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Point-contact spectroscopy of iron-based layered superconductor LaO$_{0.9}$F$_{0.1-\delta}$FeAs

Lei Shan[(a)], Yonglei Wang, Xiyu Zhu, Gang Mu, Lei Fang, Cong Ren and Hai-Hu Wen

National Laboratory for Superconductivity, Institute of Physics and Beijing National Laboratory for Condensed Matter Physics, Chinese Academy of Sciences - P.O. Box 603, Beijing 100190, China

Abstract – We present point-contact spectroscopy data for junctions between a normal metal and the newly discovered F-doped superconductor LaO$_{0.9}$F$_{0.1-\delta}$FeAs (F-LaOFeAs). A zero-bias conductance peak was observed and its shape and magnitude suggest the presence of Andreev bound states at the surface of F-LaOFeAs, which provides a possible evidence of an unconventional pairing symmetry with a nodal gap function. The maximum gap value $\Delta_0 \approx 3.9 \pm 0.7$ meV was determined from the measured spectra, in good agreement with the recent experiments on specific heat and lower critical field.

Introduction. – Copper-based layered superconductors have attracted extensive attention due to their high superconducting transition temperature (T_c) and the underlying rich physics of strong electron correlation. Although $d_{x^2-y^2}$ pairing symmetry has been proved in cuprates [1], the mechanism of high-temperature superconductivity is still not settled down. In order to open a new path to clarify this issue, much efforts have been devoted to seeking new transition-metal–based superconductors other than cuprates. Recently, superconductivity was observed in the Fe-based layered material La[O$_{1-x}$F$_x$]FeAs ($x = 0.05$–0.12) with transition temperatures $T_c \approx 26$ K [2]. Immediately after this exciting discovery, Ren *et al.* reported that T_c could reach 55 K in Sm[O$_{1-x}$F$_x$]FeAs [3]. Superconductivity at 25 K was also found in Sr-substituted samples La$_{1-x}$Sr$_x$OFeAs without F-doping [4]. This provides a new chance to understand high-T_c superconductivity in non-cuprate systems. Recent experiments indicated that La[O$_{1-x}$F$_x$]FeAs has a low carrier density [5,6] with strong electron-electron correlation [7], and its superconducting regime occurs in close proximity to a long-range–ordered antiferromagnetic ground state [8]. All these properties are much like that of high-T_c copper oxides and hence a novel superconductivity is anticipated. Theoretically, unconventional superconductivity beyond *s*-wave pairing was predicted for Fe-based

superconductors by recent calculations [9–14]. Most interestingly, a nonlinear magnetic-field dependence of the electronic specific-heat coefficient $\gamma(H)$ has been found for LaO$_{0.9}$F$_{0.1-\delta}$FeAs (F-LaOFeAs) in the low-temperature limit, indicative of an unconventional pairing symmetry with a nodal gap function [15], which is in good agreement with the linear temperature dependence of the lower critical field (H_{c1}) observed on the same material [16]. However, the possibility of a novel pairing symmetry in this new superconductor needs to be verified by more experiments especially by phase-related measurements.

In this letter, we report the study of point-contact spectroscopy on the superconducting F-LaOFeAs as a function of both temperature and magnetic field. We observed a distinct zero-bias conductance peak in the conductance-voltage characteristics of the point-contact junctions, suggesting that F-LaOFeAs has an unconventional pairing symmetry with a nodal gap. The determined maximum gap value is consistent with the results of specific heat and lower critical field.

Experiment. – The polycrystalline samples of F-LaOFeAs used here were synthesized by using a two-step solid-state reaction method. The superconducting transition temperature T_c defined as the onset of the drop in resistivity was 27 K with a transition width of $\Delta T_c \approx 3$ K (10%–90% of normal state resistivity). The detailed information about the synthesization is

