PHYSICAL REVIEW B **83**, 100513(R) (2011)

Anisotropic phase diagram and superconducting fluctuations of single-crystalline $SmFeAsO_{0.85}F_{0.15}$

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(Received 10 January 2011; published 24 March 2011)

We report on the specific-heat determination of the anisotropic phase diagram of single crystals of optimally doped $SmFeAsO_{1-x}F_x$. In zero field, we find a clear cusplike anomaly in C/T with $\Delta C/T_c = 24$ mJ/mol K^2 at $T_c = 49.5$ K. In magnetic fields along the c axis, pronounced superconducting fluctuations induce broadening and suppression of the specific-heat anomaly which can be described using three-dimensional lowest-Landau-level scaling with an upper critical field slope of –3.5 T/K and an anisotropy of $\Gamma = 8$. The small value of $\Delta C/T_c$ yields a Sommerfeld coefficient $\gamma \sim 8$ mJ/mol K^2, indicating that $SmFeAsO_{1-x}F_x$ is characterized by a modest density of states and strong coupling.

DOI: 10.1103/PhysRevB.83.100513 PACS number(s): 74.25.Dw, 74.25.Bt

Following the initial discovery[1] of superconductivity at temperatures up to 26 K in $LaFeAsO_{1-x}F_x$, superconductivity has been found in a large number of materials whose common structural motif is the presence of FeAs (or FeSe,Te) planes.[2–4] The highest values of T_c of ~56 K (resistive onset) were achieved in Sm- and Gd-based 1111 materials.[5] The high values of T_c, and the prospect of unconventional $s^{\pm}$ symmetry of the superconducting order parameter, pairing mediated by spin fluctuations, and multiband superconductivity have generated tremendous interest in these new superconductors. The FeAs superconductors have distinguishing macroscopic properties such as an enormous upper critical field combined with generally low superconducting (SC) anisotropy. The upper critical field, H_{c_2}, its anisotropy, and the specific-heat anomaly at the superconducting transition are fundamental bulk characteristics that shed additional light on the microscopic length scales, the Fermi-surface topology, and electronic structure of the superconductor.

Here we present single-crystal specific-heat measurements of $SmFeAsO_{0.85}F_{0.15}$ to determine the anisotropic phase diagram and the effect of SC fluctuations in this material. A clear cusplike anomaly is observed at the SC transition with a height of $\Delta C/T_c \approx 24$ mJ/mol K^2, which is substantially smaller than the prediction based on the scaling $\Delta C/T_c \propto T_c^2$ reported for various Ba-122-based materials.[6] The shape of the zero-field transition and its evolution in applied magnetic fields reveal pronounced SC fluctuation effects which can be consistently described in the framework of three-dimensional (3D) lowest-Landau-level (LLL) scaling, yielding an upper critical field slope of –3.5 T/K for $H \| c$ and a coherence length anisotropy $\Gamma = 8$. The strong SC fluctuations are manifested in the very large value of the Ginzburg number $G_i \sim 1.6 \times 10^{-2}$. Entropy conservation and the low value of the specific-heat anomaly imply that the Sommerfeld coefficient of the electronic specific heat, $\gamma \sim 8$ mJ/mol K^2, is lower than previously anticipated, identifying $SmFeAsO_{0.85}F_{0.15}$ as a superconductor with modest density of states and strong coupling.

We used a membrane-based steady-state ac microcalorimeter[7] enabling high-precision measurement of the specific heat of submicrogram samples. The absolute accuracy of our specific-heat data was checked against gold samples of a size similar to our pnictide crystals. $SmFeAsO_{0.85}F_{0.15}$ crystals with approximate sizes of $108 \times 95 \times 7$ μm^3 (sample I) and $130 \times 79 \times 13$ μm^3 (sample II) were grown in a high-pressure synthesis procedure using NaCl/KCl flux.[8] The sample size was deduced from optical micrographs such as those shown in the upper inset of Fig. 2. We estimate that this procedure induces an error bar of 10%–15% into the quoted specific-heat values mostly due to uncertainties arising from a nonuniform sample thickness as judged from edge-on pictures. The magnetic characterization of the samples[9] reveal a temperature-independent magnetic moment at low temperatures and a transition width of ~1.5 K, underlining the high quality of the crystals.

