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Point-contact spectroscopic studies on normal and superconducting AFe_2As_2-type iron pnictide single crystals

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Abstract

Point-contact Andreev reflection spectroscopy is applied to investigate the gap structure in iron pnictide single-crystal superconductors of the AFe_2As_2 (A = Ba, Sr) family ('Fe-122'). The observed point-contact junction conductance curves, $G(V)$, can be divided into two categories: one where Andreev reflection is present for both $(Ba_{0.6}K_{0.4})Fe_2As_2$ and $Ba(Fe_{0.9}Co_{0.1})_2As_2$, and the other with a $V^{2/3}$ background conductance universally observed, extending even up to 100 meV for $Sr_{0.6}Na_{0.4}Fe_2As_2$ and $Sr(Fe_{0.9}Co_{0.1})_2As_2$. The latter is also observed in point-contact junctions on the nonsuperconducting parent compound $BaFe_2As_2$ and superconducting $(Ba_{0.6}K_{0.4})Fe_2As_2$ crystals. Mesoscopic phase-separated coexistence of magnetic and superconducting orders is considered to explain distinct behaviors in the superconducting samples. For $Ba_{0.6}K_{0.4}Fe_2As_2$, double peaks due to Andreev reflection with a strongly sloping background are frequently observed for point contacts on freshly cleaved c-axis surfaces. If normalized using a background baseline and analyzed using the Blonder–Tinkham–Klapwijk model, the data show a gap size of ~3.0–4.0 meV with $2\Delta_0/k_BT_c \sim 2.0$–2.6, consistent with the smaller gap size reported for the LnFeAsO family ('Fe-1111'). For the $Ba(Fe_{0.9}Co_{0.1})_2As_2$, the $G(V)$ curves typically display a zero-bias conductance peak.

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

1. Introduction

The recently discovered iron-based superconductors [1] have emerged as a whole new family of high temperature superconductors, which are complimentary to the study of cuprates. They have attracted intensive studies in the community focusing on their physical properties and potential applications. Reminiscent of cuprates, the antiferromagnetic (AFM) phase in the parent compounds is suppressed with chemical doping or higher pressure, and it is proposed that superconductivity in iron pnictides may originate from AFM fluctuations [2]. It is pointed out that there are also vital differences in that the AFM ground state in the cuprates and Fe pnictides are Mott-insulator and metallic, respectively. Soon after the discovery of Fe pnictides, ARPES and quantum oscillation measurements have revealed disconnected hole-

like bands around the $\Gamma(0,0)$ point and electron-like bands around the $M(\pi,\pi)$ point on the FS [3–7]. The order parameter (OP) symmetry in the cuprates has been determined to be $d_{x^2-y^2}$, while it is still not known for the iron pnictides. A first step in unveiling the mechanism of the new family of superconductors is to determine the pairing symmetry. An extended s-wave gap structure with a sign reversal ($s\pm$) on different FSs is proposed by Mazin *et al* [2] where the hole-like and electron-like bands are fully gapped and have a π phase shift in the superconducting state. A fully gapped state has been confirmed by different experimental techniques such as ARPES [3–6], penetration depth [8], μSR [9, 10], Hc_1 [11] and specific heat measurements [12]. However, a universal power law rather than exponential behavior is observed in the London penetration depth measurement for $RFeAsO_{0.9}F_{0.1}$ (R = Nd, La) [13], and $Ba(Fe_{1-x}Co_x)_2As_2$ at various doping levels [14, 15]. The Hebel–Slichter coherent peak is absent in NMR measurements [16, 17] and it is argued that is a natural consequence of the extended $s\pm$ model in the superconducting state [18].

Point-contact Andreev reflection spectroscopy (PCARS) is a powerful tool to investigate the gap structure and OP symmetry in superconductors. A good example is the case of MgB_2, where two gaps, originating from σ and π bands, are clearly present in the point-contact spectra, and also through directional PCARS measurements, the gap dependencies on temperature and magnetic field are systematically revealed [19–22]. Two of the authors successfully applied PCARS to reveal the $d_{x^2-y^2}$ symmetry of the superconducting OP in $CeCoIn_5$ [23]. Shortly after the discovery of the superconducting LnFeAsO (Fe-1111) family, some PCARS measurements have been carried out, but the results are not yet conclusive, partly due to the polycrystalline nature of the samples. Chen *et al* report a conventional BCS-like superconducting gap with $2\Delta_0/k_BT_c \sim 3.7$ for $SmFeAsO_{0.85}F_{0.15}$ [24], while multiple gaps are claimed by other groups with different detailed structures in the initial stage [25–29]. Among those who claim multiple gaps, some merging agreements are being reached where $2\Delta_1/k_BT_c \sim 2$–3 and $2\Delta_2/k_BT_c \sim 7$–9. In this paper, we apply PCARS to different AFe_2As_2-type superconducting single crystals to elucidate their gap structure and OP symmetry.

