Gap structure in the electron-doped iron–arsenide superconductor Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$: low-temperature specific heat study

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Abstract. In this paper, we report the field and temperature dependence of low-temperature specific heat down to 400 mK and in magnetic fields up to 9 T of the electron-doped Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$ superconductor. Using the phonon specific heat obtained from pure BaFe$_2$As$_2$, we found a normal state Sommerfeld coefficient of 18 mJ mol$^{-1}$ K$^{-2}$ and a condensation energy of 1.27 J mol$^{-1}$. The temperature dependence of electronic specific heat clearly indicates the presence of low-energy excitations in the system. The magnetic field variation of field-induced specific heat cannot be described by single clean s- or d-wave models. Rather, the data require an anisotropic gap scenario that may or may not have nodes. We discuss the implications of these results.

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1. Introduction

The recent discovery of superconductivity in Fe-based pnictides $RFeAsO$ [1, 2] ($R$—rare earth) has created a new era in superconductivity research and has stimulated great interest in these compounds [3]. Subsequently, other types of superconducting materials containing FeAs layers were discovered, such as binary chalcogenides $Fe_{1+x}Se$ [4, 5], so-called ‘111’ compounds [6, 7] LiFeAs or NaFeAs, and 122 systems $AFe_2As_2$, where $A$ is an alkaline earth [8]–[10]. $BaFe_2As_2$, a prototypical member of the latter family, crystallizes with tetragonal ThCr$_2$Si$_2$-structure type, and at ambient pressure exhibits structural and spin-density-wave (SDW) transitions at about 140 K [11]. Suppression of the SDW state by either applied pressure [12] or chemical doping [8, 10] results in superconductivity. Despite a large theoretical and experimental effort (see [13, 14]) to understand the nature of superconductivity in these materials, there are still many open questions that have not been resolved, such as the pairing mechanism and the symmetry of the order parameter. Moreover, the experimental results reported so far are often contradictory, ranging from nodal to fully gapped isotropic superconductivity. Even within the Co-doped $BaFe_2As_2$ family, the situation is unclear. While surface-sensitive measurements such as ARPES (angle resolved photoemission spectroscopy) [15] and STM (scanning tunnelling microscopy) [16] claim fairly isotropic gap values, penetration depth [17], $\mu$SR (muon spin rotation) [18], NMR (nuclear magnetic resonance) [19], thermal conductivity [20]–[22], specific heat [23] and Raman scattering [24] argue that an anisotropic gap is necessary, although the details vary between these measurements as well. Often, the superconducting gap structure is discussed in terms of the $s\pm$ model, with sign reversal of the order parameter between different Fermi surface sheets [25]–[27]. However, the large sample, family and doping dependence may favor scenarios where the low-energy excitations, possibly nodal, depend strongly on the particular sample being studied and the probe used to investigate them (e.g. [28]–[30]).

In this paper, we present the results of our detailed studies on the specific heat of the electron-doped $Ba(Fe_{0.92}Co_{0.08})_2As_2$ superconductor. By subtracting the lattice contribution (obtained from measurements on non-superconducting samples), we can extract the full electronic $T$-dependence. The temperature- and magnetic field-dependent data imply an anisotropic gap structure.

2. Experimental details

Large single crystals of $Ba(Fe_{0.92}Co_{0.08})_2As_2$ were grown from FeAs flux with typical sizes of about $2 \times 1.5 \times 0.2$ mm$^3$. The samples crystallize as well-formed plates, with the [001]-direction perpendicular to the plane of the crystals. The doping level was determined by microprobe analysis. More details about the synthesis and characterization of the samples can be found in [9]. Based on electrical resistivity measurements, $T_c$ was established to be 20 K (zero resistance), in agreement with the heat capacity results presented here. Heat capacity was measured down to 400 mK and in magnetic fields up to 9 T using a thermal relaxation method implemented in a Quantum Design PPMS-9 device. All data measured in the field were field cooled.

3. Results and discussion

The temperature dependence of the specific heat of $Ba(Fe_{0.92}Co_{0.08})_2As_2$ is shown in figure 1. As can be seen from the figure, a pronounced anomaly of specific heat is observed at $T_c$. 