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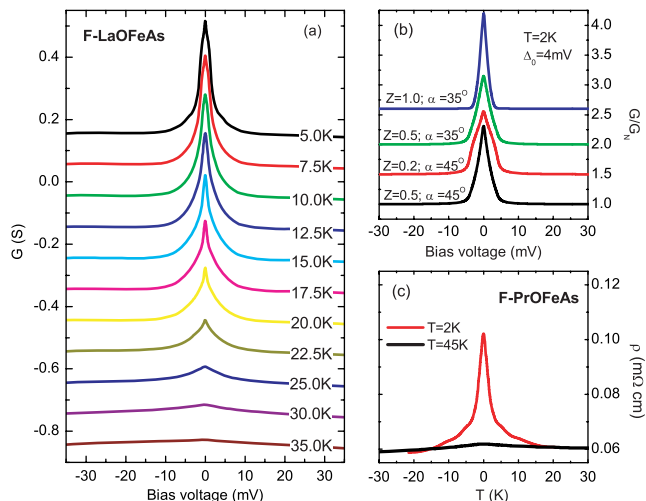
[(a)]E-mail: lshan@aphy.iphy.ac.cn

Fig. 1: (a) Conductance-voltage (*G-V*) characteristics of F-LaOFeAs measured at various temperatures up to above T_c for $H = 0$ T. All the curves except the top one are offset downwards for clarity. (b) Calculated spectra for a nodal superconductor according to the extended BTK model described in text. All the curves except the lowest one are offset upwards for clarity. (c) *G-V* curves of F-PrOFeAs measured at 2 K and 45 K (above T_c), respectively.

elaborated in a recent paper [15]. The point-contact junctions are prepared by carefully driving the Pt/Ir alloy or Au tips towards the sample surface which is polished by fine sand paper and cleaned by ultrasonic beforehand. The tip's preparation and the details of the experimental setup were described elsewhere [17]. Typical four-terminal and lock-in techniques were used to measure the conductance-voltage (dI/dV - V or G-V) characteristics. Each measurement is comprised of two successive cycles, to check the absence of heating-hysteresis effects.

Results and discussion. – As shown in fig. 1(a), fig. 3(a) (raw data) and fig. 4(d) or (e) (normalized spectra), most of the measured spectra have a zero-bias conductance peak (ZBCP) though the magnitude and shape of the peak varies from one position to another on the sample surface. The approximate percentage of the occurrence of ZBCP is more than 50% for the sample we have studied. Many other spectra are featureless and thus maybe come from the non-superconducting region on the sample surface (the superconducting volume is above 80% for the bulk of the sample). Moreover, such ZBCP was observed in the spectra with various junction resistances from 5 Ω to 80 Ω and is not related to the effect of a large contact spot. Blonder *et al.* [18] 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 parametrized by a dimensionless number Z which describes the crossover from a metallic to an ideal tunnel junction behavior by $Z = 0$ to $Z = \infty$. It was predicted that no sharp ZBCP could be observed for an *s*-wave superconductor/normal

metal junction with any barrier strength, furthermore, the normalized conductance (G/G_N) at zero bias cannot exceed 2 (the upper limit of the ideal Andreev reflection). This is obviously not the case of our measurements, in which a distinct ZBCP was observed and its magnitude can exceed 2. However, this ZBCP can be reasonably associated to the surface Andreev bound states and is one of the unique features of a nodal superconductor [19], as demonstrated in the cuprate superconductors such as YBCO and LSCO [20].

Tanaka *et al.* [21–23] extended the BTK model to deal with the issue of an anisotropic *d*-wave superconductor. In this case, another parameter α (the angle between the quasiparticle injecting direction and the main crystalline axis) was introduced in addition to the barrier strength Z. Moreover, the isotropic superconducting gap Δ in the *s*-wave BTK model was replaced by an anisotropic gap with $d_{x^2-y^2}$ symmetry, *i.e.*, $\Delta = \Delta_0 \cos(2\theta)$ in which Δ_0 is the maximum gap. It was thus predicted that, for $Z > 0$ 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 only when the quasiparticle scattering is strong enough [24]. The calculated curves using this extended BTK model are presented in fig. 1(b). A ZBCP can be clearly seen in all curves for different α and Z though its shape and magnitude varies from one to another, which is qualitatively consistent with the measured spectra in this work.

