , and the observed effects in the NMR and μ SR data. These effects are a decrease in $1/T_1T$ below T^* with a pronounced slope change below T_c and a decrease in the μ SR Knight shift already below T^* . From the theory arguments and from a separate analysis of the two contributions to the NMR spectrum, we can exclude an inhomogeneous nucleation of superconductivity as the origin of these features and instead conclude that pairing fluctuations appear at T^* homogeneously across the whole sample volume.

It is worth mentioning that a similar pseudogaplike behavior [30,31] and BTRS superconductivity [32] were reported recently as well for the Fe(Se,S,Te) system. This calls for investigations into whether an electron-quadrupling state may also exist at certain doping levels in this system. A promising direction is to search for quartic states in low-dimensional systems and monolayers of BTRS superconductors, in which the fluctuations are enhanced by reduced dimensionality. In particular, a pseudogap temperature exceeding twice the resistive T_c was reported in single-layer FeSe/SrTiO₃ [33]. Another realization of the quadrupling phase is expected on the 1×1 potassium-terminated surface of KFe₂As₂ [5]. STM data indicate that this surface is a two-dimensional (2D) superconductor with broken time-reversal symmetry. Further studies are needed to investigate the temperature range of pairing fluctuations and to search for a quadrupling phase in this novel 2D superconductor.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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