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Specific heat in strongly hole-doped Iron-based superconductors

Dmitry V. Chichinadze and Andrey V. Chubukov

1School of Physics and Astronomy, University of Minnesota, Minneapolis, MN 55455, USA

We compute specific heat $C(T)$ in a strongly hole-doped Fe-based superconductor, like KFe$_2$As$_2$, which has only hole pockets. We model the electronic structure by a three-orbital/three pocket model with two smaller hole pockets made out of $d_{xy}$ and $d_{xz}$ orbitals and a larger pocket made out of $d_{yz}$ orbital. We use as an input the experimental fact that the mass of $d_{xy}$ fermion is several times heavier than that of $d_{xy}/d_{xz}$ fermions. We argue that the heavy $d_{xy}$ band gives the largest contribution to the specific heat in the normal state, but the superconducting gap on the $d_{yz}$ pocket is much smaller than that on $d_{xy}/d_{xz}$ pockets. We argue that in this situation the jump of $C(T)$ at $T_c$ is determined by $d_{xy}/d_{xz}$ fermions, and the ratio $(C_T - C_n)/C_n$ is a fraction of that in a one-band BCS superconductor. At $T < T_c$, $C(T)$ remains relatively flat down to some $T^*$, below which it rapidly drops. This behavior is consistent with the data for KFe$_2$As$_2$ and related materials. We use one-parameter model for the interactions and fix this only parameter by matching the experimental ratio of the gaps on the two $d_{xy}/d_{xz}$ pockets. We argue that the resulting parameter-free model reproduces quantitatively the data on $C(T)$ for KFe$_2$As$_2$. We further argue that the very existence of a finite $T^* < T_c$ favors $s^+$ gap structure over $d$-wave, because in the latter case $T^*$ would almost vanish.

Introduction. Rich physics of Iron-based superconductors (FeSC) continues to attract strong attention from the condensed-matter community [1–13]. One of the most debated issues in the field is the strength of correlations. On one hand, FeSCs have Fermi surfaces, and most display a metallic, Fermi-liquid like behavior in some temperature range above superconducting $T_c$. On the other, there is a clear distinction between the observed electronic structure and the one obtained by first-principle calculations for free fermions. Some researchers believe that this difference can be accounted for by including the momentum-dependent self-energy [14], which modifies the dispersion but leaves fermions and their collective degrees of freedom fully coherent (this is often termed as “itinerant scenario”, see e.g., Ref. [9][15]). Others argue that at energies relevant to superconductivity and competing orders, fermions can be viewed as correlated yet itinerant, but collective magnetic excitations should be viewed as at least partly localized (a “Hund metal scenario”, see, e.g., Ref. [16][17]). And others further argue [18][19] that electronic excitations should be viewed as itinerant on some Fe-orbitals and as nearly localized on other orbitals (an “orbital selective Mottness” scenario).

From the perspective of Mott physics, the best candidates to display Mott behavior are strongly hole-doped FeSCs, like KFe$_2$As$_2$ [20][25], as for these systems the tendency towards electron localization has been argued to develop at a smaller Hubbard $U$ (Ref. [18][19]). Low-energy fermionic states in KFe$_2$As$_2$ are composed of fermions from three orbitals, $d_{xy}$, $d_{xz}$, and $d_{yz}$, the last two are related by Cs symmetry [23]. Specific heat measurements in KFe$_2$As$_2$ have shown that above superconducting $T_c$, specific heat coefficient $C(T)/T$ scales as $a + bT^2$, as expected in a metal, but $a$ is larger than in other FeSCs [25][30]. Because $a$ is proportional to the sum of the effective masses for different bands, large value of $a$ implies that at least one effective mass is large. Within the Mott scenario, the mass enhancement comes from frequency-dependent self-energy, $\Sigma(\omega)$.