2. Experiment

AFe_2As_2-type (Fe-122) parent compound and various superconducting single crystals are grown out of FeAs flux by a high temperature solution method, such as parent compound $BaFe_2As_2$ ($T_N \sim 135$ K), electron-doped superconducting $A(Fe_{0.9}Co_{0.1})_2As_2$ (A = Ba or Sr, $T_c \sim 22$ K) [30], and hole-doped superconducting $(Ba_{0.6}K_{0.4})Fe_2As_2$ ($T_c \sim 37$ K) and $(Sr_{0.6}Na_{0.4})Fe_2As_2$ ($T_c \sim 36$ K) [31]. Each family of the single crystals comes exclusively from one single group source. All the crystals have natural c-axis facets and are cleaved in the air to expose fresh and shiny surfaces before mounting on the sample holder. A sharp Au tip prepared by electrochemical etch [32] is engaged onto the sample surface with a conventional differential micrometer mechanism [33]. The contact between the Au normal metal and Fe-122 crystals is made after cooling to ~2 K. The junction conductance $dI/dV = G$, as a function of the bias voltage V, is recorded by the standard ac lock-in technique, where the superconductor is always biased positively. Contacts may be lost due to vibration or instability in thermal contraction. Here we report data obtained after dozens of contacts have been made on each crystal, and some with changing temperature up to the bulk T_c and magnetic field up to 9 T.

3. Results and discussions

Two different categories of $G(V)$ curves are observed for point contacts on these Fe-122 superconducting crystals: (1) for $(Ba_{0.6}K_{0.4})Fe_2As_2$ and $Ba(Fe_{0.9}Co_{0.1})_2As_2$, Andreev reflection signals are observed but distinct from each other; and (2) for $(Sr_{0.6}Na_{0.4})Fe_2As_2$ and $Sr(Fe_{0.9}Co_{0.1})_2As_2$, in the absence of Andreev reflection, a universal power law behavior is seen. The latter is also observed for the point contact on the nonsuperconducting parent compound $BaFe_2As_2$ and superconducting $(Ba_{0.6}K_{0.4})Fe_2As_2$ crystals. Results and discussions are presented here for each case separately.

3.1. $(Ba_{0.6}K_{0.4})Fe_2As_2$

For the $(Ba_{0.6}K_{0.4})Fe_2As_2$ crystals, representatives of the most frequently observed $G(V)$ curves at low temperatures (~2 K) are shown in figure 1. The prominent features are the two peaks at ~±3 meV and a strongly sloping background. A hump structure can also be noticed around ±15 meV as indicated by arrows, and a small conductance asymmetry is systematically observed. Similar broad backgrounds and asymmetries are reported in the recent point-contact measurement on $SmFeAsO_{0.8}F_{0.2}$, where the sloping background is claimed to disappear around the Neel temperature (~140 K) of the parent compound [29]. The temperature dependence as shown in figure 2(a) verifies that the low-bias conductance enhancement is due to Andreev reflection. Although the junction resistance changes with temperature, the Andreev reflection signal disappears only above the bulk T_c, giving confidence that we probe the bulk gap. The sloping background survives above T_c so it does not originate from superconductivity and must be due to some other scattering mechanism.

Since the $(Ba_{0.6}K_{0.4})Fe_2As_2$ material is known to be reactive in air, we try to minimize exposure time between cleavage and cooldown. The usual time is about 30 min. We investigate the effect of a 1 week air exposure as shown in figure 2(b). The Andreev reflection signal is lost below 16.6 K, lower than that of the bulk. This indicates the air exposure degrades the surface and suppresses the superconductivity and that cleavage minimizes the surface degradation. We note the sloping background does not change with the air exposure.