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Figure 1. Heat capacity of \( \text{Ba(Fe}_{0.92}\text{Co}_{0.08})\text{As}_2 \) measured at 0 T (circles) and 9 T (squares). The solid line describes the normal state specific heat (see text). The inset shows the difference between measured \( C/T \) and the normal state contribution. The \( T_c \) obtained by entropy balance construction is 20 K.

A magnetic field of 9 T strongly suppresses the anomaly and moves it to lower temperatures. In general, the total specific heat of any system is the sum of several different excitations:

\[
C_{\text{tot}}(T) = C_{\text{el}}(T) + C_{\text{ph}}(T) + C_{\text{mag}}(T) + \cdots,
\]

where \( C_{\text{el}}(T) \), \( C_{\text{ph}}(T) \) and \( C_{\text{mag}}(T) \) describe electronic, lattice and magnetic contributions to the total specific heat, respectively.

In order to estimate the phonon contribution in our system, we assume that the phonon part of the specific heat is independent of doping. Thus, we determine the phonon specific heat from the parent compound BaFe\(_2\)As\(_2\). BaFe\(_2\)As\(_2\) exhibits an SDW transition at about 140 K; hence one may expect the presence of a magnetic contribution to the low-temperature specific heat in the system. However, recent inelastic neutron scattering experiments show that, in the ordered state, spin-wave excitations have a gap of about 10 meV (\( \Delta \approx 116 \) K) \([31]\). Consequently, at temperatures below 40 K, \( C_{\text{mag}} \) is negligible, and we separate the contributions to the specific heat of the parent compound as \( C = \gamma T + C_{\text{ph}} \).

Thus, to describe the experimental data of \( \text{Ba(Fe}_{0.92}\text{Co}_{0.08})\text{As}_2 \) above \( T_c = 20 \) K, we adjust \( \gamma \) to obtain the best agreement between the data and \( C = \gamma T + C_{\text{ph}} \). The following four points make us confident that our determination of \( \gamma \) and \( C_{\text{ph}} \) for the doped compound is reasonable.

(i) There is good agreement of \( C = \gamma T + C_{\text{ph}} \) above 20 K (see figure 1). (ii) The \( \gamma \) that we obtain in this procedure provides accurate entropy balance for electronic specific heat in the superconducting state below \( T_c \). (iii) The condensation energy that we obtain from the resulting analysis is quantitatively consistent with other measurements (see below). (iv) Calculations and inelastic x-ray scattering measurements indicate that phonons below 10 meV are independent of doping \([32, 33]\).

The value of the normal state electronic specific heat may be compared with the density of states at the Fermi level calculated for pure BaFe\(_2\)As\(_2\). Within a single band model approximation, \( \gamma = 18 \) mJ mol\(^{-1}\) K\(^{-2}\) gives \( N(E_F) \) as 7.64 eV\(^{-1}\) f.u.\(^{-1}\) Using local density
approximation (LDA) together with generalized gradient approximation—Perdew, Burke and Emzerhof (GGA-PBE) [34] or general potential in the LAPW method [35], the calculated values of $N(E_F)$, for the parent BaFe$_2$As$_2$, are 3.93 and 3.06 eV$^{-1}$ f.u.$^{-1}$, respectively. Thus, mass renormalization is roughly a factor of 2, consistent with the mass renormalization determined by optics [36] and ARPES [37].

At 20 K, the specific heat exhibits a jump $\Delta C/T_c = 24$ mJ mol$^{-1}$ K$^{-2}$ (see the inset in figure 1), consistent with previous reports [9, 23, 38, 39]. It is also similar to $\Delta C/T_c = 28$ mJ mol$^{-1}$ K$^{-2}$ obtained for Ba$_{0.8}$K$_{0.2}$Fe$_2$As$_2$ [23, 40]. The size of the jump at $T_c$ depends on details of the superconducting state. Taking $\gamma = 18$ mJ mol$^{-1}$ K$^{-2}$, the ratio $\Delta C/T_c \gamma = 1.33$ is very close to, albeit smaller than, the weak-coupling Bardeen–Cooper–Schrieffer (BCS) value 1.43.