This self-energy narrows the dispersion and simultaneously reduces the quasiparticle residue $Z$, transferring $1 - Z$ spectral weight into Hubbard sub-bands. The effect is believed to be the strongest for the band made of fermions from $d_{xy}$ orbital [18][19]. However, band narrowing and accompanying mass enhancement can be also caused by innocuous reasons like smaller hopping integral for $d_{xy}$ fermions or closeness to a Van-Hove singularity (see [31] and references therein). In the latter case, large value of the specific heat coefficient can be understood already within the itinerant scenario. ARPES data do indeed show [31–33] that the $d_{xy}$ band is more narrow than the bands made by fermions from $d_{xz}$ and $d_{yz}$ orbitals, but Hubbard sub-bands have not been yet detected in KFe$_2$As$_2$. Furthermore, some ARPES data on KFe$_2$As$_2$ and other FeSCs show that $d_{xy}$ excitations are as sharp as excitations from $d_{xz}/d_{yz}$ bands [21][34]. This makes the interpretation of specific heat data above $T_c$ somewhat ambiguous.

In this communication we analyze whether one can separate between Mott and itinerant scenarios by analyzing specific heat data in the superconducting state. Given that $d_{xy}$ fermions have the largest mass, i.e., the largest density of states (DOS), there are four possibilities for system behavior below $T_c$. They are depicted in Fig. [1]. One possibility (panel (a)) is that superconductivity predominantly develops on the heavy $d_{xy}$ orbital because of larger DOS. If this is the case, the system’s behavior is the same as in a one-band superconductor: the specific heat jump at $T_c$, $\delta C/T_c = (C_T - C_n)/C_n$, is of order one, and $C(T)$ varies as a function of a single variable $T/T_c$ below $T_c$. Another (panel (b)) is that superconductivity develops at $T_c$ on $d_{xz}/d_{yz}$ orbitals, but the temperature dependence of $C(T)$ below $T_c$ is still determined by the heavy $d_{xy}$ orbital. In this situation $\delta C/T_c$ is small, but $C(T)$ below $T_c$ is the same as in panel (a). The third possibility (panel (c)) is that not only $(C_T - C_n)/C_n$ at $T_c$ but also the behavior of $C(T)$ in some $T$ range below $T_c$ is determined by $d_{xz}/d_{yz}$ orbitals, while
fermions on the \( d_{xz} \) orbital have smaller gap and can be treated as non-superconducting down to \( T_{xy} < T_c \). In this situation \( (C_s - C_n)/C_n \) is small, \( C(T)/T \) varies slowly between \( T_c \) and \( T_{xy} \) towards a finite value (equal to normal state \( C(T)/T \) for \( d_{zy} \) fermions), and rapidly drops below \( T_{xy} \). And the fourth possibility (panel (d)) is that fermions on the \( d_{xy} \) orbit do not pair down to \( T = 0 \), i.e., \( T_{xy} = 0 \).

The data for KFe\(_2\)As\(_2\) from several groups \cite{25,30} show that (i) the specific heat jump at \( T_c \) is much smaller than the BCS value, (ii) between \( T_c \) and approximately \( T_c/6 \), \( C(T)/T \) decreases rather slowly towards a finite value, (iii) below \( T_c/6 \), \( C(T)/T \) rapidly drops and tends to zero at \( T \to 0 \). This behavior is consistent with the one in Fig. 1(c). We analyze whether this behavior can be understood by just assuming that the \( d_{xy} \) band is heavier than the other two bands (and, hence, the DOS for this band is the largest), or one needs to additionally include the reduction of quasiparticle \( Z \) for the \( d_{xy} \) band. A momentum/frequency independent \( Z \) can be absorbed into the renormalization of the interactions involving \( d_{xy} \) fermions, hence the issue is whether mass/DOS variation between \( d_{xy} \) and \( d_{xz}/d_{yz} \) bands is sufficient to describe the data, or one needs to additionally assume that the interactions involving \( d_{xy} \) fermions are weaker than the ones between \( d_{xz} \) and \( d_{yz} \) fermions. We argue that the difference in the masses is sufficient to describe the observed behavior. Namely, we obtain the behavior in Fig. 1(c) by analyzing the model of three \( \Gamma \)-centered \( d_{xz}/d_{yz} \) and \( d_{xy} \) hole pockets in 2 Fe zone, and invoking mass difference but keeping the interactions on all three orbitals comparable in strength. If \( Z \) on the \( d_{xy} \) orbital is small in KFe\(_2\)As\(_2\), this will additionally reduce the value of \( T_{xy} \).

