Magnetic and superconducting transitions in $Ba_{1-x}K_xFe_2As_2$ studied by specific heat

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We report on specific-heat measurements in $Ba_{1-x}K_xFe_2As_2$ ($x \leq 0.6$). For the underdoped sample with $x=0.2$, both the spin-density-wave transition at $T=100$ K and the superconducting transition at 23 K can be identified. After subtraction of the normal-state specific heat by appropriate Debye and Einstein contributions and the normal-state Sommerfeld coefficient, we can describe the electronic contribution to the specific heat in the superconducting state for concentrations $0.3 \leq x \leq 0.6$ by a full single gap within the BCS limit.

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Since the discovery of superconductivity in Fe-based pnictides,[1,2] the superconducting (SC) transition temperature could be raised from $T_c=27$ K to a maximum of 55 K in the so-called 1111-systems RFeAsO (R=La-Gd).[3] Subsequently, further classes of SC compounds containing FeAs layers were reported: the 122-systems AFe_2As_2 with A=Ba, Sr, Ca, Eu and a T_c up to 38 K,[4–7] the 111-compounds LiFeAs and NaFeAs,[8,9] and the binary chalcogenide systems such as $Fe_{1+x}Se$.[10–12] Recently, compounds where the superconducting FeAs layers are separated by conducting transition-metal-oxide blocks were reported with $T_c=37$ K.[13–15] In the first two material classes, the mother parent compounds undergo a transition from a poorly metallic state with tetragonal symmetry to a spin-density-wave (SDW) low-temperature state with orthorhombic distortions.[16–19] The SC state in the 122 compounds can be reached, e.g., by substituting the A-site ions by K (Refs. 4 and 5) or doping the FeAs layers with Co.[6,7]

One exciting and controversial issue is the competition or coexistence of magnetism and superconductivity in the underdoped concentration regimes and the relation to the structural distortions. While in the 1111-systems magnetism seems to be completely suppressed before superconductivity appears,[20] in $Ba_{1-x}K_xFe_2As_2$ the coexistence of orthorhombic distortions and superconductivity has been reported up to $x \approx 0.2$ ($T_c \approx 26$ K),[17,18] and the persistence of long-range antiferromagnetic ordering up to $x=0.3$.[21] Several local-probe studies such as μSR found evidence for a phase separation into SC and antiferromagnetic domains.[22–24] Recent neutron-scattering studies on lightly Co-doped $BaFe_2As_2$ revealed that the structural and antiferromagnetic transition do not occur concomitantly for $x>0$ and that for $x=0.047$ both an atomic force microscopy and a SC transition are present.[25,26]

The symmetry of the SC order parameter is another important question under discussion: a growing number of theoretical and experimental studies seem to be consistent with theoretically suggested $s^{\pm}$-wave model[27,28] with a sign reversal between the two Fermi surfaces. However, the existence of line nodes has not been clarified yet. For example, nuclear magnetic-resonance or thermal-conductivity studies report both evidence for a fully gapped state[29,30] and for nodal lines.[31–34] angle resolved photoemission spectroscopy (ARPES) measurements revealed again nearly isotropic and nodeless gaps.[35–38] This controversy seems to be related to the effects of doping,[39] which is in agreement with the findings in $Ba_{1-x}K_xFe_2As_2$, where a fully gapped state seems to exist for $x \simeq 0.3$,[30] but KFe_2As_2 reportedly exhibits line nodes.[34,40]

Specific-heat measurements provide clear signatures of the antiferromagnetic and SC phase transitions and are sensitive to the symmetry of the SC order parameter, which determines the electronic part of the specific heat below T_c. The specific heat of several samples with a nominal composition in the vicinity of the optimal doping $x=0.4$ have been reported previously[16,18,41–43] and it became clear that a major difficulty consists in modeling the normal-state contribution to the specific heat.[44] Hence, most studies focused on the evaluation of the jump in C/T at the superconducting transition.[16,18,41,42,44] Mu *et al.*[43] extracted the electronic specific heat in the SC state and fitted the data using s-wave isotropic BCS theory with a single gap $\Delta=6$ meV in agreement with the low-energy gaps reported by other experimental techniques such as optical spectroscopy and ARPES (see e.g., Ref. 45 and references therein).