It is necessary to consider other possible origins of the ZBCP observed here before it can be ascribed to the surface Andreev bound states of a nodal superconductor. First, a remarkable ZBCP often shows up if the intergrain Josephson coupling effect is in series with the point contact [17], or if the role of critical current becomes dominant due to the significant dissipation at the point-contact microconstriction [25]. However, these conjectures also favor two sharp dips besides the central ZBCP which were not observed here. Another mechanism for the ZBCP is related to magnetic and Kondo scattering coming from the magnetic impurities in or near the barrier [26]. However, a ZBCP due to magnetic impurities should be uncorrelated with the occurrence of superconductivity. In contrast, as shown in fig. 2, both the magnitude and the integrated spectral weight of ZBCP in the present experiment were found to build up rapidly just below 26 K, very close to the bulk T_c. In addition, the magnitude of the ZBCP caused by magnetic impurities should depend on temperature logarithmically [26], which is also inconsistent with our data. Furthermore, as presented in fig. 1(c), similar ZBCP has been observed on another newly synthesized Fe-based superconductor Pr[O$_{1-x}$F$_x$]FeAs (F-PrOFeAs) with $T_c = 42$ K (the measurement and data analysis is still in process). All these considerations suggest that the ZBCP observed here is most possibly related to some type of nodal gap function. In the scanning electron microscopic picture obtained on the sample surface, it was noted that there are many tiny crystalline stacks distributed
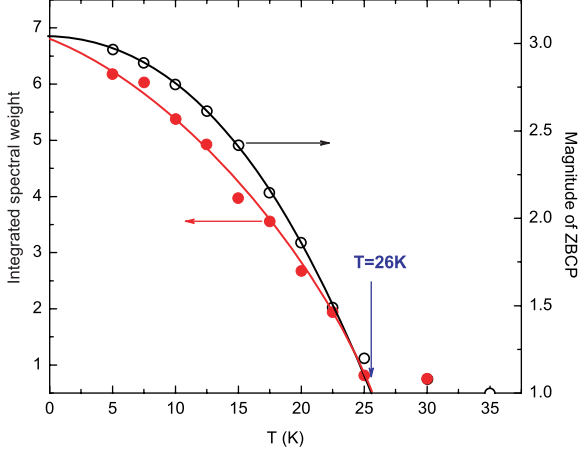
Fig. 2: Temperature dependence of the magnitude of ZBCP and the integrated spectral weight. Both parameters are obtained from the spectra shown in fig. 1(b) after normalization by the data of 35 K.

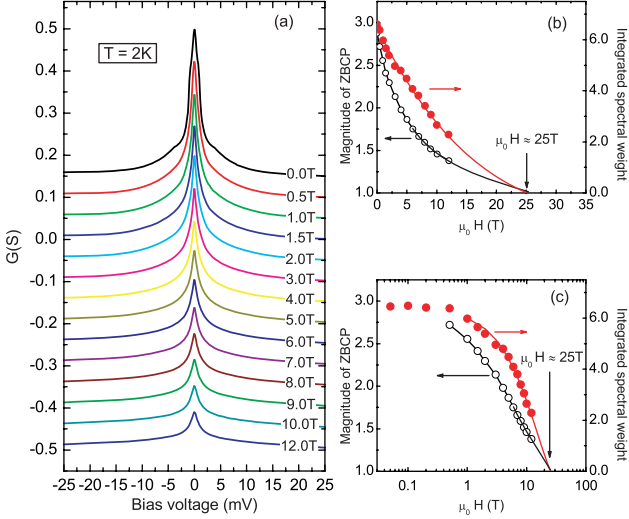Fig. 3: (a) G-V curves measured in various magnetic fields for $T = 2$ K. All the curves except the top one are offset downwards for clarity. (b) Field dependence of the magnitude of ZBCP and the integrated spectral weight. Both parameters are obtained from the spectra shown in (a) after normalization by the data of 35 K. (c) Same data as that in (b) while the horizontal axis is in logarithmic scale.

disorderly with finite space between them. Consequently, the tip is easy to penetrate through the sample surface and rests in a pit, leading to the contacts between the tip and the sides of the surrounded tiny crystals and hence increases the opportunities to detect ZBCP if a nodal pairing symmetry exists.

