Andreev spectroscopy of $LaFeAsO_{0.9}F_{0.1}$

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Current-voltage characteristics of Andreev contacts in $LaFeAsO_{0.9}F_{0.1}$ have been measured using the break-junction technique. The contacts were prepared at $T=4.2$ K from polycrystalline samples and exhibit superconductor/normal conductor/superconductor-type behavior due to multiple Andreev reflections. Two sets of subharmonic gap structures were detected indicating the existence of two distinct superconducting gaps: $\Delta_L=(5.5\pm1)$ meV and $\Delta_S=(1\pm0.2)$ meV.

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The discovery of superconductivity in the rare-earth oxypnictides RFeAsO$_{1-x}$F$_x$ (R=La, Nd, Sm, and Pr) has triggered an enormous amount of research activities (see, e.g., Refs. 1–28). The material exhibits a layered crystal structure with layers of FeAs separated by spacer layers, such as LaO, where the dopants are introduced, for example, by replacing some of the oxygen in LaFeAsO by fluorine. The parent compounds of the pnictides are poor metals with a spin-density wave (SDW) ground state,[22,23] and superconductivity emerges as soon as the orthorhombic distortion and the SDW magnetism is suppressed.[24,25] Band-structure calculations[2–4] show that in RFeAsO$_{1-x}$F$_x$ the energy bands crossing the Fermi level are formed by mainly the Fe and the As states while the R-O bands are far from the Fermi energy. Superconductivity is located in the FeAs layers and the LaO layers provide the charge reservoir when doped with F ions. Theoretical calculations[4–6] as well as some experimental observations[10,11,16,18,29] have led to the conclusion that the FeAs-based superconductors belong to the class of multiband superconductors which have been studied intensively since the original theoretical works.[30] It was found that the Fermi surface of oxypnictides consists of slightly warped tubular sections running along the $\boldsymbol{c}$ direction.[31] There are two hole sheets centered at the Γ point and two electron sheets centered at the Brillouin-zone corner M.[31] These four bands in most cases can be considered as two effective two-dimensional bands.[10,11,16,18,19]

Valuable information about the magnitude and the symmetry of the order parameter in these compounds is now available.[7–21,29] The amplitude of the superconducting gap differs for hole and electron Fermi surfaces but the gap anisotropy on each cylinder is weak.[13,29,32] The relative sign of the order parameters remains undetermined. There are indications that conventional electron-phonon coupling is insufficient to explain superconductivity in the FeAs-based superconductors.[3,26] At the same time a nearly full iron isotope effect (^{54}Fe substituted for ^{56}Fe) in $Ba_{1-x}K_xFe_2As_2$ (Ref. 33) and a strong electron-phonon coupling of the Fe-breathing mode of $LaFeAsO_{1-x}F_x$ have been found.[34] A correlation between phonon frequencies of different oxypnictides and their critical temperatures T_c was reported.[35]

Andreev spectroscopy is one of the powerful tools to measure the superconducting energy gap. A variety of such measurements on superconductor/normal conductor (SN) contacts has been performed on oxypnictides RFeAsO$_{1-x}$F$_x$.[8,10,12,15–18] However, there is an ambiguity in the interpretation of Andreev-spectroscopy data concerning the symmetry, the magnitude, and the number of superconducting gaps. Complementary information can be gained by means of break-junction tunneling data.[36] Here, we present a systematic investigation of break-junction tunneling in $LaFeAsO_{0.9}F_{0.1}$ at $T=4.2$ K. The data imply superconductor/normal conductor/superconductor (SNS)-type behavior which is attributed to multiple Andreev reflections. In the current-voltage characteristics two sets of subharmonic gap structures (SGS) were detected. This observation implies the presence of two nodeless superconducting gaps which amount to $\Delta_L=(5.5\pm1)$ meV and $\Delta_S=(1\pm0.2)$ meV, respectively.