In this work we report on the specific heat of polycrystalline $Ba_{1-x}K_xFe_2As_2$ for $x \leq 0.6$. A part of the data was presented without a detailed analysis already in Ref. 18. We model the normal-state contribution of the specific heat to access the anomalies at the SC and magnetic-phase transitions. For $x=0.2$ we observe a strongly broadened SDW anomaly and a superconducting transition at about 23 K. Concerning the symmetry of the SC order parameter we find that the electronic part of the specific heat in the SC state around the optimal doping $x=0.4$ is well described by using a single gap.

Polycrystalline samples of $Ba_{1-x}K_xFe_2As_2$ with $x=0.0$, 0.1, 0.2, 0.3, 0.4, 0.5, and 0.6 were prepared and characterized by x-ray powder-diffraction and resistivity measurements as described in Refs. 17 and 18. The magnetic properties were studied using a commercial superconducting quantum interference device magnetometer (Quantum Design MPMS-5) with external magnetic fields up to 50 kOe.

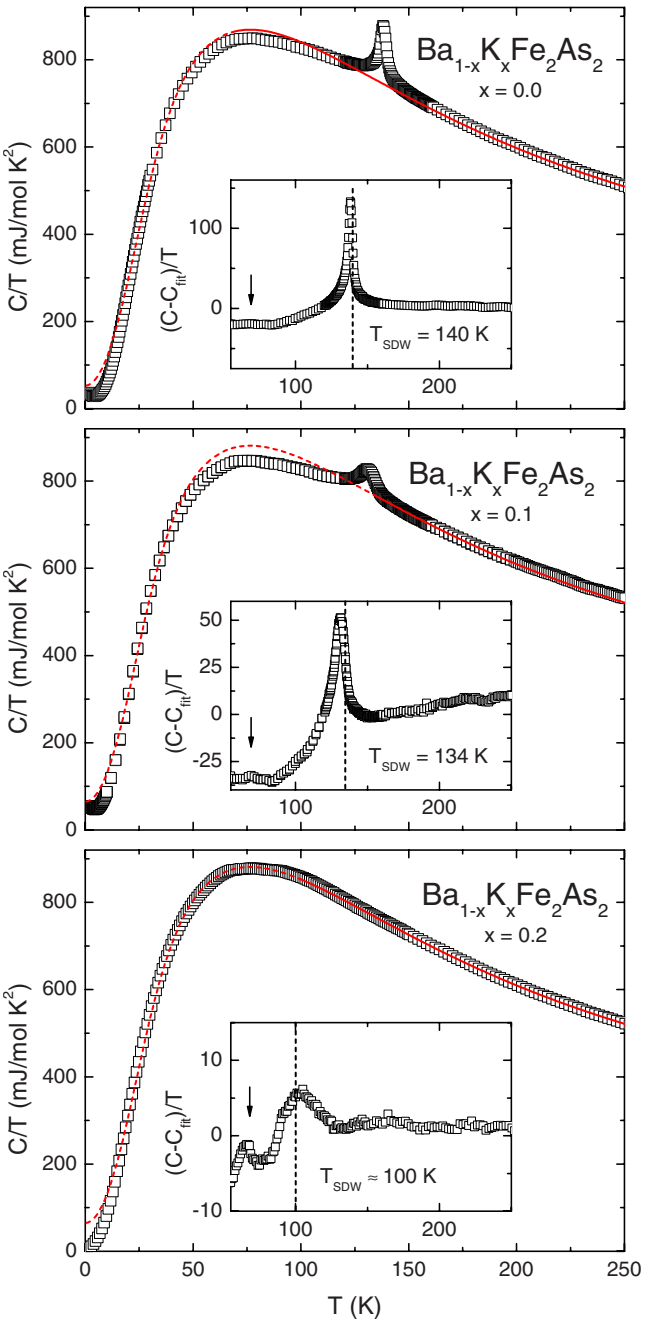

FIG. 1. (Color online) C/T vs T of $Ba_{1-x}K_xFe_2As_2$ for $x \leq 0.2$. Solid lines are fits to the high-temperature data (see text). Extrapolations below T_c are indicated by dashed lines. Insets: residual heat after subtracting the model from the experimental data. Arrows indicate contributions assigned to FeAs.

The heat capacity was measured in a Quantum Design physical properties measurement system for temperatures from $2\ \mathrm{K} < T < 300\ \mathrm{K}$.

