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Multiple gaps in $\text{SmFeAsO}_{0.9}\text{F}_{0.1}$ revealed by point-contact spectroscopy

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Abstract

We report the observation of two gaps in the superconductor $\text{SmFeAsO}_{0.9}\text{F}_{0.1}$ (F– SmFeAsO) with $T_c = 51.5$ K as measured by point-contact spectroscopy. Both gaps decrease with temperature and vanish at T_c , and the temperature dependence of the gaps is described by the theoretical prediction of the Bardeen–Cooper–Schrieffer (BCS) theory. A zero-bias conductance peak (ZBCP) was observed, indicating the presence of sign reversal of the gap function in F– SmFeAsO . Our results strongly suggest an unconventional superconductivity with multiple gaps in F– SmFeAsO .

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Recently, superconductivity was discovered in the Fe-based layered superconductor $\text{LaFeAs}[\text{O}_{1-x}\text{F}_x]$ (F– LaFeAsO) [1]. By replacing La by some other rare earth elements, T_c was improved quickly up to 55 K [2], which is substantially higher than that of the single-layered cuprate superconductors and exceeds the theoretical value predicted by the conventional BCS theory [3]. Superconductivity was also observed in the hole doped case [4, 5]. Thus a new family of high- T_c superconductors was opened up, providing an unique chance to understand high- T_c superconductivity. Detecting the gap structure and pairing symmetry is an essential step in revealing the mechanism of these Fe-based superconductors. Specific heat measurements on F– LaFeAsO showed a nonlinear magnetic field dependence of the electronic specific heat coefficient as expected for a nodal superconductor [6]. This was proved subsequently by the measurements of point-contact spectroscopy [7], lower critical field H_{c1} [8], London penetration depth λ and spin-lattice relaxation rate $1/T_1$ [9, 10]. Muon spin relaxation measurements also presented the possibility of dirty d-wave pairing [11]. Theoretically, some calculations support d-wave pairing [12–14] while others support an unconventional s-wave pairing [15, 16]. Most interestingly, a nodal superconductivity

with two-gap structure was suggested by NMR experiments on $\text{PrFeAsO}_{0.89}\text{F}_{0.11}$ [17]. Actually, multiband superconductivity has been predicted theoretically [18, 19, 20, 21] and was supported by the magnetic properties of $\text{SmFeAsO}_{0.8}\text{F}_{0.2}$ single crystal [22] and the high-magnetic field resistance of $\text{LaFeAsO}_{0.89}\text{F}_{0.11}$ [23]. The Andreev reflection and tunneling experiment is a powerful tool for measuring the superconducting gap. However, some Andreev reflection experiments favored an isotropic single gap in $\text{SmFeAsO}_{0.85}\text{F}_{0.15}$ and $\text{NdFeAsO}_{0.85}$ [24, 25], while others presented evidence of a two-gap scenario for $\text{LaFeAsO}_{1-x}\text{F}_x$ [26], F– NdFeAsO [27] and $(\text{Ba}_{0.55}\text{K}_{0.45})\text{Fe}_2\text{As}_2$ [28]. Moreover, scanning tunneling microscopy (STM) data seem to support a single-gap d-wave order parameter symmetry in $\text{SmFeAsO}_{0.85}$ [29]. Therefore, more experiments on various samples with high quality are still required in order to clarify this issue.

In this paper, we present point-contact spectroscopy data from compact and rigid $\text{SmFeAsO}_{0.9}\text{F}_{0.1}$ (F– SmFeAsO) samples. A zero-bias conductance peak (ZBCP) was observed repeatedly, indicating a sign reversal of the gap function of F– SmFeAsO , which is similar to that of F– LaFeAsO [7]. Moreover, two different gaps were observed, and both of them decrease with temperature and vanish at T_c . These results strongly suggest that F– SmFeAsO has an unconventional superconductivity with multiple gaps.