We note in passing that our theoretical scenario is different from the one presented in Ref. \cite{27} as we do not require that KFe\(_2\)As\(_2\) is close to a magnetic quantum criticality. It is also different from the one in Ref. \cite{26} where the temperature evolution of \( C(T) \) was largely attributed to the gaps on hole barrels near \( (\pi, \pi) \) in 2 Fe zone. We emphasize that the existing ARPES data didn’t detect superconducting gaps on the hole barrels, but did detect the gaps on the three \( \Gamma \)-centered hole pockets which we consider. Several earlier works \cite{28,29} analyzed the behavior of \( C(T) \) in KFe\(_2\)As\(_2\) within the phenomenological two-gap model, constructed in analogy with the two-gap model for MgB\(_2\) (Ref. \cite{39}). Our reasoning is similar to these works in the sense that we have larger gaps on \( d_{xz}/d_{yz} \) pockets and a smaller gap on \( d_{xy} \) pocket. On the other hand, our analysis is based microscopic three-band model, and we reproduce experimental \( C(T) \) with no free parameters.

The model. The electronic structure of KFe\(_2\)As\(_2\) in the physical 2-Fe Brillouin zone consists of 3 hole pockets, located at the \( \Gamma \)-point, and hole barrels near \( (\pi, \pi) \). There is no evidence of superconductivity on the hole barrels, and we neglect them in our analysis. Two inner \( \Gamma \)-centered pockets are made out of fermions from \( d_{xz} \) and \( d_{yz} \) orbitals, and the outer pocket is made out of fermions from \( d_{xy} \) orbital \cite{20}. We take as an input that the \( d_{xz} \) band has larger band mass/DOS than \( d_{xz}/d_{yz} \) bands. We follow earlier works \cite{56,39} and describe superconductivity within the low-energy model with \( H = H_0 + H_{int} \), where the quadratic Hamiltonian \( H_0 \) is given by \( 2 \times 2 \) matrix for \( d_{xz} \) and a separate term for \( d_{yz} \) fermions, and \( H_{int} \) is the Hubbard-Hund interaction, dressed by contributions from high-energy fermions.

To study superconductivity, we convert from orbital to band basis, i.e., diagonalize the quadratic form to \( H_0 = \sum \epsilon_k c_k^\dagger c_k + \epsilon_{d_{xz}} d_{xz}^\dagger d_{xz} + \epsilon_{d_{yz}} f_{xz}^\dagger f_{xz} + \epsilon_{d_{xy}} f_{xy}^\dagger f_{xy} \), where \( c_k \) and \( d_k \) are linear combinations of fermions from \( d_{xz} \) and \( d_{yz} \) orbitals, and \( f_{\alpha} \)-operators describe \( d_{xy} \) fermions. The pairing interaction has s-wave and d-wave components (see Ref. \cite{57} and Supplementary material (SM) for details). We focus first on s-wave superconductivity and discuss d-wave pairing later.
The pairing interaction in $s$-wave channel is

$$H_{SC} = \sum_{k,p,s,s'} \left[ U_{cc} c_{k}^d c_{k-s}^c c_{s-p}^c + U_{dd} d_{k}^d d_{k-s}^d d_{s-p}^d + U_{cd} (c_{k}^d d_{k-s}^d + d_{k}^d c_{k-s}^c) c_{s-p}^c + (U_{fc} f_{k}^d f_{k-s}^d + U_{fd} f_{k}^d f_{k-s}^d) d_{s-p}^d + H.c. \right]$$

where for circular hole pockets, bare interactions are $U_{cc} = U_{dd} = U_{cd} = (U + J)/2$, $U_{ff} = U/2$, and $U_{fc} = U_{fd} = \frac{U}{2}$.