The main frames of Fig. 1 show the specific heat divided by temperature C/T of the samples with $x \leq 0.2$. Anomalies attributed to the reported SDW transition can be identified at 140 and 134 K for $x=0.0$ and $x=0.1$, respectively. In the case of $x=0.2$, however, only an extremely broadened and weak feature at around 100 K can be presumed, although a structural transition was clearly identified to occur in this temperature range.[18] Similarly, susceptibility measurements showed bulk superconductivity in this sample below 23 K while no evident feature was discernible in C/T at this temperature.[18] To reveal the SDW and superconducting transitions, we model the specific heat above the corresponding SDW and superconducting transitions by

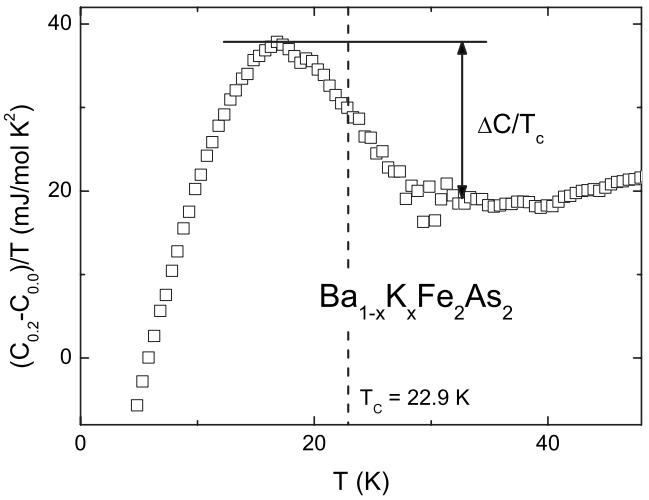

FIG. 2. Difference in the specific heat for $x=0.2$ and $x=0.0$ divided by temperature.

$$C(T) = D(\theta_D, T) + 2E(\theta_{E1}, T) + 2E(\theta_{E2}, T) + \gamma T, \qquad (1)$$

where D and E denote isotropic Debye and Einstein contributions with the corresponding Debye and Einstein temperatures θ_D, θ_{E1}, and θ_{E2} and γ is the Sommerfeld coefficient. The ratios were kept fixed for all investigated samples to comply with the lattice degrees of freedom. We started out with the sample with $x=0.3$ because at this concentration no structural or magnetic transitions seem to occur and the system remains tetragonal down to lowest temperatures.[18] The parameters obtained for $Ba_{0.7}K_{0.3}Fe_2As_2$ were then slightly adapted to describe the specific heat in the tetragonal phase for the samples with $x \leq 0.2$. The resulting curves reproduce the data nicely in the tetragonal phase and are shown as solid lines in Fig. 1 for $T \geq T_{\mathrm{SDW}}$ and the extrapolations for $T < T_{\mathrm{SDW}}$ are shown as dashed lines. The obtained fit parameters are listed in Table I.

The data after subtraction of the modeled normal-state contribution $(C - C_{fit})/T$ is shown in the corresponding insets of Fig. 1. Immediately, one recognizes the sharp SDW transitions for $x=0.0$ and $x=0.1$. For $x=0.2$ a strongly broadened anomaly at around 100 K now becomes clearly visible, in agreement with the observed SDW anomalies in the resistivity and the structural transition.[18] The weak features visible at about 70 K and present in all three samples are probably traces of binary FeAs, which reportedly orders helimagnetically in this temperature range.[46] Hence, subtracting the modeled normal-state contribution allows to reveal the SDW transition anomalies but the validity of this modeling is naturally limited by the electronic reconstruction, the possible contributions of magnetic excitations, and the structural changes occurring at the SDW transition.

Therefore, we follow a different route to uncover the SC

TABLE I. Fit parameters for the specific heat of $Ba_{1-x}K_xFe_2As_2$ at various doping levels using Eq. (1).

