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Superconducting fluctuation effect in $CaFe_{0.88}Co_{0.12}AsF$

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Abstract

Out-of-plane angular dependent torque measurements were performed on $CaFe_{0.88}Co_{0.12}AsF$ single crystals. Superconducting fluctuations, featured by magnetic field enhanced and exponential temperature dependent diamagnetism, are observed above the superconducting transition temperature T_c, which is similar to that of cuprate superconductors, but less pronounced. In addition, the ratio of T_c versus superfluid density follows well the Uemura line of high-T_c cuprates, which suggests the exotic nature of the superconductivity in $CaFe_{0.88}Co_{0.12}AsF$.

Keywords: superconducting fluctuation, iron-based superconductor, magnetic torque

(Some figures may appear in colour only in the online journal)

1. Introduction

Unconventional superconductivity in copper oxide superconductors develops from a complex normal state, where pseudogap is one of the remarkable phenomena. A gradual depletion of the density of states at the Fermi energy was observed below a crossover temperature T^*, signaling the opening of the pseudogap well above the superconducting transition temperature T_c in the underdoped systems. Although the pseudogap phenomenon has been very intensely investigated, no general consensus has been reached yet regarding the nature of pseudogap and its relationship to the superconductivity. It is in great debate if the T^*line will intercept with the superconducting phase boundary or it will merge with the superconducting boundary in the overdoped side [1]; correspondingly, the pseudogap is a competing order with superconductivity or a precursor of superconductivity.

Large Nernst effect or diamagnetic signal above T_c is widely observed in unconventional superconductors, which is associated with superconducting fluctuations/vortices persisting in the normal state. For examples, in cuprate superconductor $Bi_2Sr_2CaCu_2O_{8+\delta}$, $Bi_2Sr_{2-y}La_yCuO_6$, $La_{2-x}Sr_xCuO_4$, $La_{1.8-x}Eu_{0.2}Sr_xCuO_4$, $YBa_2Cu_3O_{6+x}$ [2–5], and in heavy fermion superconductor URu_2Si_2 [6]. The recent discovery of superconductivity in iron-based superconductors provide a new candidate to study the superconducting fluctuation effect because these compounds share many similarities with the cuprate superconductors, for example, the layered structure, the proximity to antiferromagnetic phase, and the similar evolution of the superconductivity with doping.

Superconducting fluctuation effect was studied in $Ba_{1-x}K_xFe_2As_2$ and controversial results are reached. Salem-Sugui *et al*, found that phase fluctuations are important in $Ba_{1-x}KxFe_2As_2$, which is consistent with nodes in the gap [7], while Mosqueira *et al*, observed classical Ginzburg–Landau scaling and excludes phase incoherent superconductivity above T_c in iron-based superconductors [8]. In $SmFeAsO_{0.8}F_{0.2}$, diamagnetic response was detected above T_c, where the authors concluded that the phase superconducting fluctuations of novel character and the conventional superconducting fluctuations are simultaneously present in the fluctuating diamagnetism [9]. All these above experiments are based on magnetization measurements.