The inset of Fig. 1(a) displays the specific-heat anomaly near $T_c \sim 49.5$ K of sample I in zero field. The specific heat is almost linear in temperature above T_c up to 60 K, the highest temperature measured. We use a linear extrapolation of the normal-state specific heat C_n plus a small correction due to SC fluctuations, described in detail in this paper, as the background to analyze the specific heat of $SmFeAsO_{0.85}F_{0.15}$ in the temperature range close to $T_c(H)$. At lower temperatures this background specific heat is no longer applicable since the Debye function approaches the characteristic T^3 dependence.

The main panels of Fig. 1 show the SC specific heat C_s/T of sample I in various fields applied along the c axis and ab plane, respectively. Similar data were obtained for sample II. In zero field a clear, almost cusplike anomaly is observed with a height of ~4 mJ/mol K^2, approximately twice the value reported on a polycrystalline sample[10] and close to the value of 19 mJ/mol K^2 obtained on a polycrystalline sample of oxygen-deficient F-free $SmFeAsO_{1-x}$ with $T_c = 54.6$ K.[11] However, our value for $\Delta C/T_c$ is almost an order of magnitude smaller than what would be expected on the basis of the scaling $\Delta C/T_c = \text{const} \times T_c^2$ that has been reported for various Ba-122-based materials.[6] This indicates that this scaling is not universal for all FeAs superconductors *per se*, but that different material families may follow different branches with different values

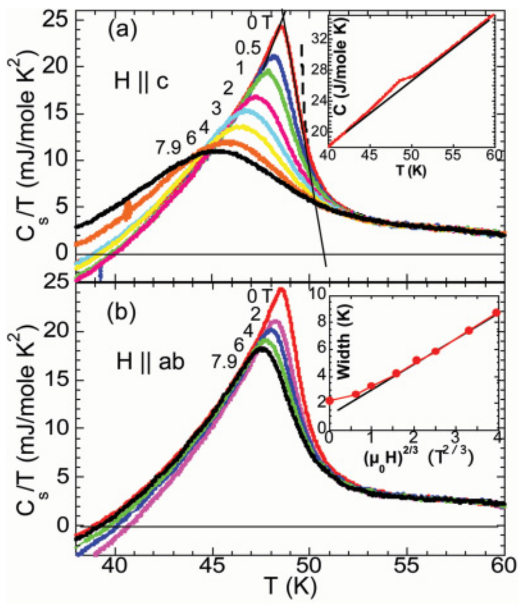

FIG. 1. (Color online) Temperature dependence of the superconducting specific heat of sample I plotted as C_s/T in various magnetic fields applied along the c axis (a) and along the ab planes (b). The dashed line in (a) represents the temperature dependence of the fluctuation contribution in Gaussian approximation. The solid lines indicate the determination of the transition width as described in the text. The inset in (a) shows the total specific heat with the solid line indicating an almost linear background. The inset in (b) shows the width of the transitions in Fig. 1(a) plotted versus $H^{2/3}$.

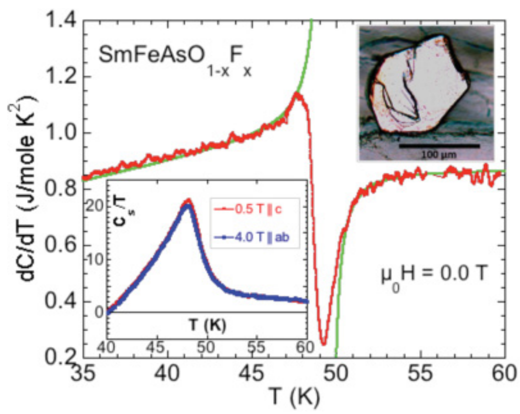

FIG. 2. (Color online) Temperature derivative dC/dT of the total zero-field specific heat. The green lines are fits to three-dimensional Gaussian fluctuations. The lower inset displays the superconducting specific heat C_s/T in units of mJ/mol K^2 in a field of 0.5 T$\|c$ and 4.0 T$\|ab$, respectively, revealing an anisotropy of 8. The upper inset shows an optical micrograph of sample I.

of the constant. The upward curvature in C/T below T_c, the sharp cusp, and the long tail above T_c are signatures of strong SC fluctuation effects. In magnetic fields applied along the c axis the peak position, T_P, of C/T shifts to lower temperatures and the peak height is strongly suppressed. Concurrently, the onset (~53 K) does not change appreciably, resulting in a strong field-induced broadening of the transition. This field dependence is reminiscent of the behavior seen in cuprate high-T_c superconductors[12] and a further indication of strong fluctuation effects in $SmFeAsO_{0.85}F_{0.15}$, as discussed in more detail in this paper. For parallel fields, $H\|ab$, this effect is much weaker, indicating strong anisotropy of $SmFeAsO_{0.85}F_{0.15}$. As shown in the lower inset of Fig. 2, the specific-heat data in 0.5 T$\|c$ virtually superimpose upon those in 4.0 T$\|ab$, showing directly that the SC anisotropy of $SmFeAsO_{0.85}F_{0.15}$ at temperatures near T_c is $\Gamma \sim 8$. This value is in good agreement with previous determinations based on torque magnetometry.[13] For comparison, the companion compound $NdFeAsO_{0.85}F_{0.15}$ has an upper critical field anisotropy of 4–5 close to T_c.[14,15]