Figure 3(a) shows the evolution of the ZBCP with increasing field from 0 to 12 T at a fixed temperature of 2 K. ZBCP is suppressed continuously whereas survives up to the highest field in our measurements. We present in fig. 3(b) both the height and integrated spectral weight of the ZBCP. The same data are re-plotted in fig. 3(c) while the horizontal axis is in logarithmic scale. By extrapolating

these two parameters to zero independently, we can obtain consistently the local upper critical field (H_{c2}) of 25 T from both figs. 3(b) and (c). This value is much smaller than $H_{c2} > 50$ T estimated from resistivity measurements [5,6]. As mentioned in ref. [6], the upper critical field determined from resistivity reflects mainly the situation of $H \parallel a$-b plane since the Cooper pairs within the grains with this configuration will last to the highest field compared to other grains. Since our point-contact configuration is a local measurement, it probes the grains with particular orientations, so the determined H_{c2} should locate between $H_{c2}^{ab} \approx 56$ T and $H_{c2}^{c} \approx 6$ T by assuming an anisotropy $\Gamma = H_{c2}^{ab}/H_{c2}^{c} \approx 10$ [6], in agreement with the result in this work.

Dagan *et al.* [27] proposed a convenient method to determine the gap value of a nodal superconductor by considering that the peak in the density of states at the gap value should be sensitive to an applied field, while other structures should not be. The method of data analysis consists simply in subtracting conductances measured in an applied field, from the zero-field conductance. Structures that are not directly related to superconductivity are, in this way, eliminated [27]. When the subtraction procedure is applied to the data of fig. 3(a), as shown in fig. 4(a), very clear dips appear symmetrically around ± 4 mV and locate at the same bias for fields up to 5 T (indicated by two vertical lines). This is very similar to the case discussed in ref. [27] (except that there is a field-dependent background here), allowing a precise determination of the gap values. Figure 4(b) shows such dip structure for various temperatures and the gap values determined from these dips are presented in fig. 4(c). Since the thermal smearing effect plays a more important role at higher temperatures, the results of higher temperatures shown in fig. 4(c) may be overestimated to some extent. Even though, the decay of the gap value with increasing temperature is obvious.

Figure 4(d) shows the estimation of the gap values by fitting the normalized spectra (measured at different locations) to the extended BTK model with a $d_{x^2-y^2}$ gap function. The obtained Δ_0 varies in a range of 1 mV, indicating a certain degree of inhomogeneity on the sample surface. Two symmetric slight shoulders have also been observed in some spectra (refer to figs. 1(a) and (c)), which cannot be fitted with the single-gap model used here. This may be related to another gap opening on other Fermi surfaces though it contributes little to the measured spectra. It should be mentioned that our data support a nodal gap function of F-LaOFeAs while cannot distinguish between a d-wave and a p-wave. Since the superconducting phase of F-LaOFeAs is located close to the ferromagnetic phase [2], it is possible that the spin triplet pairing, such as the p-wave pairing symmetry, is favored in this material. In fact a very similar ZBCP was observed in Sr_2RuO_4 single crystal with spin triplet superconductivity [28]. Figure 4(e) shows the calculations with a p-wave gap function of $\Delta = \Delta_0 \sin \theta$, which is consistent with the experimental data as well as the
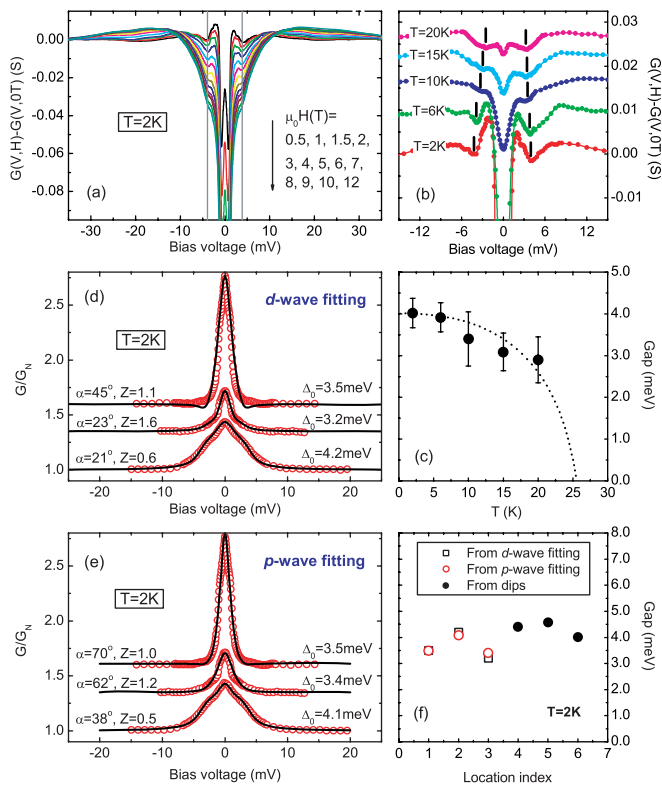
Fig. 4: (a) The conductance of various fields from fig. 3(a) after subtraction of the zero-field conductance. (b) The conductance at low field of 0.5 T or 1.0 T after subtraction of the zero-field conductance for various temperatures. The positions of the dips in each curve are indicated by short bars. (c) Maximum superconducting gap determined from the data presented in (b). (d), (e) Fitting the normalized spectra (open circles) obtained at various locations on the sample surface to the extended BTK model with *d*-wave and *p*-wave pairing symmetries, respectively. (f) Summing-up of the maximum gap values of various locations on the sample surface determined both by dips and fits.