2. Experimental details

Polycrystalline samples of superconducting F-SmFeAsO were prepared by a high-pressure synthesis method [30]. Detailed information about the synthesis is given in a recent paper [2]. As shown in figure 1(a), the DC susceptibility (measured under a magnetic field of 1 Oe) and AC susceptibility data (measured using an AC amplitude of 0.1 Oe) exhibit a sharp magnetic transition. The width defined between the 10% and 90% cuts of the transition is below 2 K, with the middle of the Meissner transition at 51.5 K, indicating the good quality of the superconducting phase. Figure 1(b) shows a scanning electron microscopy (SEM) picture of the sample surface. Many microcrystals accumulate tightly, without obvious gaps between them. For a single microcrystal, the layered structure can be seen clearly, indicating its definite orientation, while the crystalline orientation is random from one crystal to another. Compared with samples synthesized by the common vacuum quartz tube synthesis method, the samples studied here are much more compact and rigid, and thus are more suitable for point-contact spectroscopy measurements. The point-contact junctions are prepared by carefully driving Pt/Ir alloy or Au tips towards the sample surface. The tip preparation and details of the experimental setup have been described elsewhere [31]. The surfaces of the polycrystalline samples were polished with fine metallographic grinding papers and then cleaned with pure acetone in an ultrasonic bath for about 1 min just before being mounted onto the point-contact insert. Typical four-terminal and lock-in techniques were used to measure the conductance–voltage (G – V with $G = dI/dV$) characteristics. The junction resistance varies from several ohms to several tens of ohms. Each measurement is comprised of two successive cycles, to check the absence of heating-hysteresis effects.

3. Results and discussions

Figure 2 shows the G – V curves measured at various locations on the sample surface, which have been normalized by the high-bias data. A distinct ZBCP can be seen in figures 2(e), (g) and (h), which is consistent with the STM data obtained on a similar sample [29]. The ZBCP was also observed in previous experiments on some other Fe-based superconductors; it was ascribed to a non-intrinsic effect [24] or explained by an $s\pi$ -pairing model and/or a phase separated picture [27, 28]. In contrast, no ZBCP was observed in the work of Gonnelli *et al* [26]. To understand the ZBCP observed here, some possible mechanisms have to be considered. The first one is the Andreev reflection peak for a normal metal/conventional superconductor junction with a very low interface barrier. With increasing barrier strength, such a peak will evolve into a conductance dip accompanied by two coherence peaks outside [32]. This is not the case of our experiments, in which the ZBCP was observed for various barrier strengths and can be coexistent with the coherence peaks (see figures 2(g) and (h)). The second possible mechanism is the inter-grain effect [31]. However, such an effect will become increasingly significant with decreasing junction resistance while the ZBCP observed here does not depend on the junction resistance, which varies

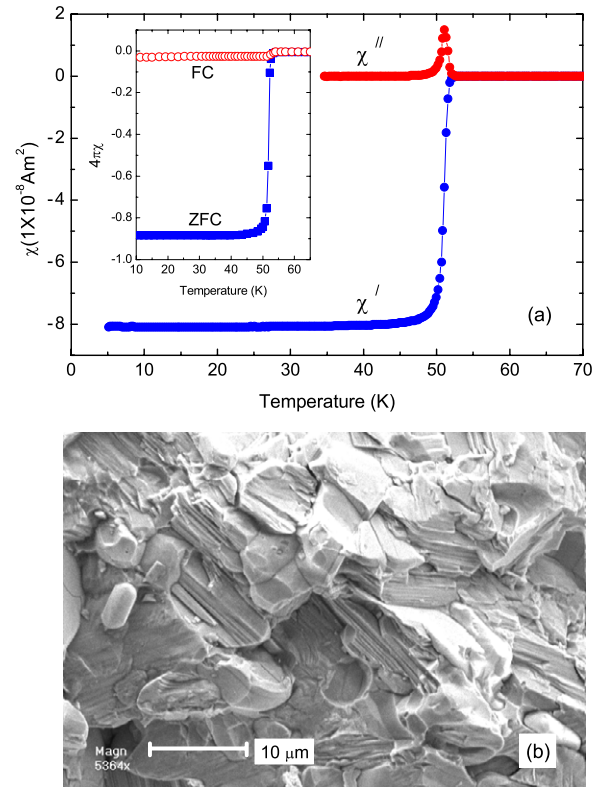


Figure 1. (a) Temperature dependence of AC susceptibility of F-SmFeAsO. Inset: DC susceptibility of F-SmFeAsO. (b) A scanning electron microscopy picture of the sample.