Polycrystalline $LaFeAsO_{0.9}F_{0.1}$ was prepared and characterized by powder x-ray diffraction, wavelength-dispersive x-ray spectroscopy, magnetization, transport, nuclear magnetic resonance, and muon spin-relaxation experiments.[23,25–28] We have used a break-junction technique[36] to generate contacts in our samples (more than 30 Andreev SNS point contacts have been studied at $T=4.2$ K). For these measurements, we have exploited three $LaFeAsO_{0.9}F_{0.1}$ samples (thin plates with typical dimensions $\approx 0.2\times0.5\times2.0$ mm^3) from the same synthesis procedure with critical temperatures $T_c=27.1$–28.1 K. Two current and two potential leads were attached to the sample by the In-Ga alloy. A microcrack in a sample was then generated at liquid helium temperature. The break-junction technique allows no detailed information about the junction geometry or intrinsic junction properties. At the same time there is a possibility to readjust the contacts easily with a micrometer screw at helium temperature. Thus the break-junction technique allows changing the junction properties during the measurements. An additional advantage of this technique is the existence of clean cryogenically cleaved surfaces used for the contact formation.

We exploit a standard modulation technique to measure the dI/dV and d^2I/dV^2 characteristics. A low-level ac modulation voltage (820 Hz) on potential leads of a contact is stabilized using a lock-in nanovoltmeter and a computer-controlled digital ac bridge (with a proportional-integral-

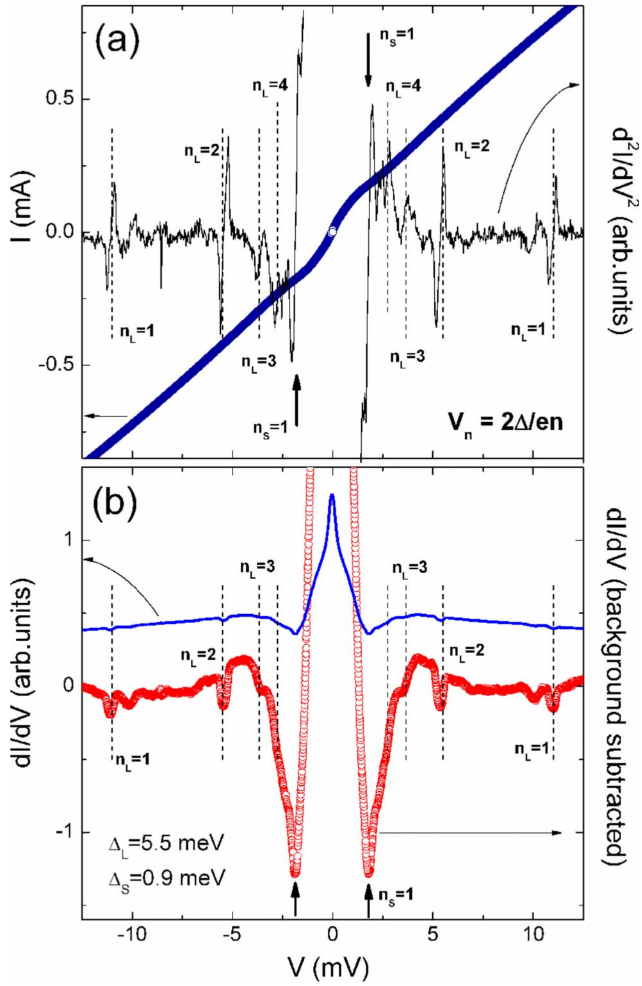

FIG. 1. (Color online) The current-voltage characteristics of an Andreev SNS point contact ($LaFeAsO_{0.9}F_{0.1}$, contact LOFA2D02) at T=4.2 K. (a) $I(v)$ (left) and d^2I/dV^2 (right) curve. (b) dI/dV curve as measured (left) and with partially suppressed background (right). Two series of subharmonic gap singularities at bias voltages $V_{nL}=2\Delta_L/en_L$ and $V_{nS}=2\Delta_S/en_S$ are detectable (Δ_L=5.5 meV and Δ_S=0.9 meV). The dashed lines display the expected bias voltages of the larger gap V_{nL} and arrows indicate V_{nS}.