After renormalizations from high-energy fermions, all couplings become different, and, most important, $U_{cd}$ becomes larger than $U_{cc} U_{dd}$ (Refs. [37] [38] [40]). This gives rise to an attraction in the $s^+$ channel.

![Diagram](image)

**FIG. 2.** Diagrammatic expressions for Gorkov’s gap equations. Triangles with different filling represent SC vertices on different bands. Solid, dashed, and dotted lines represent $c, d, f$-fermions respectively. Wavy lines represent interactions between fermions.

**Superconductivity.** Superconducting $T_c$ and $s$-wave gaps on the three Γ-centered hole pockets at $T \leq T_c$ are obtained by solving the set of coupled linearized gap equations, presented in Fig. 2. In analytical form we have

$${\Lambda}_{c} \begin{pmatrix} \Lambda_{c} \\ \Lambda_{d} \end{pmatrix} = - L \begin{pmatrix} v_c U_{cc} & v_d U_{cd} & v_f U_{fc} \\ v_c U_{cd} & v_d U_{dd} & v_f U_{fd} \end{pmatrix} \begin{pmatrix} \Lambda_{c} \\ \Lambda_{d} \end{pmatrix}$$

(2)

where $L = \ln \frac{\Delta}{\Delta_0}$, $\Lambda$ - is the upper cutoff, and $v_c, v_d, \text{ and } v_f$ are densities of states, proportional to the band masses. In our case, $v_c \sim v_d$, and $v_f$ is larger. We present the full solution for the gap in the SM and here show the result for $\tilde{\Lambda} = (\Lambda_c, \Lambda_d, \Lambda_f)^T = (1, \alpha, -\beta)^T$ to leading order in $v_{cd}/v_f$, where $\alpha$ and $\beta$ do not depend on $v_f$ (see SM for exact expressions). The key observation here is that the gap $\Delta_f$ on the $d_{xy}$ pocket is small in the ratio of $v_{cd}/v_f$. This is the consequence of the fact that $s^+$ superconductivity develops on $c$ and $d$ pockets (not to be confused with $s^+$ pairing in systems with both electron and hole pockets), while the gap on the $d_{xy}$ pocket does not exist on its own, but rather is induced by inter-orbital pairing interactions ($\Delta_f$ scales with $U_{fc}, U_{fd}$). Note that $\Delta_f$ is non-zero only when $c$ and $d$ pockets are treated as non-equivalent, otherwise $\alpha = -1$ and $\beta = 0$.

To minimize the number of parameters, below we set $U_{cd}, U_{ff}, U_{fc}, U_{fd}$ equal to their bare values in the Hubbard-Hund model (see above) and use $J = J' = 0.4U$ [39]. Then $U_{cd} = 0.7U, U_{ff} = 0.5U, U_{fc} = U_{fd} = 0.2U$ [10] [41]. We model the renormalization of $U_{cd}^2 - U_{c} U_{dd}$ into a positive variable, necessary for $s^+$ superconductivity, by a single parameter $x$, by setting $U_{cc} = U_{cc}^{bare}(1 - x) = 0.7U(1 - x), U_{dd} = U_{dd}^{bare}(1 + x) = 0.7U(1 + x)$. We used the experimental values $v_d/v_c = 1.33, v_f/v_c = 3.17$ from Ref. [30] and set $x = 0.5$ to match the experimental value of $\alpha \approx -0.4$ (Ref. [20]). The same $x$ gives $\beta v_c/v_f \approx 0.06$, consistent with [20].

**The specific heat.** To calculate $C(T)$, we compute the internal energy $E(T)$ above and below $T_c$, and use $C(T) = dE/dT$. To obtain $E(T)$ we construct a BCS Hamiltonian with anomalous terms with prefactors $\Delta_c, \Delta_d, \text{ and } \Delta_f$, and diagonalize it. This yields

$$E(T) = - \sum_{i = c,d,f} v_i \int d\epsilon \frac{\epsilon_i^2 + \Delta_i^2/2}{\sqrt{\epsilon_i^2 + \Delta_i^2}} \tanh \frac{\sqrt{\epsilon_i^2 + \Delta_i^2}}{2T} + ...$$