from several ohms to several tens of ohms. Furthermore, since our samples are very compact and rigid (see figure 1(b)), the inter-grain effect should be very weak. The third mechanism is the critical current effect [33], which is often related to the artifact due to a large contact spot, as pointed out by a recent paper [24]. In this case, a ZBCP often shows up in junctions with lower resistances, which is different from that we observed, i.e., the ZBCP is not related to the value of the junction resistance. Besides these factors, a ZBCP can also be caused by surface Andreev bound states, which is a clear signature of the pair potentials with reversal sign in momentum space and is known as an unique character of nodal superconductors [34]. The ZBCP observed here is thus most possibly associated with this mechanism.

Blonder, Tinkham and Klapwijk (BTK) [32] have proposed a simplified theory for the G – V curves of an s-wave superconductor/normal metal junction separated by a barrier of arbitrary strength. The barrier strength is parameterized by a dimensionless number Z which describes the crossover from metallic to ideal tunnel junction behavior by $Z = 0$ – ∞ . Obviously, this single-band s-wave model cannot explain the observed ZBCP, which is most possibly related to an unconventional superconductivity. Tanaka and Kashiwaya [35] extended the BTK model to deal with the issue of unconventional pairing symmetry. In this case, the angle (α) between the quasiparticle injecting direction and the main crystalline axis was introduced as another parameter. Moreover, the isotropic superconducting gap Δ was replaced by an anisotropic gap with $d_{x^2-y^2}$ symmetry: $\Delta = \Delta_0 \cos(2\theta)$,

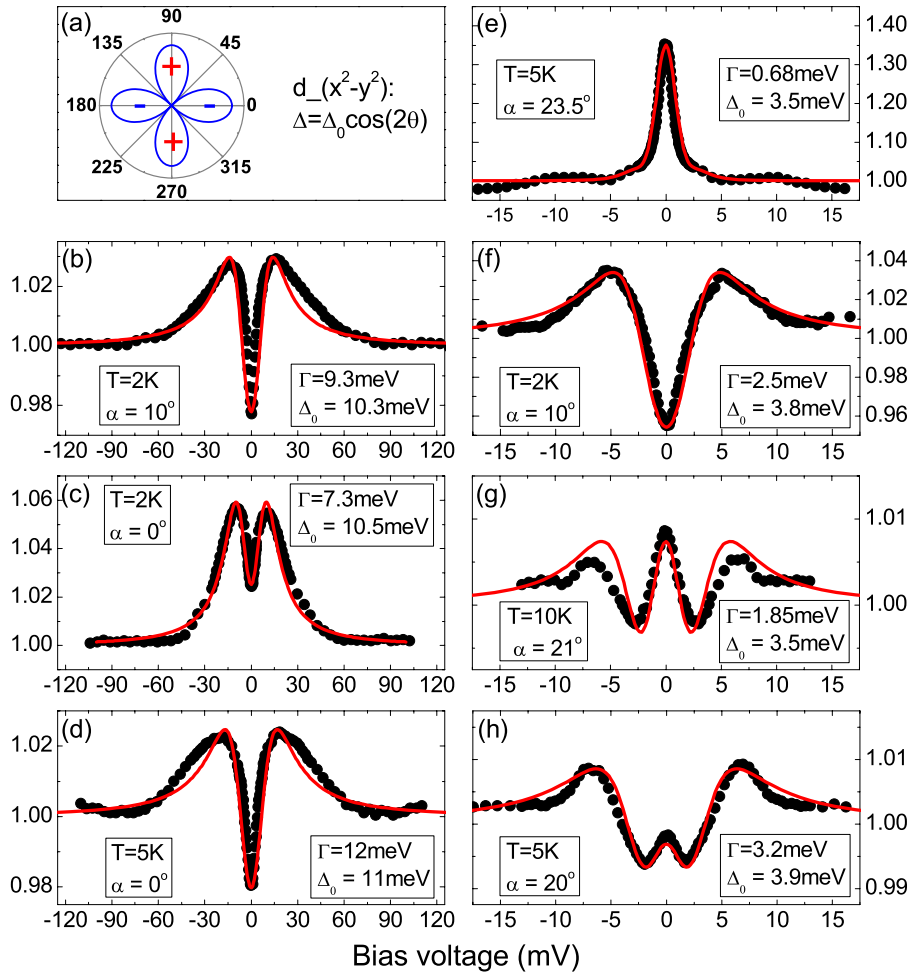


Figure 2. (a) Gap structure (d-wave type) used to fit the experimental data. (b)–(h) Experimental data (solid circles) measured at various locations on the sample surface and theoretical calculations (solid lines) with the gap function shown in (a). The quasiparticle injecting angle α presented here was determined by theoretical fitting.

