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The Carrier Specific Heat of $\text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$

F. Y. Wei,¹ B. Lv,¹ F. Chen,¹ Y. Y. Xue,¹ and C. W. Chu^{1,2}

¹Department of Physics and TCSUH, University of Houston, Houston, Texas 77204-5002, USA

²Lawrence Berkeley National Laboratory, 1 Cyclotron Road, Berkeley, California 94720, USA

ABSTRACT

The carrier contribution to the specific heat coefficient, C_e/T , of $\text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ with $0 \leq x \leq 1$ has been determined. The C_e/T at the optimal doping level appears to be T-independent above the superconducting transition temperature T_c . Systematic reductions and increases with cooling below 100 K, on the other hand, characterize the underdoped and overdoped samples, respectively. As the result, the low-T limit of the normal-state C_e/T increases almost linearly with x by a factor of three over $0 \leq x \leq 1$. However, the C_e/T suppression across the magnetostructure transition of the underdoped samples is actually negligibly small. In particular, the extracted $\gamma_H - \gamma_L$ at $x = 0$, which should include all suppression effects, is comparable to or even smaller than that of $x = 0.15$, where γ_H and γ_L are the high- and low-T limits of the normal-state C_e/T . Therefore, it appears that, while the magnetostructure transition still plays a role, it may not be the main factor behind the C_e/T evolution.

The normal-state electronic structure is one of the key issues in the investigations of the FeAs-based compounds. While various local density approximation (LDA) calculations show considerable differences in the details, they consistently suggest a quasi 2D Fermi surface made of three hole-pockets around the Γ point and two electron-cylinders around the M point.¹⁻⁴ On the experimental side, the angle resolved photoemission spectroscopy (ARPES) and quantum dHvA oscillation measurements not only confirm the suggested Fermi-surface topology, but also reveal some discrepancies on moment-dependent energy shifts, mass enhancements and band splits, indications of significant residual carrier interactions.⁵⁻⁸ Various magnetic excitations have also been observed. In particular, a static spin density wave (SDW) transition and a nearby structure transition on the underdoped side have been regarded as the key factors behind the metal-superconductor evolution.^{6,9,10}

How such evolution is related to macroscopic observations, *e.g.* the carrier part of the normal-state specific heat coefficient, C_e/T , is an interesting issue. The C_e/T observed has been used to support the band-structure calculation from the very beginning.¹⁻³ The band calculations suggest an x-insensitive C_e/T in $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, but the change of its low-T limit, γ_L , with doping is later attributed to the suppression of DOS by the combined SDW-structure transitions around T_{SDW} .^{3,11} Whether this magnetostructure transition opens a significant gap is a debatable issue. For example, the data demonstrate that the entropy suppression at T_{SDW} is rather small for the similar SrFe_2As_2 .¹² While optical data suggest large gaps, *i.e.* at the order of $2k_B T_{\text{SDW}}$, open at the Fermi surface, the ARPES data either report no noticeable gap or a much more complicated Fermi surface reconstruction.^{6,13} Recent investigations on the overdoped KFe_2As_2 further suggest that the more general interband interactions also play a significant role in its C_e/T .^{7,14} A 20 meV

Fermi-level shift in $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ from $x = 0.4$ to $x = 1$, for example, doubles the carrier mass. Calculations on a similar compound of LaFePO further reveal that the mass enhancement is mainly caused by the interband interactions between the e -pockets and the p -pockets, *i.e.* sensitive to the Fermi-level shifts, which shrink (expand) the e -pockets (p -pockets) with the K doping.¹⁵ The even larger 130-180 meV shift reported from $x = 0$ to 0.45,⁸ therefore, might also play a role. The roles of the magnetostructure transition and the mass enhancement will be an important issue in understanding the specific-heat data. Here we report our C_e/T measurements on $\text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ with $0 \leq x \leq 1$ up to 300 K. The extracted γ_L increases with x continuously and monotonically without any anomaly around the optimal doping level $x \approx 0.45$. Such smooth evolution across $x = 0.45$ may suggest similar mechanisms over a broad doping range. The high-T limit γ_H of the C_e/T , on the other hand, shows a peak at $x = 0.15$ and decreases with x monotonically over higher doping levels. It is especially interesting to note that the difference $(\gamma_H - \gamma_L)$, presumably the C_e/T suppression associated with the magnetostructure transition, at $x = 0$ is comparable to or lower than that at $x = 0.15$. These are rather different from the simplified SDW suppression model. In addition, the C_e/T of all samples appear to share a common temperature dependency: being T-independent above 100 K (except the overlapped SDW peaks), but dropping (enhancing) by the amount of $(\gamma_H - \gamma_L)$ with further cooling. Such trends suggest that the DOS suppression of the underdoped samples may not merely be the result of the magnetostructure transition.