x	θ_D/K	θ_{E1}/K	θ_{E2}/K	$\gamma/\frac{\text{mJ}}{\text{mol K}^2}$	$\frac{S}{T}\vert_{T_c}/\frac{\text{mJ}}{\text{mol K}^2}$
0.0	144	183	365	53	
0.1	144	183	365	65	
0.2	144	183	365	65	
0.3	144	183	365	53	52.9
0.4	144	185	378	49	48.2
0.5	145	186	374	54	56.8
0.6	155	188	359	58	61.3

transition in the $x=0.2$ sample which was revealed by susceptibility measurements to occur at about 23 K.[18] We used the data for the pure $BaFe_2As_2$ as a reference for a system in the SDW state but without superconductivity at low temperatures. Moreover, the normal-state parameters for both compounds are very similar. The resulting low-temperature electronic specific heat is shown in Fig. 2. A clear anomaly with a midpoint temperature of $T_c=22.9$ K and a jump $\Delta C/T_c=19.2$ mJ/mol K^2 is in good agreement with the SC transition temperature determined from susceptibility measurements.[18] We would like to emphasize that both the SDW transition and the SC anomaly are very broad and indicate a distribution of the corresponding transition temperatures, which may be a result of chemical inhomogeneity within the Ba/K layers[18] or the presence of strong magnetic and SC fluctuations similarly to the results reported for Co doped $BaFe_2As_2$.[25,26]

Having discussed the data for the samples which exhibit a SDW transition, we now turn to the samples where magnetic ordering and orthorhombic distortions are absent and the structure remains tetragonal down to lowest temperatures: the main frames in Fig. 3 show C/T vs T of $Ba_{1-x}K_xFe_2As_2$ for $x=0.3$, 0.4, 0.5, and 0.6. The transitions into the superconducting states are clearly visible as peaks in the experimental data for all four compounds. Again, the lines are the modeled electronic and lattice contributions of the normal state according to Eq. (1) using the parameters given in Table I. The model reproduces the data above T_c nicely.

The electronic part obtained by subtracting the modeled normal-state contribution is shown in the corresponding insets. The residual electronic specific heat C_{el} can be well described by the BCS derived α model[47,48]

$$C_{el} = T\frac{\mathrm{d}S_{el}}{\mathrm{d}T}, \tag{2}$$

$$S_{el} = -\frac{6\gamma}{\pi^2 k_B}\int_0^\infty \mathrm{d}\epsilon[f\ln f + (1-f)\ln(1-f)]. \tag{3}$$

Here, the ratio of the energy gap Δ_0 at 0 K and T_c is not fixed ($2\Delta_0/k_BT_c=3.53$ in the BCS theory) but left as a free fitting parameter. The Fermi-Dirac distribution $f=f(E,T)=[\exp(E/k_BT)+1]^{-1}$ is determined by the energy $E=\sqrt{\epsilon^2+\Delta^2(T)}$, where ϵ denotes the energy of independent fermion quasiparticles measured relative to the Fermi surface. The temperature dependence of the gap $\Delta(T)=\Delta_0\delta(T/T_c)$ is assumed to be the same as calculated by Mühlschlegel,[49] where δ is the normalized BCS gap in the limit of weak-coupling superconductors. The obtained fit curves are shown as solid lines in the insets of Fig. 3. The discrepancies of the fit and the data at lowest temperatures might originate from chemical disorder and inhomogeneity in the samples.

For the Sommerfeld coefficient γ we fixed the value to the parameters given in Table I for the normal state. This procedure is justified by the good agreement of our γ values with the values obtained from the electronic entropy $S=\int_0^T d\vartheta(C-C_{latt}^{fit})$ divided by temperature at the superconducting transition $S/T|_{T_c}$ given in Table I. Moreover, our normal-state Sommerfeld coefficient is in agreement with the recent result of $\gamma\approx 47$ mJ/mol K^2 for a sample with $x=0.3$ by Storey *et al.*[50] The above model leads to the fit parameters presented in Table II. As expected the optimally doped sample with $x=0.4$ exhibits the highest values for $T_c=37.3$ K and $\Delta_0=78$ K. The critical temperatures for the compounds with $x=0.3$ and $x=0.5$ are close to this optimal value but the zero-temperature gaps are already reduced (Fig. 4). The coupling parameters $2\Delta_0/T_c$ are in good agreement with the BCS value of 3.53. For the sample with $x=0.4$, the ratio $2\Delta_0/T_c=4.14$ is enhanced signaling stronger coupling (see inset of Fig. 4). The corresponding gap value $\Delta_0=6.7$ meV is in good agreement with the 6 meV derived by Mu *et al.* from specific heat and corresponds to the small-gap value observed also by ARPES, optical spectroscopy, and other techniques (for an overview see, e.g., Ref. 45 and references therein).