as shown in figure 2(a). It was then predicted that, for $Z \neq 0$, the ZBCP is formed for all directions in the a – b plane except when tunneling into the (100) and (010) planes. This ZBCP will be suppressed when the quasiparticle scattering or surface roughness is strong enough [36, 37]. Since point-contact spectroscopy is a local measurement, detecting ZBCPs becomes possible on our polycrystalline samples if they have unconventional superconductivity. It was found that all the spectra shown in figure 2 can be described very well by the extended BTK model. Moreover, various quasiparticle injecting angles (α) are obtained from theoretical fitting for the spectra measured at different positions, indicating that our measurement is indeed a local detection. Thus, the variation of the tunneling curves depending on the location on the sample (as shown in figure 2) is partially due to the variation of the orientations of the microcrystals. The broadening parameter Γ/Δ_0 [38] in the calculations is between 0.19 and 1.09, coming from some unclear scattering mechanisms at the interface, which is similar to the case of cuprate superconductors. However, it does not thwart the determination of the gap magnitude in our analysis. To demonstrate this point, we present in figures 3(a) and (b) the best fitting results (for the data shown in figures 2(a) and (b)) when Γ was fixed

to be some consecutive values. It can be clearly seen that the gap value obtained does increase with increasing Γ value. However, when the gap value drops from around 10 meV to 8 meV, both the peak positions and the shape of the calculated curves have deviated from the experimental data significantly (for the gap value of 6 meV, such deviation becomes more serious). Therefore, the gap values obtained here are reasonable, with a small uncertainty. It should be mentioned that a gap structure with reversal sign is necessary to explain our data, while the details of the gap function cannot be distinguished although the $d_{x^2-y^2}$ -wave symmetry was accepted here (for example, d_{xy} -wave symmetry [39] can explain our data as well). It is worth noting that the ZBCP observed here is also consistent with the recent calculations for an unconventional s-wave pairing state with an internal π phase ($s\pi$ state), that is, nodeless gaps on each band but with the opposite signs on different bands [15, 40]. Nonetheless, whether the ZBCP is a common feature of the oxypnictides still needs to be clarified by more experiments on various high-quality samples.

Another remarkable finding in the calculations presented in figure 2 is that the determined maximum gap Δ_0 can be divided into two groups, namely, a big gap of

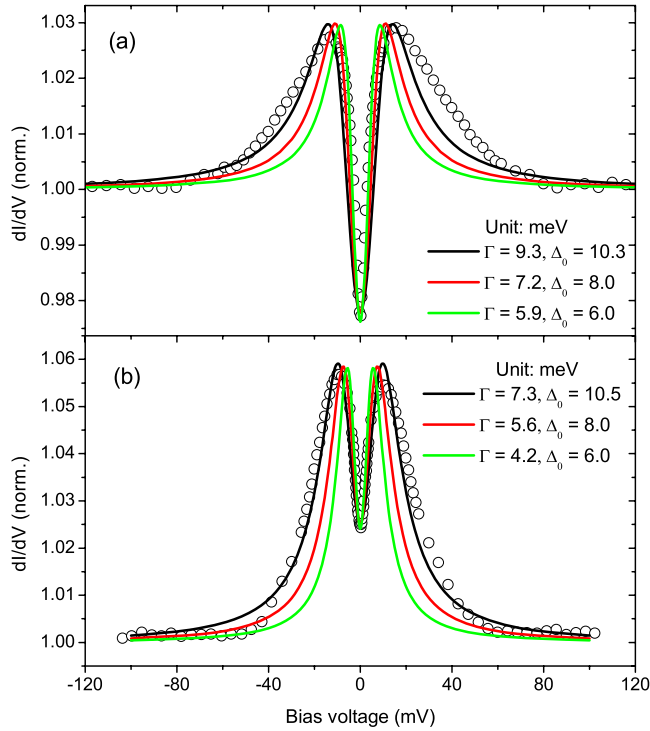


Figure 3. The fitting process using the extended BTK model with different broadening parameters. (a) and (b) correspond to the curves shown in figures 2(b) and (c), respectively.