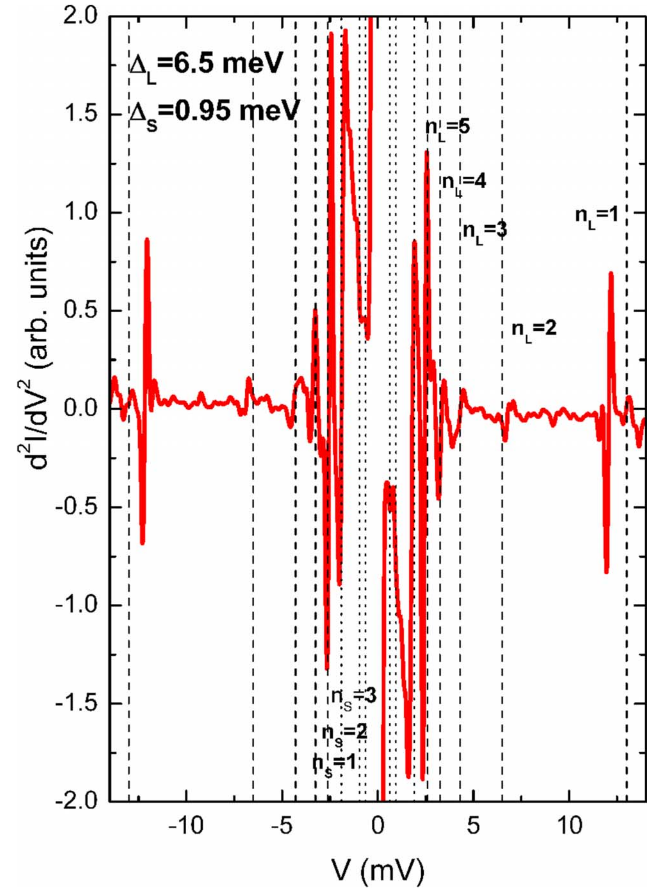

FIG. 2. (Color online) The d^2I/dV^2 characteristic of Andreev SNS point contact ($LaFeAsO_{0.9}F_{0.1}$, contact LOFA2D06) at T=4.2 K. Two series of subharmonic gap singularities at bias voltages $V_{nL}=2\Delta_L/en_L$ and $V_{nS}=2\Delta_S/en_S$ are detectable (Δ_L=6.5 meV and Δ_S=0.95 meV). The dashed lines display the expected bias voltages of the larger gap V_{nL} and the dotted lines indicate V_{nS}. A slight deviation of V_{1L} from the expected value is most probably caused by overheating at high current densities.

derivative feedback signal). The differential conductance of a contact is proportional to the amplitude of the ac feedback current through the contact.

In Fig. 1 representative data for the current-voltage characteristics $I(V)$, dI/dV, and d^2I/dV^2 of a $LaFeAsO_{0.9}F_{0.1}$ break junction are displayed. The data are typical for the SNS-type of behavior which is expected for clean classical Andreev microcontacts with excess-current characteristics.[37,38] The main features of the current-voltage characteristics of such point contacts comprise a pronounced excess current at low bias voltages and a SGS, showing sharp dips of a differential conductance dI/dV at bias voltages:[38]

$$V=\frac{2\Delta}{en} \quad \text{with} \quad n=1,2,\ldots. \tag{1}$$

Usually, this SGS is explained by multiple Andreev reflections at the point contact SN interfaces.[38] This type of structure was earlier observed in excess-current characteristics of SNS break junctions in high-T_c superconductors[39] and MgB_2 (Ref. 40), and must be distinguished from the SGS in the current-deficit characteristics of superconducting quantum point contacts (SQPC) with a relatively low interface transparency.[41,42] In the latter case the SGS exhibits a series of *maxima* in the differential conductance at bias voltages $V=2\Delta/en$.[43] It was shown that with increasing transparency the series of maxima turns into a series of *minima* in the conductance.[43] This final result for SQPC with high transparency and excess-current characteristics agrees with calculations of Kümmel *et al.*[38] for clean classical SNS contacts. In the present investigation we assume that the theoretical model of Kümmel *et al.*[38] is applicable to our break junctions with excess-current characteristics.