For the gap analysis, we normalize the conductance for the cleaved surfaces to the extrapolated baseline as shown in figure 3(a). The normalized data are then analyzed by the single-gap Blonder–Tinkham–Klapwijk (BTK) model [34, 35]. The best fit for the energy gap is ~3.0–4.0 meV, so $2\Delta_0/k_BT_c \sim 2.0$–2.6, smaller than the BCS weak

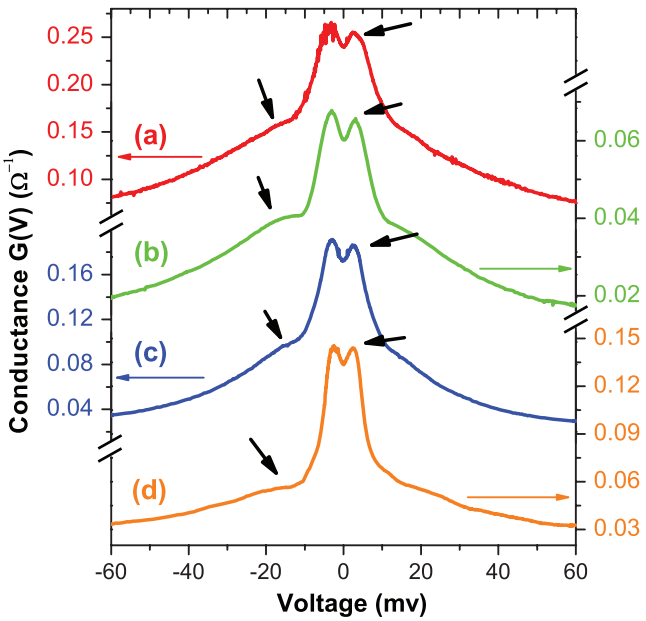

Figure 1. Differential conductance spectra $G(V)$ for the Au/$(Ba_{0.6}K_{0.4})Fe_2As_2$ point-contact junctions at low temperatures $T \sim 2$ K. The peak and hump structures are indicated by arrows nearby.

coupling ratio of 3.52. This value is comparable with the smaller gap size probed by PCARS on polycrystalline samples of $LaFeAsO_{1-x}F_x$ [26], $NdFeAsO_{0.9}F_{0.1}$ [27], and $SmFeAsO_{0.8}F_{0.2}$ [29]. We stress that our materials are 'Fe-122' single crystals, and we probe in the c-axis orientation. This may account for us not observing the larger gap as follows: an ARPES study on $(Ba_{0.6}K_{0.4})Fe_2As_2$ reveals an isotropic but FS sheet-dependent gap structure, where the β band has an averaged gap size $\sim 5.8 \pm 0.8$ meV with $2\Delta/k_BT_c \sim 3.6 \pm 0.5$ and the α, γ and δ bands have comparable gap sizes around 11–13 meV with $2\Delta/k_BT_c \sim 7.0$–8.0 [4]. Considering the FSs from band structure calculations, the α, γ and δ bands are highly two-dimensional with cylindrical shapes while the β band is strongly three-dimensional due to the $d_{3z^2-r^2}$ component [36]. The small gap observed here may correspond to the 3D β band. Because the Fermi velocity on the α, γ, and δ bands is mostly in the ab plane and perpendicular to the c-axis, these bands contribute a relatively small spectral weight for current flowing in the c-axis, and the coherent peaks from these larger-gap bands are almost absent in PCARS, similar to the case of MgB_2 [20]. Both gaps should then be observable if junction current is flowing within the ab plane. Szabó *et al* report two superconducting gaps for PCARS measurement in the ab plane with $2\Delta_1/k_BT_c = 2.5$–4 and $2\Delta_2/k_BT_c = 9$–10 [37]. However, Andreev reflection is totally absent for their point-contact junctions in c-axis. This is distinct from our results and may be due to the sample difference.

The elastic and inelastic electron mean free paths, l_{el} and l_{in}, respectively, are not known for $(Ba_{0.6}K_{0.4})Fe_2As_2$ single crystals. However, it is generally believed that they are bad metals and l_{el} could be a few tens of nanometers, making it difficult to form a contact in the Sharvin ballistic limit, (contact diameter $d < l_{el}$). For the contact in the diffusive regime ($l_{el} < d < \sqrt{l_{el}l_{in}}$), dips may arise from the extra finite resistance of the superconducting electrode when the junction current at a finite voltage bias exceeds the critical current [38]. The humps in figure 1 are a possible signature of shallow dips, possibly dimmed by the sloping background. As the point contact is moved further away from the ballistic regime, the dip structure becomes more pronounced and a zero-bias conductance peak (ZBCP) is observed, rather than the usual double-peak. This is likely the case as shown in figures 4(a)

