Superconducting fluctuation effect in CaFe$_{0.88}$Co$_{0.12}$AsF
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 $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$, $\text{Bi}_2\text{Sr}_2\text{La}_x\text{CuO}_6$, $\text{La}_2\text{Sr}_x\text{CuO}_4$, $\text{La}_{1.8-\delta}\text{Eu}_0.2\text{Sr}_x\text{CuO}_4$, $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ [2–5], and in heavy fermion superconductor $\text{URu}_2\text{Si}_2$ [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 $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ and controversial results are reached. Salem-Sugui et al., found that phase fluctuations are important in $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$, 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 $\text{SmFeAsO}_0.8\text{F}_{0.2}$, 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.
2. Experimental details

The high-quality single crystal sample of CaFe$_{1-x}$Co$_x$AsF 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$_{1-x}$FxFeAs 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 $T_c \approx 21$ 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 $\theta$ 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 $\tau = M \times H$. For paramagnetic response, torque can be written as $\tau_p = \chi_p H^2 \sin 2\theta$ where $\chi_c$ and $\chi_a$ are the susceptibility along the $c$ and $a$ axes of the crystal [15, 16]. In other words, $\tau$ has a $\sin 2\theta$ angular dependence.
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$_{0.85}$Co$_{0.15}$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 2θ 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$_x$AsF, 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 $\tau$ and summarize the $T$ dependence of $\tau/H^2$ 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 $\tau$ 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$_2$Ba$_2$CuO$_{6+\delta}$ [17], Bi$_2$Sr$_2$−$_x$La$_x$CuO$_{6+\delta}$ [18] and Bi$_2$Sr$_2$−$_x$CaCu$_2$O$_{6+\delta}$ [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 $\tau/H^2$ 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 $\tau_{sc}$. Figure 2(b) shows the fluctuation-induced diamagnetism, $M_{sc} = \tau_{sc}/H$ versus $T/T_c − 1$ curves. Note that $\tau$ is the amplitude of the sin 2θ term, therefore $M_{sc} = \tau_{sc}(\pi/4)[H \sin(\pi/4)] = M_c − M_a$, where $M_c$ 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 represents 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., $|M_{sc}| \propto e^{−b(T/T_c−1)}$, 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$_2$Sr$_{2−x}$La$_x$CuO$_{6+\delta}$ [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)$_2$CuO$_{6+\delta}$ 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$_x$CuO$_4$, Bi$_2$(Sr,La)$_2$CuO$_{6+\delta}$ and HgBa$_2$CuO$_{4+\delta}$ [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
superconducting fluctuations should scale with each other. However, our observation of exponential temperature dependence of the fluctuations is not consistent with Gaussian fluctuation theory which gives a divergence of Nernst coefficient at $T_c$ since $\xi \propto (T - T_c)^{d-4}/2$ (see [22]).

The Ginzburg number $Gi = (\pi \lambda^2 / h T_c) (\xi / \phi_0)^2$ is a measure of the importance of thermal fluctuations (where $\lambda_0$ is the London penetration depth, $\xi$ is the coherence length, $k_B$ is the Boltzmann constant and $\phi_0$ is the flux quantum) [25]. In our case, $\lambda_0 = 400 \text{nm}$ (see [26]), $T_c = 21 \text{K}$ and $H_{c2}^{ab} = 124 \text{T}$ (estimated by Werthamer–Helfand–Hohenberg (WHH) relation: $H_{c2}^{ab}(0) = -0.7T_c(dH_{c2}/dT_c) = 124 \text{T}$, where $dH_{c2}/dT_c = 8.5$). Hence, $Gi$ is estimated to be $7.87 \times 10^{-3}$. 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$_5$ $(10^{-2} – 10^{-6})$ ([29]). The critical interval above $T_c$ is estimated to be $0.17 \text{K}$ since $\Delta T_0 = (T_0 - T_c) / \Delta Gi$ ([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.

$$\frac{\xi}{T} = \frac{\hbar}{2m^*e},$$

where $\Delta$ is the superconducting gap, $\tau$ is the scattering rate, $\lambda$ is the mean free path of free electron, $\hbar = h/2\pi$ ($h$ is the Planck constant), and $\xi$ is the coherence length. If $\xi/\ell \gg 1$, then the material is in the dirty limit; if $\xi/\ell \ll 1$, then the material is in the clean limit. In our case, the resistivity around $T_c$ is $\rho = 0.6 \text{m}\Omega \text{cm}$. Since $\rho = \frac{\sigma}{m^*e^2} = \frac{\lambda}{\xi \ell}$ and the superconducting gap can be estimated to be $1.7k_B T_c$, therefore we obtain $\lambda = \frac{\hbar}{2m^*eT} = \frac{h \rho}{\sigma (\lambda h \phi_0)^2} \approx 2$, 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$_2$–Sr$_x$CuO$_4$ [31] and Bi$_2$Sr$_2$–La$_2$CuO$_{6+x}$, 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$_1$–F$_x$FeAs. 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 cannot 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 sensitive to both SDW and superconducting fluctuations, 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$_x$AsF 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 $T_F = c_F / k_B$ (where $k_B$ is the Boltzman constant, $c_F$ is the Fermi energy and $c_F = (\hbar^2 / 2m^*)^{1/2}$ $\text{Im}^*$, see [35]) to be about 240 K. Thus the $T_c$ of this 1111 material is in the range between 1/1000 of $c_F / k_B$, in strong contrast to ordinary BCS superconductors which have $T_c$ of less than 1/1000 of $c_F / k_B$ [35].

4. Conclusions

In summary, an abnormal superconducting fluctuation effect is observed above $T_c$ in CaFe$_{1-x}$Co$_x$AsF. 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.

Acknowledgments

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References

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