d-wave fits. In both approaches, the nodal characteristic and the maximum value of the gap function of F-LaOFeAs can be confirmed consistently. Most recently, NMR data present strong evidence of singlet pairing in $PrFeAsO_{0.89}F_{0.11}$ [29], thus the singlet pairing symmetry such as the *d*-wave seems to be more possible than the *p*-wave pairing for the sample studied here. In fig. 4(f), we can see that the gap values determined both by fitting procedure and from the dip structure are well consistent with each other, which can be summed up as $\Delta_0 = 3.9 \pm 0.7$ meV. This result is in good agreement with $\Delta_0 = 3.4 \pm 0.5$ meV from our specific-heat measurement [15] and $\Delta_0 = 4.0 \pm 0.6$ meV from our H_{c1} measurement [16] on the same material. Thus, we can calculate the Bardeen-Cooper-Schrieffer coherence length $\xi_{BCS} = \hbar\nu_F/\pi\Delta_0$, where the in-plane Fermi velocity $\nu_F^{ab} = 1.30 \times 10^7$ cm/s [30]. Using $H_{c2}^c \approx 6$ T mentioned above [6], we can calculate the Ginzburg-Landau coherence length $\xi_{GL}^{ab} = \sqrt{\phi_0/2\pi H_{c2}^c}$ with ϕ_0 the flux quantum. The obtained

$\xi_{BCS} = 70$ Å is in good agreement with the calculated $\xi_{GL}^{ab} = 74$ Å within the experimental errors. We also estimated the mean free path $l \approx 100$ Å from resistivity by using $k_F l = hc/e^2\rho$ (where $k_F \simeq m\nu_F/\hbar$, $c = 8.7$ Å is the lattice constant along the normal to the FeAs plane, and $h/e^2 = 26$ KΩ is the quantum resistance). These estimations self-consistently suggest that F-LaOFeAs is in the moderate clean limit, which is different from the dirty-limit case of the electron-doped cuprates with similar T_c values [31]. It is also the reason why a clear ZBCP can be observed in this work and the Δ_0 can be derived properly from the specific-heat data [15] for F-LaOFeAs.

Conclusion. – In summary, we have studied point-contact spectroscopy of the junctions built up between a normal metal tip and the newly discovered Fe-based layered superconductor $LaO_{0.9}F_{0.1-\delta}FeAs$. A distinct zero-bias conductance peak was observed and can be related to the surface Andreev bound states. Our data present a possible evidence that $LaO_{0.9}F_{0.1-\delta}FeAs$ is a nodal superconductor with a maximum gap of 3.9 ± 0.7 meV, which is in good agreement with our recent specific heat and H_{c1} data.

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