10.5 ± 0.5 meV (figures 2(b)–(d)) and a small gap of 3.7 ± 0.4 meV (figures 2(e)–(h)), though the barrier strength Z and quasiparticle injecting angle α are random due to the diverse configuration of the grains in the polycrystalline samples. The small gap cannot be explained as a degradation of the sample surface since its value is distributed in a narrow range far below that of the big gap. It should be pointed out that these two distinct gap values were determined mainly by the peak positions of the measured spectra by considering the scattering- and thermal-induced broadening; thus the two-gap characteristic observed here does not depend on which model is applied. To get further insight into this point, we have measured the temperature dependence of these two gaps with distinct energy scales.

Figure 4 shows the temperature dependence of two types of spectra corresponding to the two different gaps as mentioned above. At lower temperatures, two coherence peaks can be seen clearly, accompanied by a low-energy depression of the quasiparticle density of states. With increasing temperature, the peaks are suppressed and smeared continuously, and finally the spectra become smooth featureless curves around T_c . These data were normalized (by the data of $T = 60$ K) in order to be compared with theoretical models, as shown in figures 5(a) and (b). It was found that all the data can be fitted very well to the extended BTK model mentioned above. Figure 5(c) shows two spectra measured at the same location but with different junction resistances and hence different barrier strengths. A similar gap value of about 10 meV was obtained from these spectra, indicating that the detected superconductivity does not depend on the junction resistance. Figure 5(d) summarizes the

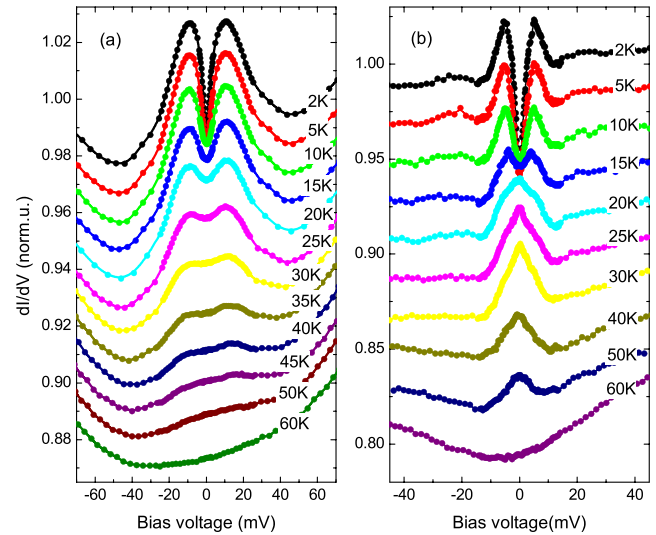


Figure 4. Conductance measured for various temperatures from 2 K to above T_c . (a) Large gap, (b) small gap. All the curves except the top one are offset downwards for clarity.

gap values obtained from figures 5(a) and (b). The temperature dependence of both the large gap and the small one can be described by the prediction of BCS theory. The gap value of $\Delta_0 = 10$ meV leads to $2\Delta_0/k_B T_c = 4.6$, a bit larger than the prediction of weak-coupling d-wave BCS theory. Moreover, both gaps are closed around T_c , reflecting the inter-band coupling existing in this material.