The measured specific heat, C, contains several contributions: $C(T,H) = C_n(T) + C_s(T,H)$, where the normal-state background signal $C_n(T) = C_{\rm ph} + \gamma T + C_{\rm mag}$ results from phonons, the normal electrons, and magnetic contributions due to the magnetic Sm ions, and the SC signal is given as $C_s(T,H) = C_{\rm MF}(T,H) + C_{\rm fl}(T,H)$. Here, $C_{\rm MF}(T,H)$ describes the conventional mean-field step at the superconducting transition, and $C_{\rm fl}(T,H)$ are corrections to the mean-field signal resulting from fluctuation effects. SC fluctuation phenomena may be described using the Ginzburg-Landau (GL) free-energy functional.[16,17] Assuming a two-component SC-order parameter, the fluctuation contribution to the specific heat (per volume) in zero applied field for a three-dimensional superconductor is given in Gaussian approximation as $C_{\rm fl}(T) = C^+|t|^{-1/2}$, where $t = (T - T_c)/T_c$ and $C^+ = k_B\Gamma/8\pi\xi_{ab}^3(0)$ is the amplitude of the fluctuation specific heat for $T > T_c$. For $T < T_c$, the amplitude is $C^- = \sqrt{2}C^+$. $\xi_{ab}(0)$ is the zero-temperature value of the in-plane GL coherence length and $\Gamma = \xi_{ab}/\xi_c$ is the coherence length anisotropy. At temperatures very close to T_c the fluctuation contribution becomes large, signaling the transition to critical fluctuations and the breakdown of the Gaussian approximation. The extent of this critical regime is given by the Ginzburg number $G_i = [k_B\mu_0\Gamma T_c/4\pi\xi_{ab}^3(0)B_c^2(0)]^2/2$. In sufficiently strong applied magnetic fields, fluctuation effects are enhanced as expressed by the field-dependent Ginzburg number $G_i(H) = [H/H_{c2}(0)]^{2/3}G_i^{1/3}$. Expressions for the fluctuation specific heat and for other thermodynamic and transport quantities in magnetic fields near H_{c_2} can be obtained within the LLL approximation in which the SC-order parameter is confined to the LLL of the Cooper pairs.[18] This approximation is valid as long as $H > G_i H_{c2}(0)$. These quantities are found to depend on temperature and magnetic field only through scaling variables, which for a 3D and a two-dimensional (2D) superconductor read $[T - T_c(H)]/(TH)^{2/3}$ and $[T - T_c(H)]/(TH)^{1/2}$, respectively.[18]

A challenge in the interpretation of specific-heat results has been the fact that the SC contribution to the specific heat amounts to only a few percent of the total specific heat, $C_s \ll C_n$, implying that the normal-state background contribution has to be known with very high precision in order to achieve a definitive interpretation of fluctuation effects.[19] Alternatively, the temperature derivative of the specific heat can highlight the strong temperature variation associated with the SC transition over the smooth normal-state background. Figure 2 shows the temperature derivative dC/dT of the total zero-field specific heat. The value of ~0.85 J/mol K^2 at high temperatures corresponds to the slope of the data in the inset of Fig. 1(a). The green lines in Fig. 2 are the fits according to the predictions based on 3D Gaussian fluctuations, $dC_{fl}/dT = -C^+/2T_c|t|^{-3/2}$, yielding the amplitude $C^+ = 71.4$ mJ/mol K and $T_c = 49.5$ K. This fit describes the data well at temperatures >50 K. For the fit at $T < T_c$, a linear dependence has been added to account for the temperature dependence of C_s below T_c. The integration of the result for dC_{fl}/dT yields up to a constant the Gaussian fluctuation contribution to the specific heat as shown by the dashed line in Fig. 1(a). Since this contribution is still detectable at temperatures of ~10 K above T_c, a simple linear extrapolation of the background specific heat does not account for it. The linear dependence has to be corrected by ~0.4% to yield the Gaussian fluctuations correctly, resulting in the data as shown in Figs. 1(a) and 1(b). In particular, the nonzero SC specific heat at 60 K is a direct consequence of the Gaussian fluctuations.