Ceramic $\text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ samples with $x = 0, 0.15, 0.2, 0.25, 0.3, 0.45, 0.7, 0.8$ and 1 were synthesized from high temperature reactions of stoichiometric high-purity Sr, K, Fe, and As, as previously reported.¹⁶ Two samples were independently synthesized and tested at each x value of

0, 0.3, 0.45 and 0.7 to verify the reproducibility. The X-ray diffraction of the polycrystalline samples indicates single phase Sr122-like structure with impurity phases of less than a few percent. The specific heat was measured using a Quantum Design Physical Properties Measurement System (PPMS) over the temperature range of 1.8 and 300 K. Ceramic samples of about 6-8 mg were placed on the platform with Apiezon N-grease. The random fluctuations were verified through repeated measurements and presented as error bars when they are larger than the symbol size in the figures. Another main uncertainty is the sample mass, which strongly affects the high-T limit, γ_H , of C_e/T . Care has been taken to limit this influence to below 1 mJ/mol K².

While the estimation of the carrier contribution $C_e(T)/T$ is strongly model-dependent, both its high-T limit, γ_H , and the low-T limit, γ_L , can be objectively obtained. Actually, it is observed that the γ_L value of low- T_c superconductors can be robustly extracted due to the simple reason that the carrier contributions to the entropy S and the specific heat C are rather different. The corresponding fractions, *i.e.* $\approx \gamma_L / (\gamma_L + \beta T_c^2 / 3)$ and $\gamma_L / (\gamma_L + \beta T_c^2)$ at T_c , respectively, may differ by a factor of three, where β is the expansion coefficient of the phonon part $C_{ph}/T = \beta T^2 + \dots$. Deviations from the C_{ph}/T modeling, therefore, will be significantly suppressed if the entropy-conservation constraint is applied. Different models, for example, lead only to a few mJ/mol K² difference in the γ_L of LiFeAs.¹⁷ On the high-T side, both the Debye (Einstein) function $C_v \approx 3NR[1 - (T_D/T)^2/12 + \dots]$ and the correction term $\gamma^{an} = (C_p - C_v)/T \approx VB\beta^2$ associated with the anharmonic phonon/dilation are model insensitive, where C_v (C_p), N , T_D and R are the specific heats under constant volume (pressure), the number of atoms, the Debye temperature and the gas constant, respectively.^{11,18} The $(C_p - 15R)/T$ observed above 100 K, therefore, are plotted against $1/T^3$ for $0 \leq x \leq 1$ (symbols in Fig. 1). For a better view, vertical shifts are used. Thin reference

lines of the smoothed $x=0.45$ data are also added for each plot to clarify the doping dependency. Several characteristics are noticeable even at such rough scales: 1) The plots are linear above 150-200 K with the deviations less than the experimental uncertainty of a few mJ/mol K² (thick line for the $x = 0.45$ data in Fig. 1). This is expected from the Debye (Einstein) function above $\theta_D/2$ with the extracted Debye temperature θ_D around 300-400 K. The extraction of the intercepts, therefore, is straightforward. 2) The intercept, *i.e.* $\gamma_H + (C_p - C_v)/T$, systematically decreases with x : the symbols are above the $x = 0.45$ reference line for the underdoped samples but below for overdoped samples. The splits, $|\gamma_H(x) - \gamma_H(0.45)|$, can be as large as 20 mJ/mol K² and should be independent of the models used. 3) The symbols outside the SDW peaks are parallel to the reference line above 150 K for $x \leq 0.7$, suggesting that the members may share not only a common phonon baseline C_{ph}/T but also a similar T-dependence of the C_e/T there. A similar conclusion, *i.e.* the C_e/T is T-independent above 150 K, has also been reached in Ba_{1-x}K_xFe₂As₂ with $x \leq 0.3$.¹¹ 4) Crossovers between the symbols and the reference lines occur for $x \geq 0.7$, suggesting a higher γ_L but lower γ_H for the overdoped samples.