It is interesting to note that some spectra exhibit a two-gap feature similar to that of MgB_2 . As shown in figure 5(e), compared with the high-temperature spectrum measured around T_c , the low-temperature one has three distinct features: a ZBCP, two coherence peaks, and two symmetric humps at higher energy. In figure 5(f), we try to simulate the low-temperature spectrum after normalization according to the high-temperature one. In the calculation, a simple two-component BTK model was applied in which the normalized conductance is expressed by $G = w_1 G_1 + w_2 G_2$, where G_1 and G_2 are the conductances associated with the large gap and the small gap, respectively; w_1 and w_2 ($w_1 + w_2 = 1$) are the corresponding weights of these two gaps contributing to the total conductance. In order to reduce the number of fitting parameters, s-wave symmetry was assumed for the large gap while $d_{x^2-y^2}$ -wave symmetry for the small gap. It was found that the main features in the spectrum can be fitted very well. It should be emphasized that the s + d model adopted here is not claimed as evidence of s + d order parameters in the material but merely as a helpful model with which to fit the data. Although this is a rough approximation, it captures the main physics of multiple gaps and an order parameter with a sign change. Most interestingly, the gap values determined from the calculation are 11 and 4.5 meV, very close to the results obtained from the spectra measured at other locations.

Besides the sample of F-SmFeAsO which is focused on in this work, we have also measured the point-contact spectra of F-LaFeAsO [7] and F-NdFeAsO. In those samples, a more prominent ZBCP was often observed, while some confused