The inset of Fig. 1(b) displays the field dependence of the width of the transitions shown in Fig. 1(a). The width is determined by extrapolating the line of steepest descent of the peaks to the normal-state baseline ($C_s = 0$) and to a linear extrapolation of the low-temperature side of the peaks,[20] respectively, as shown for the $H = 0$ data in Fig. 1(a). The field dependence obtained in this way comes out to be proportional to $H^{2/3}$. This is the field dependence expected in GL theory for a 3D superconductor, suggesting scaling of the in-field specific-heat data according to the 3D-LLL scheme. In analogy to Fig. 2, Fig. 3 shows the data from Fig. 1(a) in the scaling form of $dC/dT(\mu_0 H)^{2/3}$ vs $[T - T_c(H)]/(TH)^{2/3}$ using $\mu_0 dH_{c2}^c/dT = -3.5$ T/K. In fields higher than 3 T the data show good scaling, demonstrating that the shape of the in-field specific-heat transitions is determined by strong fluctuations in an anisotropic 3D superconductor. We note though that 3D and 2D scaling may be difficult to distinguish because of the scatter in the data and because the critical exponents, 2/3 and 1/2, are not strongly different. However, the observation of 3D scaling and of 3D Gaussian fluctuations, in conjunction with the value of the c-axis coherence length that is larger than the FeAs-layer spacing in the temperature range covered in Fig. 1 (see below), indicate 3D critical behavior. In theoretical analysis of the specific heat[18] the scaling properties for the quantity C_s/C_{MF} are obtained. The field and temperature dependences of C_{MF} are not known for Sm-1111; however, experimentally we observe that the coefficient $(\mu_0 H)^{2/3}$ accounts for the field evolution of the specific-heat anomaly very well. A similar relation has been previously found for yttrium barium copper oxide.[21] The scaling property is insensitive to some variability in the upper critical field slope resulting in an uncertainty of order 25% in the value of $\mu_0 dH_{c2}^c/dT$. Our result is larger than the value obtained from torque magnetometry[13] on a crystal with $T_c \sim 45$ K, which yielded $\mu_0 dH_{c2}^c/dT = -1.9/\eta$ T/K, where η is a coefficient typically taken to be of order unity. From magnetotransport data on polycrystalline samples[22] a value of $\mu_0 dH_{c2}/dT = -12$ T/K and 2D scaling of the fluctuation conductivity were deduced. However, the interpretation of such data may be complicated by the mixture of the largely different behaviors for $H\|c$ and $H\|ab$, respectively. Magnetotransport on a focused ion beam (FIB)-patterned $SmFeAsO_{0.70}F_{0.25}$ crystal[23] yielded $\mu_0 dH_{c2}^c/dT \sim 3$ T/K if one chooses as the criterion the 90% point of the normal-state resistance.

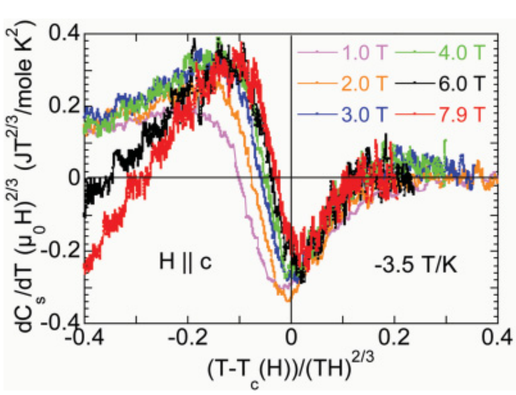

FIG. 3. (Color online) Scaling plot of $dC_s/dT(\mu_0 H)^{2/3}$ vs $[T - T_c(H)]/(TH)^{2/3}$ using an upper critical field slope of –3.5 T/K.