Fig. 1. $(C_p - 15R)/T$ vs. T^{-3} . The data (symbols) for $\text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ at $x=0, 0.15, 0.2, 0.25, 0.3, 0.45, 0.7, 0.8$ and 1 (from top to bottom). Vertical shifts are used for a better view. Smoothed $x = 0.45$ data (thin solid line) are also attached to each plot to show the x -dependence. The thick line at $x = 0.45$ is a linear fit.

To further explore the issue, γ_H is deduced from the intercepts using the same $300 \text{ K } \gamma^{\text{an}} = V B \beta^2 \approx 12 \text{ mJ/mol K}^2$ of $(\text{Ba,K})122$ (Table 1; inset, Fig. 2).¹¹ The difference of the molecular volumes of BaFe_2As_2 and SrFe_2As_2 is less than 10% (*i.e.* with a net effect less than 1 mJ/mol K^2), and the two series share a common member of KFe_2As_2 . The data spread between the independently synthesized samples further demonstrates repeatability better than 5 mJ/mol K^2 (the overlapped data at $x = 0.45$ and 0.7 cannot even be distinguished in the scale used). The deduced γ_L is also

included with comparable repeatability. Both γ_L and γ_H show strong doping dependence, although the two vary with x in opposite directions (Table 1). The result is that $\gamma_L \gg \gamma_H$ in the overdoped sample but $\gamma_L \ll \gamma_H$ in the underdoped samples. Strong T-dependence of the C_e/T is therefore expected except around $x = 0.45$. This shows that the x -independent DOS suggested by LDA calculations may be an oversimplified scenario.

Table 1. The parameters extracted

(the errors are the statistical fluctuations of several independently synthesized samples)

x	γ_L mJ/molK ²	γ_H mJ/mol K ²	γ_0 mJ/mol K ²
0	12(2)	42(2)	—
0.15	17	54	—
0.2	22	49	—
0.25	20	42	12
0.3	24(2)	34(2)	4(2)
0.45	34(1)	29	1.5(1.5)
0.7	51(4)	18(2)	23(5)
0.8	73	8	33
1	67	0	—

Some γ_L data of $\text{Sr}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ have been reported previously, *i.e.* 6.5 and 69.1 at $x = 0$ and 1, respectively.^{14,19} Both are in rough agreement with the observation here. Unfortunately, there are no reported γ_H data to the best of our knowledge. Many investigations, however, have been done

for similar $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ (*e.g.* Refs. 11 and 20). Most of the data are in line with the trend in the inset of Fig. 2.

Fig. 2. The C_e/T of the $x = 0.45$ sample (see text). (circles): from the fit of an $x = 0.45$ sample (left scale). The mark “A” indicates the kink of the possible two-gap feature (see text). Grey crosses: from the fit of an $x = 0$ sample (right scale). Inset: γ_L (open triangles) and γ_H (solid circles) as the functions of the doping level x . Thin lines are simply guides for the eye.

Two features appear to be interesting. First, both γ_L and γ_H appear to be smooth and monotonic functions of x except for the γ_H below $x = 0.2$. In particular, there is no noticeable anomaly around the crossover point $x \approx 0.45$. The same mechanism that dominates the underdoped region, therefore, seems to extend into the overdoped region. Second, the difference $\gamma_H - \gamma_L$ and the low- T limit γ_L change with x in rather different ways below 0.2. The $\gamma_H - \gamma_L = 30 \text{ mJ/mol K}^2$ at $x = 0$ is even smaller than that of 37 mJ/mol K^2 at 0.15 (or comparable considering the possible data uncertainties). This is rather different from simple SDW suppression, which expects an x -insensitive γ_H and a $\gamma_H - \gamma_L$ that decreases with x .