(3)

where dots stand for temperature-independent terms. We express $\Delta_d$ and $\Delta_f$ via $\Delta_c$ and $E(T)$ in powers of $\Delta_c$. To first order in $v_{cd}/v_f$ we obtain $E(T) = E(T_c) - (v_c + v_d \alpha^2)\Delta_c^2/2$. The contribution from the $f$ band is small in $v_{cd}/v_f$ despite that the DOS for this band is large. Using $\Delta_c(T) \approx \sqrt{T_c - T}$, we then obtain that the magnitude of the jump of the specific heat at $T_c$ does not depend on $v_f$. The specific heat above $T_c$, on the other hand, comes primarily from the $d_{xy}$ band simply because DOS for this band is the largest. As a result, $\partial C/\partial T \propto v_{cd}/v_f$ is small, unlike in a one-band BCS superconductor, where it is $O(1)$. We present the full expression for $\partial C/\partial T$ in the SM.

To obtain $C(T)$ below $T_c$, we assume, following [12] that the ratios $\Delta_d/\Delta_c$ and $\Delta_f/\Delta_c$ remain the same as near $T_c$, and $\Delta_c(T)$ has the same temperature dependence as in BCS superconductor. We then find from (3) that in the $T$ range where $\Delta_f(T) \approx -(\beta v_c/v_f)\Delta_c(T)$ is smaller than $T$, the contribution to the specific heat from the $d_{xy}$ band remains the same as in the normal state. As the consequence, $C(T)/T$ evolves from its maximal value right below $T_c$ to a finite value equal to the specific heat coefficient from non-superconducting $d_{xy}$ band. This behavior changes below $T = T_{xy}$, at which $\Delta_f(T_{xy}) = T_{xy}$. At such low temperatures the gap on the $d_{xy}$ band cannot be neglected, and the contribution to the specific heat from this band rapidly drops, and, as a result, $C(T)/T$ rapidly drops towards zero value at $T = 0$.

In Fig. 3 we show the result of numerical calculation of
the specific heat coefficient, using experimental values from the DOS’s from Ref. [30]. The behavior is the same as presented schematically in panel (e) of Fig. 1 and agrees quantitatively with the experimental data for KFe$_2$As$_2$ (Refs. [25, 28, 30, 42, 44]). We emphasize that we fixed the only interaction parameter $x$ by matching the measured [20] ratio of $\Delta_d/\Delta_c$, hence our $C(T)$ is obtained with no fitting parameters. We reproduce the experimental location of $T_{xy}$, and the overall behavior of $C(T)$ below $T_c$.

$d$–wave pairing. Some experimental data, most notably on the thermal conductivity [45, 46], have been interpreted as evidence for $d$–wave pairing symmetry in KFe$_2$As$_2$. This is in variance with laser ARPES study [20, 24], whose results were interpreted as evidence for the $s$–wave pairing. Theoretical results show that $s$–wave and $d$–wave pairing components are both attractive and comparable in strength, with RPA calculations [38] favoring $s^+$ superconductivity and early functional RG calculations [47] favoring $d$–wave pairing. By all these reasons, it is instructive to analyze $C(T)$ for $d$–wave pairing.

Within our model of circular pockets, $d$–wave pairing involves only $c$ and $d$– pockets. The $d$–wave component of the pairing interaction is

$$H_{SC} = \sum_{k,p,\uparrow\downarrow} \left[ \tilde{U}_{cc} c_{\uparrow k \sigma}^\dagger c_{\uparrow k \sigma'} c_{\downarrow p \sigma'} + \tilde{U}_{dd} d_{\uparrow k \sigma}^\dagger d_{\downarrow p \sigma'} + c_{\downarrow k \sigma'} d_{\uparrow p \sigma} + H.c. \right] \cos 2\phi_c \cos 2\phi_d,$$