Naturally, one can expect that the existence of two or more superconducting gaps as evidenced by ARPES should show up in the electronic part of the specific heat, and it was suggested that the transport properties with $s^{\pm}$ symmetry can be considered the same as for a conventional two-gap superconductor.[27] Hence, we also tried to describe our data

TABLE II. Parameters determining the electronic specific heat for $Ba_{1-x}K_xFe_2As_2$ according to the model described in the text.

	$x=0.3$	$x=0.4$	$x=0.5$	$x=0.6$
T_c/K	36.5	37.3	36.0	29.7
Δ_0/K	68	78	66	53
$2\Delta_0/k_BT_c$	3.73	4.14	3.67	3.57

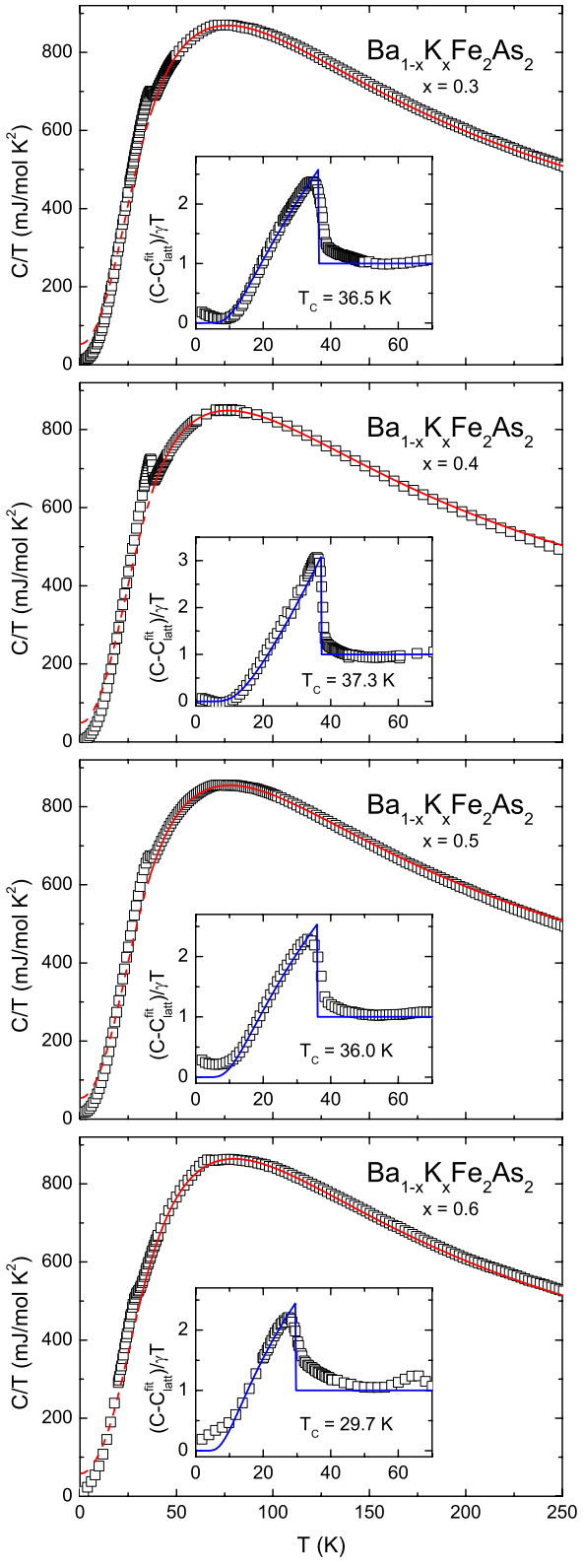

FIG. 3. (Color online) Temperature dependence of C/T of $Ba_{1-x}K_xFe_2As_2$ for $x=0.3$, 0.4, 0.5, and 0.6. The lines in the main frames are fits using Eq. (1) in the normal conducting state. Extrapolation of the fits to lower temperature is dashed. The insets show the difference between C/T and the modeled phonon contribution normalized by the normal-state Sommerfeld coefficient γ together with a fit according to Eqs. (2) and (3).