Using the standard single-band GL relation, the upper critical field slope can be converted into an in-plane coherence length of $\xi_{ab}(0) \approx 1.4$ nm. Combined with the anisotropy coefficient $\Gamma = 8$ this allows for an independent determination of the amplitude of Gaussian fluctuations yielding $C^+ \approx 69$ mJ/mol K, in very good—possibly fortuitous—agreement with that obtained from the fit in Fig. 2. In any case, the analysis presented here yields a consistent description of fluctuation effects in $SmFeAsO_{0.85}F_{0.15}$ crystals in terms of 3D GL theory. We estimate a c-axis coherence length of $\xi_c(0) = \xi_{ab}(0)/\Gamma \approx 0.18$ nm, which is clearly smaller than the repeat distance of the FeAs layers of $d = 0.85$ nm and may even be smaller than the FeAs-layer thickness. Clearly, 3D GL theory is no longer applicable under these circumstances and a 2D description, for instance, in terms of the Lawrence-Doniach model,[17] is required at low T. Near the SC transition, however, our 3D GL analysis applies and the relevant length scales are given by $\xi_c(T)$ and $\xi_{ab}(T)$. The crossover temperatures T_x from the 3D into the 2D regime in zero field can be estimated according to $2\xi_c(T_x) = d$ (Ref. 24), yielding $T_x \approx 41.5$ K for $T < T_c$ and $T_x \approx 65.5$ K for $T > T_c$. Thus, the majority of the data shown in Fig. 1 fall into the 3D regime, consistent with the analysis presented above.

With the help of the general thermodynamic relations $\mu_0 \frac{\Delta C_s}{T_c} = (\mu_0 \frac{dH_c}{dT})^2|_{T_c} = \frac{1}{\beta_A(2\kappa^2-1)}(\mu_0 \frac{dH_{c2}}{dT})^2|_{T_c}$ we can obtain, within GL theory, the thermodynamic critical field $B_c(0) = \mu_0 H_c(0) \approx 1.24$ T and the GL parameter $\kappa_c \approx 99$. Here, $\beta_A = 1.16$ is the Abrikosov number. Similar to other members of the FeAs family, $SmFeAsO_{0.85}F_{0.15}$ is in the limit of extreme type-II superconductivity. With this value for $B_c(0)$ a very high Ginzburg number of $G_i \approx 1.6 \times 10^{-2}$ can be deduced which is substantially larger than that reported for other FeAs superconductors[14,15,20] and is a consequence of large anisotropy, high T_c, and short coherence lengths.

Entropy conservation yields further constraints on the low-temperature electronic specific heat as the integral of C_s/T taken at temperatures above the zero crossing of C_s/T equals the integral from zero up to the zero crossing. The presence of strong fluctuations introduces uncertainty in the evaluation of this integral; however, we believe that the data in Fig. 1(a) account for the majority of the entropy, ~155 mJ/mol K. Although the explicit temperature variation of C_s at low temperatures is not known, with the zero-temperature limit $C_s/T = -\gamma$, a rough estimate based on entropy conservation yields $\gamma \sim 8$ mJ/mol K^2. Here we consider negligible any residual density of states that might arise due to nonsuperconducting phase fractions[25] or to pair-breaking scattering.[26] There is a large variation in reported values of γ for $SmFeAsO_{1-x}F_x$ ranging from $\gamma \sim 137$ mJ/mol K^2 (Ref. 27) to 44 mJ/mol K^2 (Refs. 10 and 28) and 19 mJ/mol K^2.[11] This discrepancy may arise from magnetic contributions to the specific heat associated with the magnetic ordering of the Sm^{3+} ions near 4.5 K. Notwithstanding the uncertainties in our estimate, it appears that such high values of γ are inconsistent with the rather small size of the specific-heat anomaly at T_c. Our results indicate that $SmFeAsO_{1-x}F_x$ has a modest value of γ, i.e., modest density of states $N(E_F)$, which is in contrast to Ba-122 compounds where γ values of ~50 mJ/mol K^2 have been reported.[29] Extensive compilations[3] do show that, on average, the density of states of Ba-122 based compounds is two to three times larger than that of 1111 and of 11 compounds.

We conclude that $SmFeAsO_{1-x}F_x$, and by extension, the other members of the 1111 family, are characterized by a modest density of states and strong coupling which induces high T_c. Furthermore, the small value of ΔC_s promotes a high value of the Ginzburg number, $G_i \sim 1/\Delta C_s^2$, leading to strong fluctuations. The exact value of G_i depends on additional materials parameters such as Γ^2 and ξ_{ab}^{-6}, which in the case of $SmFeAsO_{1-x}F_x$ conspire to yield extraordinarily high values of $G_i \sim 1.6 \times 10^{-2}$.

This work was supported by the US Department of Energy, Basic Energy Science, under Contract No. DE-AC02-06CH11357. Work at ETH was supported by the Swiss National Science Foundation NCCR Materials with Novel Electronic Properties (MaNEP), and work at University of Zurich was partially supported by the Swiss National Science Foundation.

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