Following the empirical algebra proposed by P. J. Baker et al.,²¹ the C_p data of the $x = 0.45$ sample were fit as $\gamma T + A_D F_D(T/T_D) + A_E F_E(T/T_E) + \gamma^{an} \cdot [C_v(T)/C_v(300K)]^2$, where γ , A_D , A_E , F_D , F_E , T_E and $\gamma^{an} = 12 \text{ mJ/mol K}^2$ are the Sommerfeld coefficient, two amplitudes, the Debye function, the Einstein function, the Einstein temperature and the anharmonic term at 300 K, respectively. The γ value is fixed to $(\gamma_H + \gamma_L)/2 \approx 35 \text{ mJ/mol K}^2$ for the above analysis. The fitting parameters $T_D = 205 \text{ K}$ and $T_E = 358 \text{ K}$ lead to a good agreement. The deviations are far less than 1% over whole temperature range between T_c and 300 K. The calculated $A_D F_D(T/T_D) + A_E F_E(T/T_E) + \gamma^{an} \cdot [C_v(T)/C_v(300K)]^2$ was then used as the phonon background. The carrier part C_e/T so deduced is almost T -independent, as expected from the $\gamma_H \approx \gamma_L$ observed (Fig. 2). It is interesting to note that a similar T -independent C_e/T also occurs in cuprates around the optimal doping level.¹⁸ This may not be a pure coincidence, considering the competitive excitations are likely to be absent at this doping level. No quantitative analysis of the superconductive gaps was attempted. Both the ratio $\Delta C_e/\gamma_L T_c \approx 3$ and the obvious kink, marked as **A** in the figure, around $6-8 \text{ K} \approx 0.2 T_c$ strongly suggest a two-gap structure similar to that of LiFeAs.¹⁷

The same phonon baseline was then applied for all other samples. Bumps/dips up to 10 mJ/mol K² occasionally appear. Trim procedures, which alternate the phonon spectrum, have been previously proposed to cancel such irregularities.^{11,18} However, such procedures may also unintentionally change the intrinsic trends. We would argue, however, that systematic trends are still clear without such trims (Fig. 3). To further verify the procedure adopted, the same fitting procedure was also used for other doping levels. The result of an $x = 0$ sample, for example, is also shown in Fig. 2 as grey crosses. The obtained $C_e/T \approx 10$ mJ/mol K² below 50 K is in good agreement with that based on the phonon background of the $x = 0.45$ sample, suggesting that there is no significant shift of soft-phonons with doping. Significant T-dependency, however, appears at higher T, in line with above γ_L - γ_H deduced.

Fig. 3. a) The carrier contribution C_e/T for underdoped samples with $x = 0$ (solid circles), 0.15 (open triangles) and 0.3 (open inverted triangles). b) The carrier contribution C_e/T for overdoped samples with $x = 0.7$ (solid squares), 0.8 (open circles) and 1.0 (open inverted triangles). The larger open symbols are the respective γ_L . Inset: raw C_p/T vs $(T/T_c)^2$ for $x = 0.45$ (open circles); $x = 0.7$ (open inverted triangles) and $x = 0.8$ (open squares).

For a clear view, only three data sets at $x = 0, 0.15$ and 0.3 are shown in Fig. 3a. The other four underdoped samples show essentially the same trend. The $x = 0.15$ data is included partly to represent the worst scenario of the bumps/dips without trim. As suggested by the large split between γ_L and γ_H in the inset of Fig. 2, all data with $x \neq 0.45$ show significant T-dependences. It is especially interesting to note that the main C_e/T drop of all underdoped samples occurs below 100 K. The C_e/T drop associated with the magnetostucture transition, however, should occur right below T_{SDW} if the associated gap $\Delta \geq k_B T_{SDW}$, but around $T' \approx \Delta/k_B$ if narrower. Undoped $SrFe_2As_2$ with its higher T_{SDW} of 204 K, therefore, may serve as a good testing ground. The C_e/T of the $x = 0$ sample is roughly the same on both sides of the 204 K peak, however, suggesting that any C_e/T suppression associated with the magnetostucture transition has to be either negligibly small or associated with a gap narrower than $0.5k_B T_{SDW}$ (Fig. 3a). The later scenario, however, is in disagreement with the optical data.¹⁰ The data, therefore, suggest that the magnetostucture transition may not be the direct cause of the γ_L suppression. A similar T-independent C_e/T above 150 K has also been reported for underdoped $Ba_{1-x}K_xFe_2As_2$, although the two systems have very different T_{SDW} ,¹¹ *i.e.* there is no clear correlation between the C_e/T suppression temperature and the T_{SDW} .