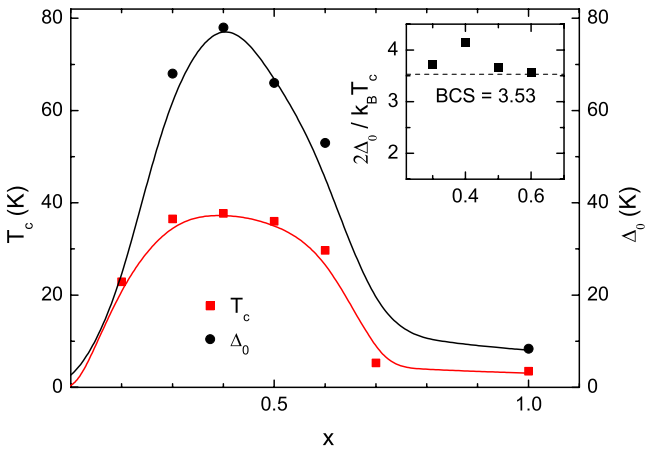

FIG. 4. (Color online) Critical temperatures and SC gaps plotted vs doping level x. Lines are drawn to guide the eye. Data for $x=1.0$ is taken from Ref. 40 and the value of T_c for $x=0.7$ is taken from Ref. 17. Inset: coupling parameters $2\Delta_0/T_c$ vs x compared to the BCS value of 3.53.

using two different gaps Δ_i with the same temperature dependence and corresponding Sommerfeld coefficients γ_i $(i=1,2)$ for the case of $x=0.4$. However, the obtained single-gap fit is already very good and we find that the additional fit parameters seem to overdetermine the available experimental data hindering a free convergence of all fit parameters. Therefore, we fixed the gap parameters $\Delta_1=70$ K and $\Delta_2=139$ K using the values determined by ARPES measurements[38] and varied only the Sommerfeld coefficients γ_i. The resulting best-fit curve with $\gamma_1=39.8$ and $\gamma_2=9.2$ mJ/mol K^2 is compared to the single-gap fit in Fig. 5. We believe that the two-gap model is not superior to the single gap in the sense that the electronic specific heat is dominated by the low-energy excitation across the smaller gap. A theoretical treatment of the thermodynamic properties of pnictides by Benfatto *et al.* in a multiband approach

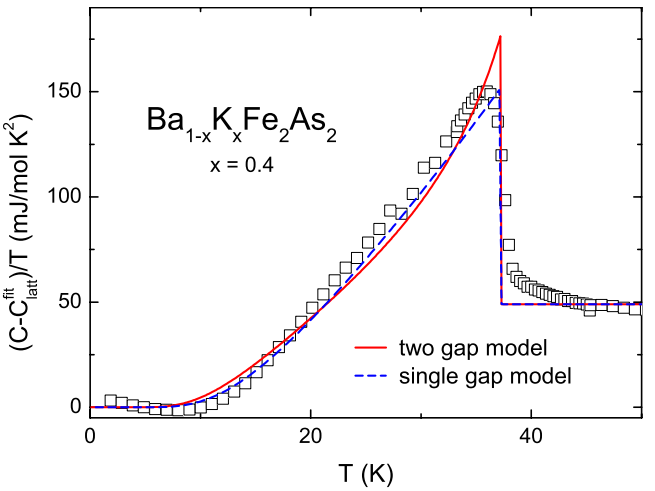

FIG. 5. (Color online) Specific heat of $Ba_{0.6}K_{0.4}Fe_2As_2$ divided by temperature after subtraction of the phononic contributions. Solid line is a fit utilizing a sum of two BCS-like terms while the dashed line is the fit according to the single-gap model.

showed that, although intermediate coupling strengths are necessary, the specific heat does not deviate significantly from the weak-coupling single-band BCS behavior.[51] As a consequence, however, a gap ratio close to the single-gap BCS $2\Delta_0/T_c=3.53$ does not reflect the weak-coupling scenario anymore. Finally, we want to mention that our data of $\Delta C/T_c$ are in agreement with the universal behavior of this quantity $\propto T_c^2$ suggested in Ref. 44.

In summary, our specific-heat measurements revealed the existence of two phase transitions for the sample with $x=0.2$, a broad transition at about 100 K associated with magnetic ordering and a broad anomaly at transition to the superconducting ground state at 23 K. We found that the phonon and electronic contribution in the normal state are almost independent of the concentration. For the superconducting samples with $0.3\leq x\leq 0.6$ the normal state γ is in agreement with the average value $S/T|_{T_c}$ in the superconducting sate. The electronic part of the specific heat of $Ba_{1-x}K_xFe_2As_2$ below T_c is well described by the phenomenological α model for a *s*-type superconductor with a single full gap and coupling parameters $2\Delta_0/T_c$ close to the weak-coupling BCS limit.

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