The quality of the SGS strongly depends on the ratio of the mean-free path l to the radius of the contact area, a.[38] For example, the normal resistance R of a ballistic Sharvin contact is given by[44]

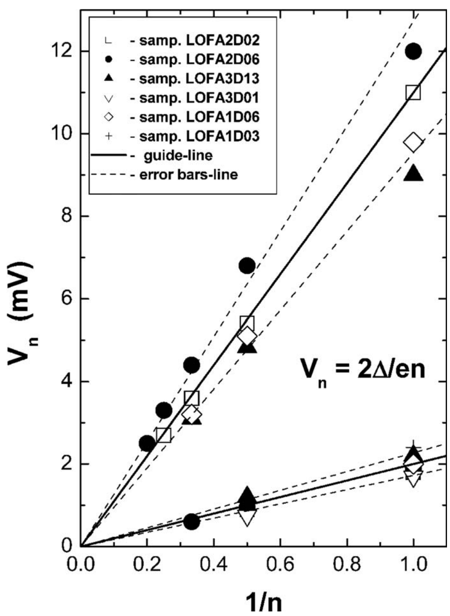

FIG. 3. The dependences of bias voltages $V_{nL}=2\Delta_L/en$ and $V_{nS}=2\Delta_S/en$ on (1/n) for investigated Andreev SNS contacts (cf. Table I).

$$R=(4/3\pi)(\rho l/a^2), \quad (2)$$

where ρ is the bulk resistivity of the metal. For $LaFeAsO_{0.9}F_{0.1}$ the product ρl roughly amounts to 1×10^{-10} $\Omega\,cm^2$ and $l \approx 1\times 10^{-6}$ cm.[8,45] From the typical normal resistance R of our contacts at $T=4.2$ K, which are in the range of 10–30 Ω, one can estimate the radius of the contact area a to $(1–2)\times 10^{-6}$ cm. Thus for our contacts $l \approx a$, which is in agreement with a limited number n of Andreev singularities in the subharmonic gap structure (Figs. 1–3).

The dI/dV characteristic in Fig. 1 exhibits a series of anomalies which were highlighted after a simple polynomial background function has been subtracted from the data. According to Eq. (1), the superconducting gap can be extracted quantitatively from the SGS in the current-voltage characteristics of Andreev SNS point contacts.[38] The observed anomalies imply the presence of two distinct gaps, i.e., a large gap $\Delta_L=5.5$ meV and a small gap $\Delta_S=0.9$ meV. In Fig. 1, the anomalies corresponding to the large gap are labeled by $n_L=1$, 2, and 3 (vertical dotted lines) and the dip for the small gap is labeled by $n_S=1$ (dashed line).

A better resolution is achieved if the second derivative d^2I/dV^2 is considered. This is shown for two representative examples from the series of 30 point contacts under study (Figs. 1 and 2). Here, the series of anomalies arising from a large gap Δ_L (clear anomalies for $n_L=1,2,3,4$) and from a small gap Δ_S ($n_S=1,2$) are displayed by dotted and dashed vertical lines, respectively. The extracted values of the dip voltages at $T=4.2$ K of the SGS in the current-voltage characteristics are summarized in Table I for six representative microcontacts. Depending on the particular SNS contact, up to five anomalies related to a large gap could be extracted and up to three anomalies could be extracted for a small gap.

A summary of the experimental data for the various microcontacts is given in Fig. 3 in which the dip voltages V_n are plotted versus the inverse number $1/n$ of the anomaly in the SGS. As suggested by Eq. (1), $V_n(1/n)$ forms a straight line through zero for each of the two gaps which strongly confirms the presence of two gaps. A slight deviation of V_{1L} from the expected value, which was observed in some cases (Figs. 2 and 3), is most probably caused by overheating at high current densities.