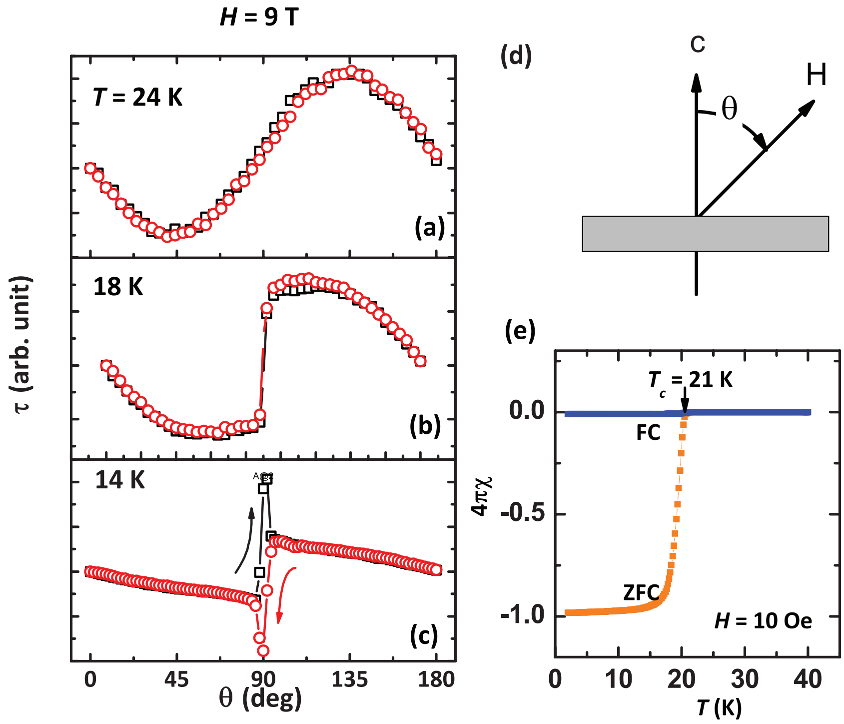

Figure 1. (a)–(c) Typical angular θ-dependent torque τ at temperatures $T = 24$, 18 and 14 K, respectively with a magnetic field $H = 9$ T. (d) Sketch of the single crystal with the orientation of the magnetic field H. (e) Temperature-dependent magnetization data under both ZFC and FC conditions.

$CaFe_{1-x}Co_xAsF$ is a 1111-type of iron-based superconductor, similar to the fluoride doped arsenide $LaFeAsO_{1-x}F_x$, but being oxygen-free. There are limited reports available on this 1111 material due to the difficulties in obtaining sizable single crystals. The typical size of the single crystals of the 1111 family is about several tens of micrometer [10]. The $CaFe_{1-x}Co_xAsF$ single crystal samples examined in this work are millimeter-sized [11, 12], even though the mass of the sample is still too small to make a strong magnetization signal. Instead, torque magnetometers provide a sensitive tool to detect magnetic response, with better resolution compared to magnetization measurements. So we performed angular-dependent torque measurements on the newly found $CaFe_{1-x}Co_xAsF$ and observed in the sample an abnormal superconducting fluctuation effect, displayed by a magnetic-field-enhanced and exponential-temperature-dependent diamagnetism. This suggests a similar origin as the superconducting fluctuation effect in cuprates, but with a much more limited region above T_c compared to the cuprate superconductors, which have short coherence length, low carrier density, high T_c and large anisotropy, leading to enhanced superconducting fluctuations. In addition, we found that the ratio of T_c and the superfluid density n_s ($\propto \lambda_{ab}^{-2}$) follows the Uemura law well, which works reasonably well for underdoped cuprate superconductors [13]. Our results suggest the exotic nature of the superconductivity in $CaFe_{0.88}Co_{0.12}AsF$, different from the conventional BCS superconductors.

2. Experimental details

The high-quality single crystal sample of $CaFe_{1-x}Co_xAsF$ was grown using the self-flux method with CaAs as the flux. The sample used in this work is with $x = 0.12$ and has a dimension of $\sim$1.2 $\times$ 1 $\times$ 0.1 mm^3 (with a mass of 120 μg). Note this millimeter-sized single crystal is already large within 1111 families, where the typical size is about several tens of micrometer. For example, the reported typical size of $LaO_{1-x}F_xFeAs$ is about 80 $\times$ 60 $\times$ 5 μm^3 [14]. Details of the sample growth procedure and characterization can be found in [11, 12]. Magnetization measurements were performed by using a magnetic property measurement system (MPMS). The temperature-dependent magnetization M curves were measured under field-cooled (FC) and zero-field-cooled (ZFC) conditions with a magnetic field H of 10 Oe. and the superconducting transition temperature is determined to be $T_{c0} \approx 21$ K (see figure 1(e)). Note that the mass of the sample is too small to produce a strong enough magnetic signal for MPMS, unless it is in the superconducting state. A torque magnetometer, which is more sensitive to the magnetic signal, instead allowed us to study the normal state response of small samples. Out-of-plane torque measurements were performed using a piezoresistive torque magnetometer in a physical property measurement system (PPMS). The angle θ is defined as the angle between the magnetic field H and the c axis of the single crystal, see figure 1(d).