Using the normal state specific heat, we can extract the condensation energy and relate it to the thermodynamic critical field. This quantity can be obtained by integrating the entropy difference:

$$ U = \int [S_n(T) - S_s(T)] dT, $$

where $S_n$ and $S_s$ denote entropy of the normal and superconducting states, respectively. In the case of Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$, this analysis gives $U = 1.27$ J mol$^{-1}$ and the thermodynamic critical field $H_c = 0.23$ T. Using the penetration depth $\lambda = 325$ nm obtained from magnetic force microscopy (MFM) [41] at a slightly different doping level and consistent with $\mu$SR results [43] and coherence length $\xi = 27.6$ obtained by scanning tunnelling spectroscopy (STS) [16] gives a Ginzburg–Landau parameter $K = \lambda/\xi = 118$. Using this value and our thermodynamic critical field $H_c$, we obtain $H_{c2} = 38$ T, from the expression $H_{c2} = \sqrt{2}K H_c$, in reasonable agreement with the published value of about 40 T [44] and the value of 39 T obtained from the slope of the upper critical field measured by specific heat and the expression $H_{c2} = 0.69 (dH_{c2}/dT_c)T_c$ [42]. This provides additional confidence in our phonon subtraction.

We begin our investigation on the possible symmetry of the superconducting gap by examining the temperature dependence of electronic specific heat. Figure 2 shows the non-lattice part of the specific heat of Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$ obtained by subtracting the phonon contribution together with a small Schottky contribution below 1 K (see below). At low temperatures, a sizeable residual specific heat coefficient $\gamma_0 = 3.7$ mJ mol$^{-1}$ K$^{-2}$ is observed in this system. Similar behavior has also been reported in Ba$_{0.8}$K$_{0.4}$Fe$_2$As$_2$ ($\gamma_0 = 7.7$ mJ mol$^{-1}$ K$^{-2}$) [45] and Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ ($\gamma_0 \approx 3$ mJ mol$^{-1}$ K$^{-2}$ for optimal-doped samples) [23]. Interestingly, a sizeable value of the residual specific heat coefficient has also been observed in superconducting cuprates [46, 47]. For Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$, $\gamma_0 = 3.7$ mJ mol$^{-1}$ K$^{-2}$ amounts to 20% of $\gamma$. We rule out that $\gamma_0$ originates from a non-superconducting portion of the sample based on the fact that x-ray analysis limits impurity phases to less than 5%, and we observe full diamagnetic shielding from magnetization measurements. Alternative explanations for the origin of the residual $\gamma_0$ include pair breaking effects of an unconventional superconductor [56], crystallographic defects or spin glass behavior.

To fit the $C_{el}/T$ data in figure 2, we use the BCS expression for specific heat:

$$ C_{BCS} = \frac{\Delta}{T_c} \frac{d}{dt} \int_0^\infty dy \left( \frac{6\gamma \Delta_0}{k_B \pi} \right) [f \ln f + (1 - f) \ln (1 - f)], $$

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Figure 2. Temperature dependence of the electronic specific heat of Ba(Fe_{0.92}Co_{0.08})_2As_2. The dashed and solid lines are theoretical curves based on BCS theory (equation (3)) with one and two s-wave gaps, respectively (see text).

where \( t = T / T_c \), \( f \) is the Fermi function \( f = 1/(e^{E/k_B T} + 1) \), \( E = \sqrt{\epsilon^2 + \Delta^2} \) and \( y = \epsilon / \Delta \) (see [48]).

The results of our analysis using a single s-wave gap [49] (dashed blue line) and two separate s-wave gaps (solid red line) are shown in figure 2. In an s-wave model, a residual linear term must be an extrinsic contribution to the specific heat, and hence we subtracted \( \gamma_0 \) from \( \gamma \) for the purposes of these fits. The normal state Sommerfeld coefficient \( \gamma = (18 - 3.7) \) mJ mol\(^{-1}\) K\(^{-2}\) and \( T_c = 20 \) K were held fixed during the fits. The gap value obtained from the single gap fit is 3 meV and may be compared with \( \Delta_0 = 6 \) meV derived for hole-doped Ba_{0.6}K_{0.4}Fe_2As_2 with \( T_c \approx 37 \) K [45]. Taking \( \Delta_0 = 3 \) meV and \( T_c = 20 \) K gives \( \Delta_0 / (k_B T_c) = 1.74 \), close to the weak coupling value of 1.76. However, as can be seen from figure 2, the single gap fit does not describe the data sufficiently and clearly indicates the presence of low-energy excitations in the system below 8 K. A much better description is obtained by fitting the data to \( C = (1 - A)C_{BCS}(\Delta_1) + AC_{BCS}(\Delta_2) \), which gives the solid red line and the parameters \( \Delta_1 = 1.65 \) meV, \( \Delta_2 = 3.75 \) meV and \( A = 0.62 \). While this fit provides a reasonable description of the data, we emphasize that there are multiple anisotropic gap descriptions that could provide a similarly good fit. From these data alone, we cannot determine whether or not nodes exist.