By applying Eq. (1) the experimentally determined voltage values V_n can be converted into superconducting gaps Δ_L and Δ_S for each of the investigated Andreev SNS contacts. The results of this analysis, shown in Table II, imply $\Delta_L = (5.5\pm 1)$ meV and $\Delta_S=(1\pm 0.2)$ meV. Table II also exhibits the superconducting transition temperatures measured *in situ* for the samples under study. This allows evaluating the ratio $2\Delta/k_BT_c$ which amounts to 3.52 for BCS superconductors. For the large gap, our data yield $2\Delta_L/k_BT_c = (3.6–5.6)$ meV thereby surpassing the BCS value. On the other hand, the ratio $2\Delta_S/k_BT_c$ for the small gap is less than one, i.e., it is significantly smaller than the BCS value, which resembles situation in MgB_2.[40]

The results of the present investigation reflect a multigap nature of the bulk superconductivity in $LaFeAsO_{1-x}F_x$. At the same time there are two less obvious reasons for the presence of a multigap structure in the current-voltage characteristics of the break junctions in $LaFeAsO_{0.9}F_{0.1}$. First, the multigap

TABLE I. Subharmonic gap structure in current-voltage characteristics of SNS contacts at $T=4.2$ K from Andreev-spectroscopy measurements on different microcontacts.

	Bias voltage V_n (mV) ($V_n=2\Delta/en$)							
	Large gap Δ_L					Small gap Δ_S		
Microcontact	$n_L=1$	$n_L=2$	$n_L=3$	$n_L=4$	$n_L=5$	$n_S=1$	$n_S=2$	$n_S=3$
LOFA2D02	11.0	5.4	3.6	2.7		1.8	0.9	
LOFA2D06	12.0	6.8	4.4	3.3	2.5	1.9	0.94	0.6
LOFA3D13	9.0	4.83	3.1			2.2	1.2	
LOFA3D01	9.1	4.3	2.7			1.7	0.75	
LOFA1D06	9.8	5.1	3.2			2.0		
LOFA1D03						2.4	1.18	

TABLE II. Superconducting gaps Δ_L and Δ_S in $LaFeAsO_{0.9}F_{0.1}$ at $T=4.2$ K from Andreev-spectroscopy measurements. Our experimental results derived at different microcontacts are compared with theoretical data in Ref. 6 as well as experimental data on $Ba_{1-x}K_xFe_2As_2$ (Refs. 20 and 21).

Microcontact	T_c (K)	Δ_L (meV)	Δ_S (meV)	$2\Delta_L/kT_c$	$2\Delta_S/kT_c$
		Our experiment			
LOFA2D02	27.1 (mid)	5.5	0.9	4.71	0.77
LOFA2D06	27.1 (mid)	6.5	0.95	5.56	0.81
LOFA1D06	27.7 (mid)	5.0	1.0	4.2	0.84
LOFA1D03	27.7 (mid)		1.2		0.93
LOFA3D13	28.1 (mid)	4.7	1.1	3.9	0.91
LOFA3D01	28.1 (mid)	4.3	0.8	3.55	0.66
		Theory[a]			
LOFA (at optimal doping)	47	7.9	2.1	3.9	1.04
		Experiment ($Ba_{1-x}K_xFe_2As_2$)			
Ref. 20	32	9	1.5	6.5	1.1
Ref. 21	23–27	9–11	2–5		

[a]Reference 6.

structure might be due to very strong inhomogeneities of the samples under study. We argue that the good reproducibility of Δ_L and Δ_S in the investigated Sharvin contacts (Table II) excludes strong inhomogeneities in our $LaFeAsO_{0.9}F_{0.1}$ samples. This is confirmed by local probe studies by means of μSR, Mössbauer, and NMR on samples from the same batches.[23,25,26] Second, there might be a suppression of the superconducting gap at the surfaces within the crack, which causes the formation of a superconductor-normal metal-constriction-normal metal-superconductor (SNcNS) contact. A comprehensive theoretical analysis of multiple Andreev reflections in SNcNS contacts[46] shows the presence of four SGS at bias voltages $V_n=2\Delta_L/en$, $V_n=2\Delta_S/en$, $V_n=(\Delta_L+\Delta_S)/en$, and $V_n=(\Delta_L-\Delta_S)/en$ for this kind of junction. In contrast, our data exhibit only two distinct SGS. If we assume that these SGS correspond to $V_n=(\Delta_L+\Delta_S)/en$ and $V_n=(\Delta_L-\Delta_S)/en$, we get $\Delta_L=6.5$ meV and $\Delta_S=4.5$ meV, which is suggested in Refs. 4 and 5. This scenario, however, contradicts the experimentally observed negative peaks (minima) in the dynamic conductance (Fig. 1) since a multiple Andreev-reflections process with $V_n=(\Delta_L-\Delta_S)/en$ should produce *positive* peaks of dynamic conductance.[46] Note that the negative anomalies found in our data are typical for multiple Andreev reflections in clean classical SNS contacts with excess-current characteristics.[38] Hence, we have to conclude that only assumption of multigap bulk superconductivity fits the experimental results of the present study.