3. Results and discussion

The torque of a sample of magnetic moment M placed in a magnetic field H is given by $\vec{\tau} = \vec{M} \times \vec{H}$. For paramagnetic response, torque can be written as $\tau_p = \frac{\chi_c - \chi_a}{2} H^2 \sin 2\theta$ where χ_c and χ_a are the susceptibility along the c and a axes of the crystal [15, 16]. In other words, τ has a $\sin 2\theta$ angular

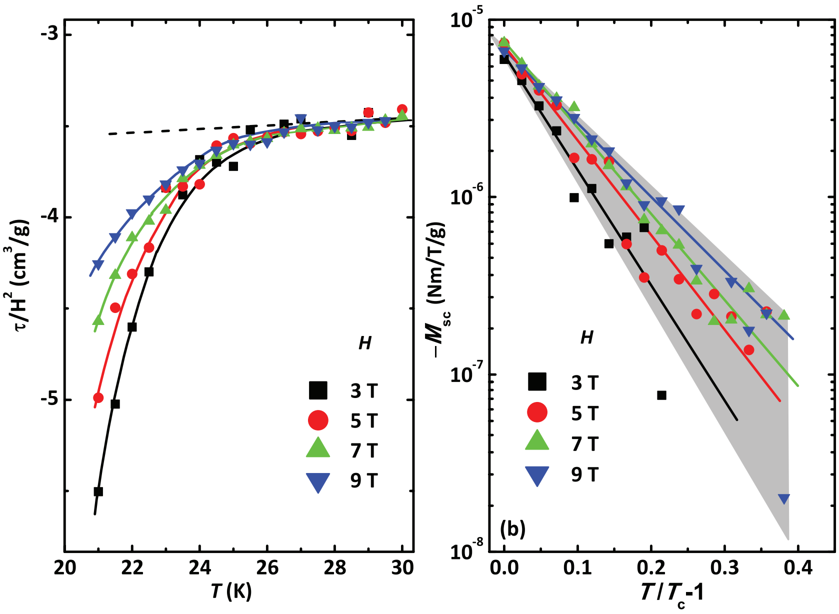

Figure 2. (a) Temperature T-dependent τ_0/H^2 curves for $H = 3, 5, 7, 9$ T. (b) $-M_{sc}$ versus $T/T_c - 1$ curves for $H = 3, 5, 7$ and 9 T.

dependence and a H^2 magnetic field dependence. This is the case for the temperatures above T_c, as shown in figure 1(a), for $T = 24$ K and $H = 9$ T in $CaFe_{0.88}Co_{0.12}AsF$. At temperatures below T_c, typical torque data is shown in figure 1(c), where a sharp peak around 90° is observed and large hysteresis is present between the torque data measured with increasing and decreasing angles. At intermediate temperatures, for example $T = 18$ K, both the $\sin 2\theta$ term and Abrikosov vortex torque contribute to the signal, as shown by figure 1(b).

In order to study the superconducting fluctuation effect in $CaFe_{1-x}Co_xAsF$, we focused our study on the temperature region above T_c, where the torque signal can be well fitted by $\tau \sin 2\theta$. We extract τ and summarize the T dependence of τ/H^2 curves as shown in figure 2(a). From bottom to top, the curves correspond to $H = 3, 5, 7$ and 9 T, respectively. Note that the data for different H merge with each other at high temperatures and then start to deviate from each other, where τ no longer has a H^2 magnetic field dependence. For each fixed field, the torque data show linear behavior (weak temperature dependence) at high temperatures, then deviate from this behavior and shows a sharp decrease at low temperatures. The sharp decrease is due to superconducting fluctuations, as reported in cuprate superconductors $Tl_2Ba_2CuO_{6+\delta}$ [17], $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ [18] and $Bi_2Sr_2CaCu_2O_{8+\delta}$ [3]. Note that the $T_c(0)$ of this material determined from magnetization measurements is 21 K, while the paramagnetic response is above 28–30 K (the temperature where the curves of τ/H^2 merge with each other). The generation of fluctuating Cooper pairs above T_c results in the appearance of the diamagnetic contribution to the magnetization $-M_{sc}$ besides the paramagnetic contribution from the fermionic carriers. This figure 2(a) explicitly shows that superconducting fluctuation exists above T_c and survives to temperature 28–30 K. The data shown here is somewhat scattered due to the small mass of the single crystal sample.