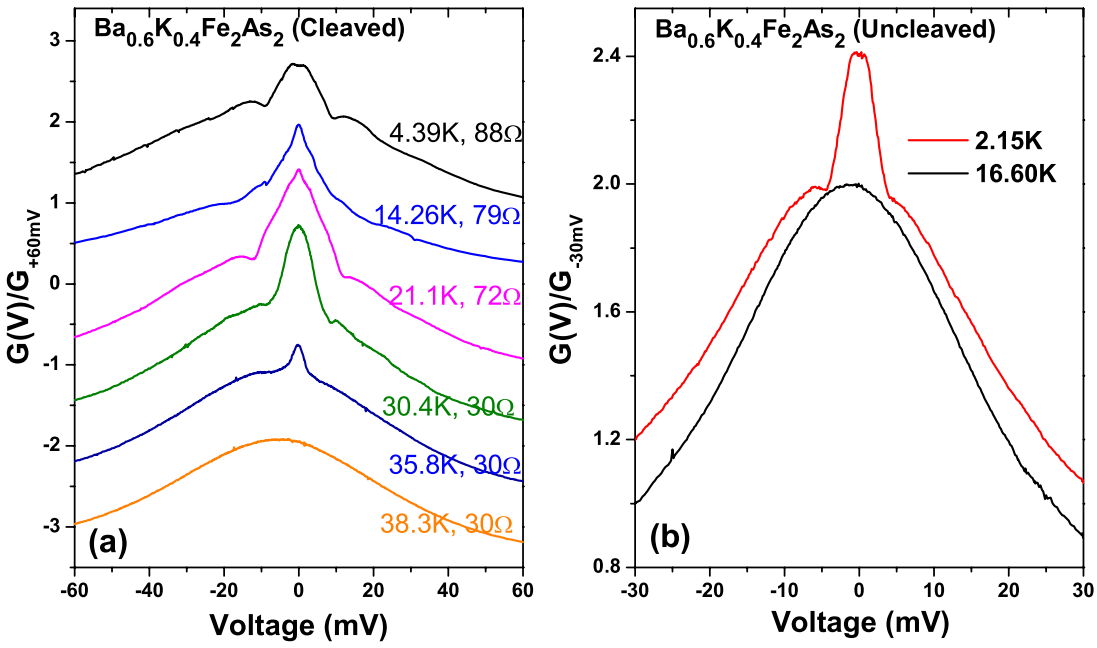

Figure 2. Temperature dependence of the conductance curves $G(V)$ for the Au/$(Ba_{0.6}K_{0.4})Fe_2As_2$ point-contact junctions on a (a) fresh-cleaved surface (the junction resistance changes with temperature due to instability of the contact) and (b) uncleaved surface. The curves are vertically shifted for clarity.

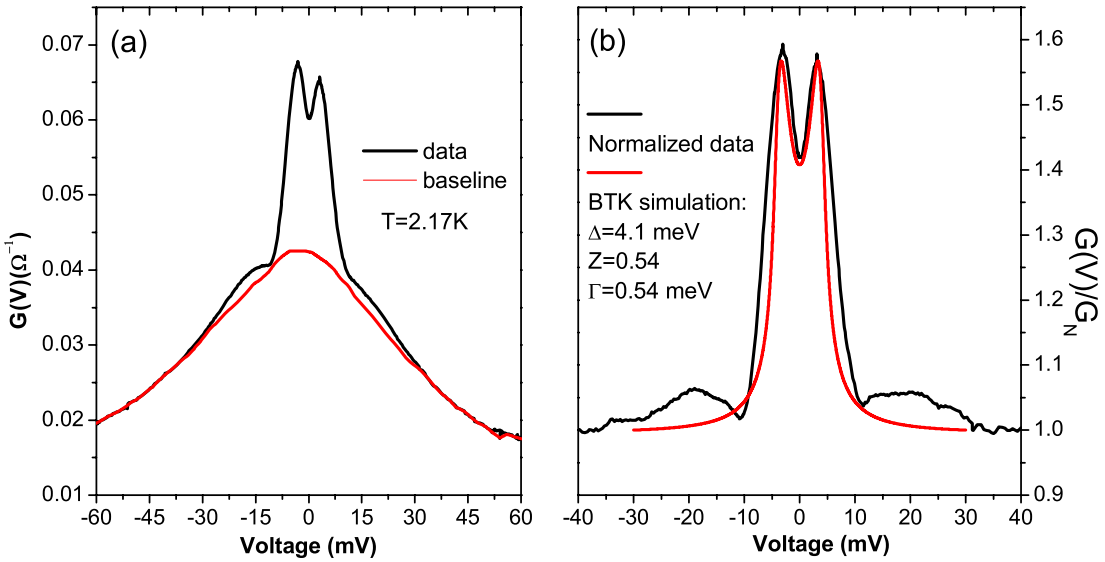

Figure 3. (a) Differential conductance curve $G(V)$ of a Au/$(Ba_{0.6}K_{0.4})Fe_2As_2$ point-contact junction and extrapolated background baseline; (b) the normalized conductance data and best fitting curve by BTK model.