The low-temperature specific heat of Ba(Fe_{0.92}Co_{0.08})_2As_2 measured in several magnetic fields is presented in figure 3. Below 1 K, an upturn in \( C/T \) is observed. Such behavior has already been observed in Ba(Fe_{0.92}Co_{0.08})_2As_2 samples grown from In flux as well as in Ba_{0.6}K_{0.4}Fe_2As_2 and FeSe single crystals [45, 50, 51]. The origin of the anomaly is most probably related to the presence of a small amount of magnetic impurities. With increasing magnetic field, the upturn shifts to higher temperatures and transforms into a maximum, which gradually broadens and diminishes in magnitude with further rising field. Such behavior indeed resembles that expected for a degenerate ground state split due to the Zeeman effect in internal and external magnetic fields. Thus, to avoid the effects of this magnetic contribution, we rely...
Figure 3. Heat capacity of Ba(Fe_{0.92}Co_{0.08})_2As_2 plotted as C/T versus T^2 with magnetic field applied along the c-axis.

on a linear extrapolation of the data from above 1.5 K. The red solid lines in figure 3 display the linear tendency of low-temperature specific heat displayed as C/T(T^2). We have used it to determine the electronic specific heat at 0 K by extrapolating the low-temperature data to zero temperature.

The so-obtained $\Delta \gamma = [C(H) - C(0)]/T$ derived at 0 K, as described above, as well as at several other temperatures is presented in figure 4. Similar data were obtained in [23]. The data were presented in the form of $\Delta \gamma / \gamma$ versus $H/H_{c2}$ with $\gamma = 18 \text{ mJ mol}^{-1} \text{ K}^{-2}$ and $H_{c2} = 39 \text{ T}$. In fully gapped superconductors, the localized quasi-particle states in vortex cores result in $\Delta \gamma$ proportional to $H/H_{c2}$, because the number of vortices is proportional to the magnetic field. As can be seen from the figure, the specific heat rises much faster with the field than is expected for a simple s-wave gap. This could indicate an anisotropic gap or point to a field-dependent coherence length [52, 53]. The latter scenario, however, is unlikely to be the sole explanation as the temperature dependence indicates low-energy excitations in the system (see figure 2). Thus, we further explore the expectation of an anisotropic gap for an explanation of the $\Delta \gamma(H)$ dependence.

It was shown theoretically [55] using microscopic quasi-classical theory that an anisotropic gap structure can display a significant field dependence in $\Delta \gamma(H)$. Taking into account this model, the experimental data may be well described at low magnetic field (see the blue solid line in figure 4) using the gap anisotropy ratio $\alpha = 0.5$ ($\Delta_{\text{min}}/\Delta_{\text{max}} = 0.5$), which is in reasonable agreement with the two-gap fit of the temperature dependence in figure 2.

An extreme anisotropic gap is that of a nodal superconductor as occurs in the cuprates. For clean d-wave superconductors, it has been shown by Volovik [54] that the quasi-particle excitation spectrum is shifted by the superfluid velocity, resulting in $\Delta \gamma \propto \sqrt{H/H_{c2}}$. The quasi-particles that contribute to the density of states come from regions far from the vortex cores and close to the nodes. This situation is presented in figure 4 by the red dashed line. As can be seen from the figure, $\Delta \gamma$ is not increasing as strongly as expected for clean d-wave superconductors.
Figure 4. Field-induced change in the low-temperature specific heat of Ba(Fe_{0.92}Co_{0.08})_2As_2, obtained at 0 K by extrapolating the experimental data of figure 3 to zero temperatures (see text). The green dotted line and red dashed line represent field dependences expected for s-wave and d-wave descriptions, respectively. The blue solid line is a theoretical curve for anisotropic s-wave superconductors (see text).