The superconducting parameters from Refs. 6, 20, and 21 are added to Table II for comparison. In accordance with a two-band model accepted in Ref. 6 the intraband coupling in one of the bands is very weak and superconductivity in this band is induced by the "driving" band through the interband coupling. As a result the zero-temperature gap ratio $2\Delta_S/k_BT_c$ is smaller than the weak-coupling BCS value ~ 3.5 (Table II). At the same time the intraband coupling in a driving band can be quite significant.[47]

We note that the sharp line shape of the Andreev singularities composing the SGS (Figs. 1 and 2) points to nodeless gaps Δ_L and Δ_S (see also Ref. 48). This conclusion is supported by the comparison of experimental data with the theoretical results of Ref. 49 for an *s*-wave superconductor with a nodeless gap. Figure 4 presents both experimental and theoretical plots of the derivatives of the dynamic conductance d^2I/dV^2 vs the normalized bias voltage eV/Δ. Both data sets display rather *symmetric* behavior (see also the structures for $n_L=1$ in the d^2I/dV^2 characteristics in Figs. 1 and 2). In the

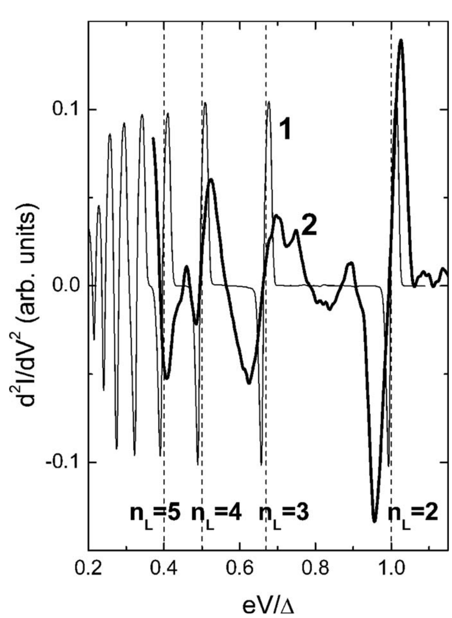

FIG. 4. The derivative of the dynamic conductance d^2I/dV^2 of Andreev SNS contact plotted as a function of normalized bias voltage eV/Δ. The theoretical curve for isotropic gap (*s* wave), after Ref. 49 (curve 1), experimental curve for the contact LOFA2D02 (curve 2).

case of a *d*-wave superconductor a strong asymmetry in the form of the Andreev singularities develops due to the presence of nodes in $\Delta(\varphi)$.[49]

In several cases an additional fine structure in the current-voltage characteristics of the break junctions was observed (Fig. 1). This is probably an indication that the two-band model might be insufficient.[50] A detailed conclusion about possible extensions of the two-band model applied for the data analysis, however, cannot be deduced from the conductivity measurements at hand.

In conclusion, current-voltage characteristics in polycrystalline $LaFeAsO_{0.9}F_{0.1}$ have been measured using the break-junction technique. The contacts exhibit SNS-type behavior due to the multiple Andreev reflections. Two sets of SGS were detected indicating the existence of two distinct superconducting gaps: $\Delta_L=(5.5\pm1)$ meV and $\Delta_S=(1\pm0.2)$ meV

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