After subtracting the linear behavior as a background for the paramagnetism (see dashed straight line), we can obtain the superconducting torque τ_{sc}. Figure 2(b) shows the fluctuation-induced diamagnetism, $M_{sc} = \tau_{sc}/H$ versus $T/T_c - 1$ curves. Note that τ is the amplitude of the $\sin 2\theta$ term, therefore $M_{sc} = \tau_{sc}(\pi/4)/[H \sin(\pi/4)] = M_c - M_a$ (M_c and M_a are the c and a components of the diamagnetic magnetization) since $\tau_{sc} = M_c H \sin\theta - M_a H \cos\theta$. Generally for layered superconductors, $|M_c|$ is larger than $|M_a|$, therefore M_{sc} is negative and respresents the superconducting diamagnetism [19]. For a fixed temperature, $|M_{sc}|$ is increasing with increasing magnetic field, which is abnormal, since in conventional superconductors, the fluctuation signal from amplitude fluctuations is suppressed by weak magnetic field and the fluctuations become unresolved above 1000 Oe [3, 20]. In addition, we found that M_{sc} has an exponential dependence on $T/T_c - 1$ for all the magnetic field examined, i.e. $|M_{sc}| \propto e^{-b(H)(1-T/T_c)}$, where b is the slope of the linear curve. All the curves converge and reach one fixed value of M_{sc} when the temperature approaches T_c from above. A constant value of M_{sc} is also reached in $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ [19]. Nevertheless, the constant M_{sc} at T_c might be very important and requires further study to explore the physics behind it.

The exponential temperature dependence of M_{sc} is an experimental observation and is not routed in any theory. However, in many cases, superconducting diamagnetism vanishes in this unusual exponential fashion above T_c. For example, we measured a series of $Bi_2(Sr,La)_2CuO_{6+\delta}$ samples and found same exponential dependence of M_{sc} (data not shown here). The same situation is also reported in the literature for $La_{2-x}Sr_xCuO_4$, $Bi_2(Sr,La)_2CuO_{6+\delta}$ and $HgBa_2CuO_{4+\delta}$ [21]. This exponential dependence reflects the exotic nature of the superconducting fluctuation which is beyond the framework of GL theory and might come from preformed Cooper pairs. We notice that the giant Nernst effect above T_c is claimed to be within the framework of Gaussian fluctuation theory [22–24]. It is expected from Gaussian fluctuation theory that the Nernst and magnetization responses from

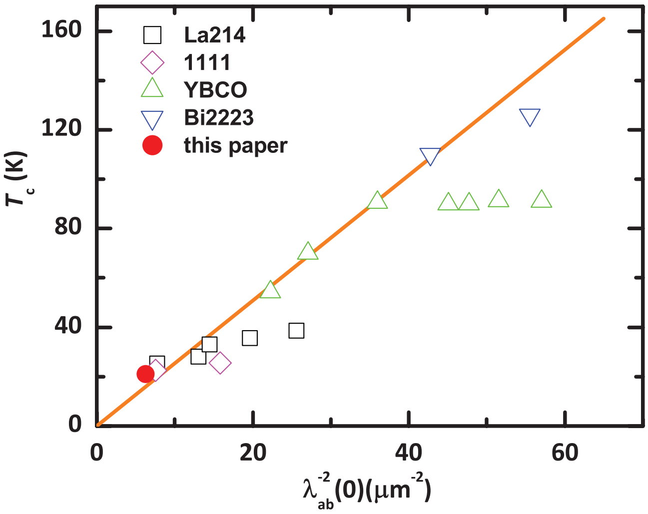

Figure 3. Uemura plot for hole-doped cuprate superconductors. The solid circle represents data from the present work. The diamonds are data for $LaFeAsO_{1-x}F_x$ taken from [34]; the data for cuprate superconductors are taken from [13].

superconducting fluctuations should scale with each other. However, our observation of exponential temperature dependence of the fluctuations is not consistent with Guassian fluctuation theory which gives a divergence of Nernst coefficent at T_c since $\alpha_{xy}^{SC} \propto (T - T_c)^{(d-4)/2}$ (see [22]).

The Ginzburg number $Gi = (\pi\lambda_0^2 k_B T_c \mu_0 / 2\xi_c \phi_0^2)^2$ is a measure of the importance of thermal fluctuations (where λ_0 is the London penetration depth, ξ_c is the out-of-plane coherence length, k_B is the Boltzmann constant and ϕ_0 is the flux quantum) [25]. In our case, $\lambda_0 = 400$ nm (see [26]), $T_c = 21$ K and $H_{c2}^{ab} = 124$ T (estimated by Werthamer–Helfand–Hohenberg (WHH) relation: $H_{c2}^{ab}(0) = -0.7T_c(dH_{c2}/dT_c) = 124$ T, where $dH_{c2}/dT_c = 8.5$). Hence, Gi is estimated to be 7.87×10^{-3}. This value is comparable with cuprate superconductors, where typical values of $Gi \approx 10^{-2}$ ([27]); but much larger than that of MgB_2 (10^{-5}) ([28]) and heavy fermion superconductor $CeCoIn_5$ (10^{-7}–10^{-6}) ([29]). The critical interval above T_c is estimated to be 0.17 K since $\Delta T_G (= T_G - T_c)/T_c \sim Gi$ ([30]). This is much smaller than the fluctuation region observed in our torque measurements.