where $\phi_c$ and $\phi_d$ are angles along the Fermi surfaces. At the bare level (i.e., without integrating out high-energy fermions) $\tilde{U}_{cc} = \tilde{U}_{dd} = \tilde{U}_{cd} = (U - J^\prime)/2$. There also exists the sin $2\phi_c$ sin $2\phi_d$ interaction component, but it does not give rise to new physics and we skip it. After renormalization $\tilde{U}_{cc}$ and $\tilde{U}_{dd}$ split, and, most importantly, $\tilde{U}_{cd}$ becomes larger than $\tilde{U}_{cc} \tilde{U}_{dd}$ [48]. Like for the $s^+$ case, the enhancement of the inter-pocket pairing interaction gives rise to an attraction and a non-zero $T_c$ for $d$–wave pairing. The matrix equation for the $d$–wave gap is

$$\begin{pmatrix} \Delta_c \\ \Delta_d \end{pmatrix} = \frac{-L}{2} \begin{pmatrix} v_c \tilde{U}_{cc} - v_d \tilde{U}_{cd} \\ -v_c \tilde{U}_{cd} - v_d \tilde{U}_{dd} \end{pmatrix} \begin{pmatrix} \Delta_c \\ \Delta_d \end{pmatrix}. \tag{5}$$

Evaluating the eigenfunctions, substituting them into the expression for the internal energy $E(T)$, and differentiating over $T$, we obtain the behavior as in panel (d) of Fig. 1. Namely, the jump $\delta C/C_n$ at $T_c$ is small, and $C(T)/T$ below $T_c$ drops but tends to a finite value at $T = 0$, equal to $C(T)/T$ for non-superconducting $d$–band. This does not agree with the data, which clearly show that $C(T)/T$ drops below $T_{xy} < T_c$. This result holds for arbitrary $C_t$–symmetric dispersion, as long as the interaction in the orbital basis is local, and the larger hole pocket can be approximated as pure $d$–band. By all accounts (see e.g., Ref. [12]), the admixture of $d_{xc}/d_{yz}$ orbital states to the composition of this pocket is very small (a percent), so $T_{xy}$, even if finite, should be truly small.

Conclusions. In this paper we studied the specific heat of KFe$_2$As$_2$. We argued that $C(T)$ in the normal state is chiefly determined by the heavy $d$–band pocket, however superconductivity predominantly involves $d_{xc}/d_{yc}$ pockets, while the gap on the $d_{xy}$ pocket is either induced, but is small (for $s$–wave pairing), or not induced at all (for $d$–wave pairing). This gives rise to the behavior when (i) the jump of $C(T)$ at $T_c$ is much smaller than the BCS value, and (ii) below $T_c$ specific heat coefficient $C(T)/T$ initially evolves towards a finite value, equal to normal state contribution from $d_{xy}$ band. For $s$–wave pairing, $C(T)/T$ eventually drops below a certain $T_{xy}$ (Figs. 1(c) and 3). If the pairing is $d$–wave, $T_{xy} = 0$ in our analysis, and is likely quite small in a more general case. The experimentally detected behavior of $C(T)/T$ (Refs. [25, 28, 30, 42, 43]) is more consistent with $s$–wave pairing. We used the detuning of interactions on $d_{xc}$ and $d_{yc}$ pockets from their bare values as a single adjustable parameter to reproduce the data on gap ratio on the two small pockets [20]. After that, our theory has no free parameters. It reproduces the magnitude of the jump at $T_c$, the shape of $C(T)/T$ below $T_c$, and the value of $T_{xy}$. We emphasize that we did not assume that interactions involving $d_{yz}$ fermions are additionally reduced due to potentially small quasiparticle residue $Z$ for fermions on the $d_{yz}$ band. The reduction of $Z_{xy}$ under hole doping follows from quite solid theoretical arguments [17, 18], what is less clear is whether the reduction is strong enough to affect $C(T)$. If it is, the overall behavior of $C(T)/T$ will not change compared to our analysis, but $T_{xy}$ will decrease further compared to $T_c$. A systematic study of $C(T)$ in doped K$_{1-x}$Ba$_x$Fe$_2$As$_2$ is needed to determine the influence of $Z_{xy}$ on the specific heat.
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