The C_e/T over the overdoped range $x \geq 0.7$ appears to be even more unusual (Fig. 3b). Its value is rather small above 150 K but raises quickly with cooling below 100 K. Although residual phonon interferences, *i.e.* an x -dependent C_{ph}/T , cannot be totally ruled out, the extracted γ_L (large open symbols in Fig. 3b) suggests that the dominant part of this low-T enhancement should come from carriers. It is also interesting to note that both the suppressions over $x \leq 0.3$ and the enhancement above $x = 0.7$ occur below a similar temperature threshold. This again suggests a common underlying mechanism.

Both the ARPES and the dHvA oscillation data suggest that the changes of the electric structure from $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ to KFe_2As_2 may be associated with the interaction between the hole pockets and the electron pockets, which are expanding and shrinking with the K-doping, respectively.^{7,14,22} As a result, large mass enhancements up to $18 m_e$ are observed in the C_e/T and the dHvA data. The KFe_2As_2 data in Fig. 3b, therefore, reveal that this renormalization is a rather delicate process: the ten-fold mass-enhancement disappears at a moderate temperature of 100 K $\approx 8 \text{ meV}/k_B$, an energy scale far lower than the observed Fermi level shift of 20 meV; the C_e/T between 120 and 300 K is almost zero. The effect of the proposed e - p interactions on the C_e/T seems to be much more than a T -independent enhancement. On the other hand, the similar trend over $0.7 \leq x \leq 1$ demonstrates that similar mass enhancement at least extends to the whole overdoped range. The similar T -dependency, the smooth x -dependencies of both γ_L and γ_H discussed above may further suggest an important role of such mass enhancement on the undoped side as well.

To further explore the overdoped side, the residual C_p/T at the zero-temperature limit, γ_0 , is estimated for $x = 0.45, 0.7$ and 0.8 (inset, Fig. 3b). The C_p/T vs. $(T/T_c)^2$ plots appear to be linear

between $0.1T_c$ and $0.4T_c$, and the intercept was taken as γ_0 assuming that the superconducting gap is not dominated by components narrower than $0.2k_B T_c$. While the γ_0 of the optimally doped sample is essentially zero, both the overdoped 0.7 and 0.8 samples possess a large value of ≈ 20 - 30 mJ/mol K², *i.e.* $0.4\gamma_L$ or larger. Similar trends can also be found in the literature. The reported $C_p/T \approx 45$ mJ/mol K² of KFe₂As₂ at 0.3 K $\approx 0.1T_c$,¹⁴ for example, is more likely associated with a large γ_0 without a dominating gap component narrower than $0.2k_B T_c$. A scenario of mesoscopic phase-separation is strongly suggested. This should make the situation more complicated.

Smooth second-order transitions, *e.g.* a quantum critical point, would be unlikely to occur.^{23,24}

Such complicated doping dependency is actually expected based on the multiband structure of pnictides. Previous data have already revealed that the orbit-dependent subsystems of pnictides may have comparable strengths but different dispersions (or the psuedogap) at Fermi level.^{10,13,25} Both their relative weight and the effective mass may evolve with doping in complex ways. The C_e/T , on the other hand, is only a weighted average. While both the superconductivity and the magnetostructure transition may be closely related to interband interactions, the data suggest that interpreting C_e/T as a simple SDW-suppression is an oversimplification.

In summary, the carrier C_e/T is deduced from 1.8 to 300 K for Sr_{1-x}K_xFe₂As₂ with $0 \leq x \leq 1$. The C_e/T of the optimally doped $x = 0.45$ sample is T-independent above the T_c , and that of the underdoped samples drops below 70-100 K. The overdoped samples, on the other hand, have a C_e/T which is rather small above 100 K but raises to unusually high values at lower temperatures. Competitive interband interactions are suggested to interpret the data.

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