Note that the absolute value of lambda depends on the ratio n/m^* (n is the charge carrier density and m^* is the effective mass) but also on the impurity scattering. We estimate the mean-free-path and how this compares to the gap as follows. $\frac{\xi_0}{l} = \frac{\hbar v_F}{\pi\Delta(0)l} = \frac{\hbar}{\pi\Delta(0)\tau}$, where Δ is the superconducting gap, τ is the scattering rate, l is the mean free path of free electron, $\hbar = h/2\pi$ (h is the Planck constant), and ξ is the coherence length. If $\xi_0/l \gg 1$, then the material is in the dirty limit; if $\xi_0/l \ll 1$, then the material is in the clean limit. In our case, the resistivity around T_c is $\rho = 0.6$ mΩcm. Since $\rho = \frac{m^*}{n_s e^2 \tau} = \frac{\lambda^2 \mu_0}{\tau}$ and the superconducting gap can be estimated to be $1.7k_B T_c$, therefore we obtain $\frac{\xi_0}{l} = \frac{\hbar}{\pi\Delta(0)\tau} = \frac{\hbar\rho}{\pi(1.7k_B T_c)\lambda^2\mu_0} \approx 2$, which is in the intermediate region between the clean and dirty limit.

To understand the physics behind this exponential temperature dependence of M_{sc}, further study is required. Nevertheless, this magneti-field-enhanced magnetization is reported previously in cuprate superconductors and was attributed to phase fluctuations, instead of conventional amplitude fluctuations [3, 19]. For $T > 1.35\ T_c$, the data is scattered and within the level of noise. So the superconducting fluctuation can be detected up to about 1.35 T_c, a much more limited region compared to that of cuprate superconductors, for example, $La_{2-x}Sr_xCuO_4$ [31] and $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$, where the superconducting fluctuations can be detected up to several times of T_c [18]. This may be related with the fact that cuprates are more two-dimensional than FeSCs, since the fluctuation effect is affected by anisotropy [20, 32]. Another possible reason is that the sample examined in this work is $x = 0.12$, which is slightly underdoped and close to the optimum doping level, where the superconducting fluctuation effect is weaker compared to the very underdoped samples. Note that an abnormal Nernst effect is reported in the normal state of $LaO_{1-x}F_xFeAs$. The anomalous change in the Nernst signal between T_c and 50 K is argued to be related to the spin-sensity wave (SDW) but the contribution of superconducting fluctuations can not be excluded [33]. Although both samples belong to the 1111 family, the temperature range of the anomaly in the Nernst signal is much larger than this work. The reason might be that the Nernst signal is senstive to both SDW and superconducting flucutations, while torque measurement is only sensitive to the latter one.

The Uemura law describes that the superconducting transition temperature T_c is proportional to the superfluid density n_s/m^* in cuprates, organic, Chevrel phase and heavy fermion systems. Figure 3 shows the Uemura plot taken from [13]. We found that n_s and T_c of $CaFe_{1-x}Co_xAsF$ follow the Uemura law well (the data from the present work is indicated as solid circles), suggesting the exotic nature of the superconductivity in this material. We estimate the Fermi temperature $T_F = \epsilon_F/k_B$ (where k_B is the Boltzman constant, ϵ_F is the Fermi energy and $\epsilon_F = (\hbar^2/2)(3\pi^2)^{2/3}/m^*$, see [35]) to be about 240 K. Thus the T_c of this 1111 material is in the range between 1/10–1/100 of ϵ_F/k_B, in strong contrast to ordinary BCS superconductors which have T_c of less than 1/1000 of ϵ_F/k_B [35].

4. Conclusions

In summary, an abnormal superconducting fluctuation effect is observed above T_c in $CaFe_{1-x}Co_xAsF$. The strength of fluctuation is exponentially dependent on $T/T_c - 1$. The diamagnetic magnetization M_{sc} above T_c reaches a constant value when approaching T_c. The fact that 1111 follows the Uemura law and demonstrates similar superconducting fluctuation behavior as cuprates suggest that pnictides could belong to the same unique group of superconductors as cuprates.

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