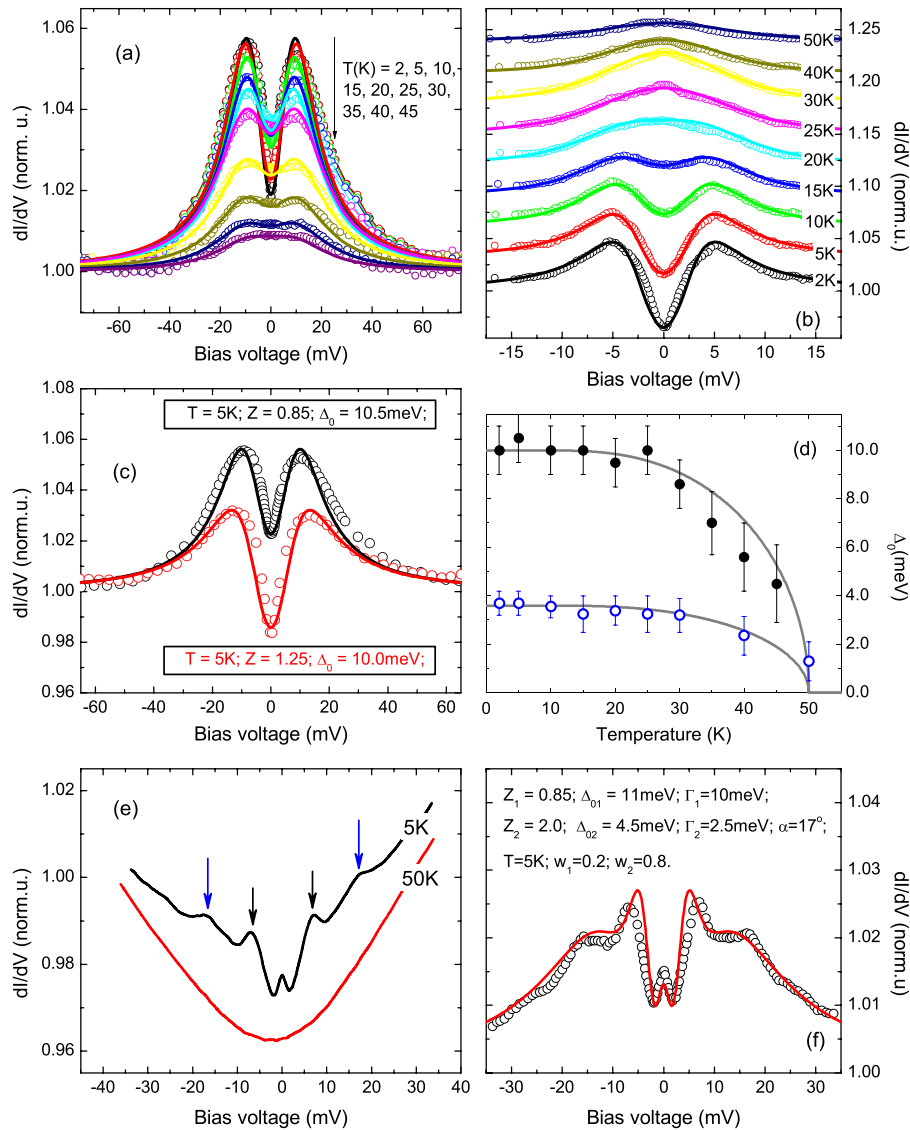


Figure 5. (a) and (b) Temperature dependence of normalized spectra corresponding to figures 4(a) and (b), respectively. The solid lines are theoretical calculations according to the extended BTK model. (c) Two spectra measured at the same position but with different junction resistance; the solid lines are theoretical calculations. (d) Temperature dependences of the gap values determined from fits as shown in (a) and (b); the solid lines are the prediction of BCS theory. (e) The spectra measured at low temperature and around T_c on the same position of the sample surface. (f) The low-temperature spectra after normalization according to the high-temperature one as shown in (e); the solid line is the theoretical fit.

backgrounds can also be observed occasionally compared with the data of F-SmFeAsO; thus quantitative analysis is more difficult. A most possible explanation for this difference is that the F-SmFeAsO studied here was prepared by a high-pressure technique while the other samples were synthesized at atmospheric pressure and hence are more fragile and loose. Therefore, there is often a space between adjacent grains, which can be seen clearly in the SEM images. Consequently, the tip can easily penetrate through the sample surface and rest on a pit, leading to multiple contacts between the tip and surrounding microcrystals. On the one hand, this increases the opportunities to detect a strong ZBCP if a sign reversal exists in the gap function. On the other hand, it will induce a more confused background due to the complicated interface effect. In this case, the gap values can still be estimated by using a

convenient method proposed by Dagan *et al* [41]. The method of data analysis consists simply in subtracting conductances measured in an applied field from the zero-field conductance. Since the maximum of the density of states at the gap value should be sensitive to an applied field, such subtraction will lead to a dip around the gap value in the subtracted spectra [41]. Using this method, we have estimated $\Delta_0 \approx 3.9$ meV for F-LaFeAsO, which is consistent with the results from both specific heat [6] and lower critical field [8].

In figure 6, we sum up the gap values of some 1111-phase Fe-based superconductors with different T_c , which were obtained from our point-contact spectroscopy measurements. The result for PrFeAsO_{0.89}F_{0.11} from NMR experiments [17] is also presented for comparison. It was noted that, for all these Fe-based superconductors, the superconducting gaps can

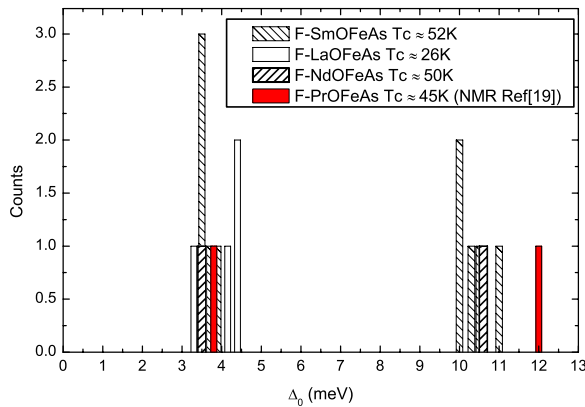


Figure 6. Statistical chart of the Δ_0 -values determined from the spectra measured at various positions on the sample surfaces. The vertical axis denotes the occurring times for a given gap value specified by the horizontal axis.

be divided into two groups with distinct energy scales centered at 4 meV and 11 meV, respectively. Therefore, the high T_c that can be achieved now is most possibly dominated by the big gap. However, the detailed gap structure is still an open issue until high-quality single crystals can be obtained.

4. Summary

In summary, we have studied point-contact spectroscopy measurements of the junctions built up between a normal metal tip and the newly discovered 1111-phase Fe-based layered superconductor $\text{SmFeAsO}_{0.9}\text{F}_{0.1}$. A zero-bias conductance peak was observed and demonstrated to be related to the sign reversal of the gap function. Two superconducting gaps with different energy scales were observed which depend on the temperature in a similar way as predicted by BCS theory. Our data present strong evidence that $\text{SmFeAsO}_{0.9}\text{F}_{0.1}$ is an unconventional superconductor with multiple gaps.

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