Finally, we consider an additional alternative. The finite density of states \( \gamma_0 \) observed in Ba(Fe_{0.92}Co_{0.08})_2As_2 may also be related to the impurity scattering effect as expected for a dirty d-wave superconductor. It has been shown by Kübert and Hirschfeld [56] that in the dirty d-wave limit, \( \Delta \gamma \) behaves like \( H \log H \) at the lowest fields \( (H_{c1} \leq H \ll H_{c2}) \). In this approach, the field dependence of field-induced specific heat may be expressed as [57]

\[
\frac{\Delta \gamma}{\gamma_n} = A \left( \frac{H}{B} \right) \log \left( \frac{B}{H} \right),
\]

where \( A = 0.322(\Delta_0/\Gamma)^{1/2} \), \( B = \pi H_{c2}/2a^2 \), \( a \approx 0.5 \), and \( \Delta_0 \) and \( \Gamma \) denote a maximum of the gap and impurity scattering rate, respectively. The solid line in figure 5 is a fit of equation (4) to the experimental data, resulting in the parameter \( \Delta_0/\Gamma = 78 \). During this analysis, the value of \( H_{c2} \) was fixed to 40 T. The inset in figure 5 shows the data for Ba(Fe_{0.92}Co_{0.08})_2As_2 together with a curve calculated for a clean d-wave superconductor. As can be seen, the field dependence of \( \gamma(H)/\gamma_n \) as well as the magnitude of the residual specific heat may be consistent with the dirty d-wave scenario. The point of these two fits (figures 4 and 5) is to demonstrate that we cannot distinguish between these two different interpretations. The quality of agreement between the two-gap analysis in figure 4 and the dirty d-wave analysis of figure 5 is comparable.

From the above analysis, we can determine that a single isotropic s-wave gap is incapable of describing the specific heat data. The form of the anisotropic gap, however, cannot be determined by our data alone. We see that two extreme cases of multiband s-wave and a dirty d-wave scenario are each in reasonable agreement with the data. Results from NMR [19],

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Figure 5. Normalized field-induced change in the low-temperature specific heat of Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$, obtained at 0 K, versus magnetic field. The red solid line is a fit of equation (4) to the experimental data. Inset: the same data plotted versus $a(H/H_c^2)^{1/2}$ with $a = 0.5$ and $H_c^2 = 40$ T.

thermal conductivity [20]–[22], ARPES [15], penetration depth [17], $\mu$SR [18] and Raman [24] also conclude that an anisotropic gap is necessary to describe their data on Co-doped BaFe$_2$As$_2$, although the extent to which varies from gapless to mild multiband behavior. A nodal gap imposed by symmetry, as in the case of the cuprates, is ruled out by the vanishingly small residual linear term of the thermal conductivity. Hence, the dirty d-wave analysis applied above should not be directly applicable. However, accidental nodes as anticipated in some spin-fluctuation models of the pnictides (see [28]–[30]) cannot be ruled out by the thermal conductivity results [58] and are also consistent with our specific heat data. The accidental node scenario has the favorable aspect that the nodes can be lifted by disorder and/or doping, which would help reconcile some of the seemingly contradictory results [59]. Further measurements as a function of doping and disorder are necessary to help elucidate the gap structure, not to mention the variations between different families and dopant atoms.

4. Summary

In summary, we have used low-temperature specific heat and its magnetic field response to explore details of the symmetry of the superconducting gap in the electron-doped Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$ superconductor with $T_c = 20$ K. Using the phonon part of the specific heat of pure BaFe$_2$As$_2$, we determine the normal state Sommerfeld coefficient in Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$ to be 18 mJ mol$^{-1}$ K$^{-2}$. The temperature variation of the electronic specific heat below $T_c$ may be well described by the presence of two superconducting gaps, pointing to complex gap structure in the system. The field-induced low-temperature specific heat cannot be explained by simple clean s- or d-wave descriptions. Its behavior also indicates a strongly anisotropic gapped superconductor.
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Note added. During the completion of this manuscript, we became aware of [60], which used a similar procedure to determine the phonon contribution to the specific heat of a similar crystal. The resulting temperature dependence was analyzed within a two-band model, with results in good agreement with ours.

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