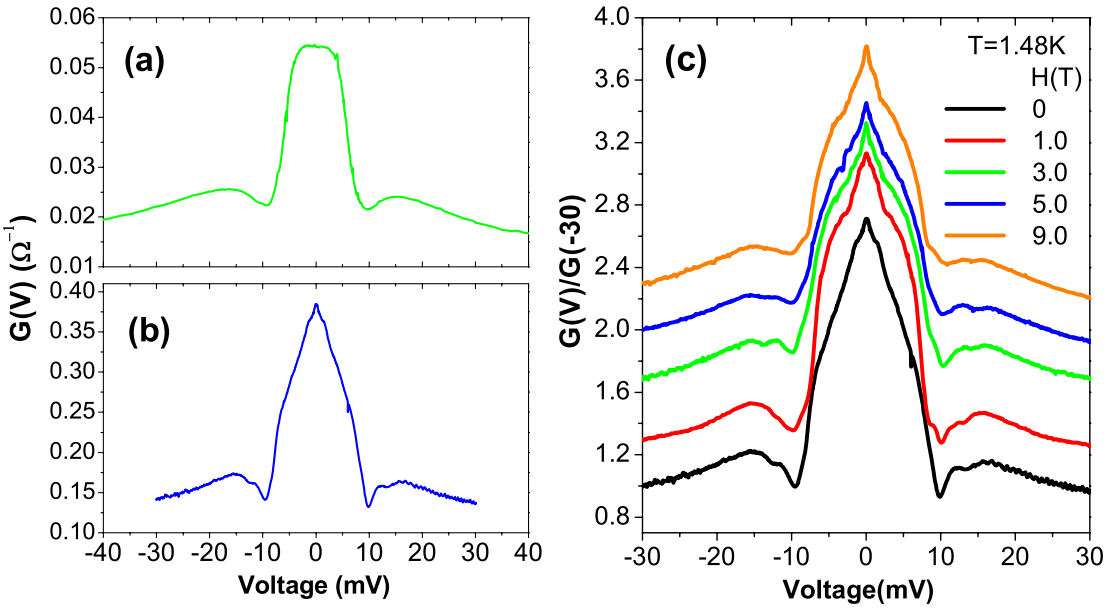

Figure 4. (a) and (b) Differential conductance curves $G(V)$ without double-peak structure for the Au/ $(Ba_{0.6}K_{0.4})Fe_2As_2$ point-contact junctions. (c) The field dependence of the $G(V)$ in (b) at $T = 1.48$ K. The curves are vertically shifted for clarity.

and (b) where a ZBCP is observed and the dips at ± 10 meV are more pronounced than in figure 1. Although a peak at zero-bias may arise from Andreev bound states in a d-wave superconductor [39], the width, the lack of field dependence as shown in figure 4(c), and the pronounced dip structures together indicate the ZBCP is due to the contact not being in the Sharvin limit.

3.2. $Ba(Fe_{0.9}Co_{0.1})_2As_2$

Conductance curves for the point-contact junctions on cleaved $Ba(Fe_{0.9}Co_{0.1})_2As_2$ single crystals are shown as a function of contact resistance and temperature in figures 5(a) and (b), respectively. For most contacts, a conductance enhancement at zero-bias with broad shoulders is observed. Note that with increasing temperature, the ZBCP disappears at the bulk T_c indicating it originates from Andreev reflection. A V-shape $G(V)$ is obtained when the tip is in gentle contact with the crystal, reaching hundreds of ohms junction resistance. With increased tip pressure reducing the junction resistance, the conductance curve changes to the general ZBCP feature.

3.3. $BaFe_2As_2$, $(Sr_{0.6}Na_{0.4})Fe_2As_2$ and $Sr(Fe_{0.9}Co_{0.1})_2As_2$

Figures 6(a) and (b) show the typical $G(V)$ curves with reduced conductance at lower voltage bias observed for the point-contact junctions on cleaved $(Sr_{0.6}Na_{0.4})Fe_2As_2$ and $Sr(Fe_{0.9}Co_{0.1})_2As_2$ surfaces. Similar features are also reported

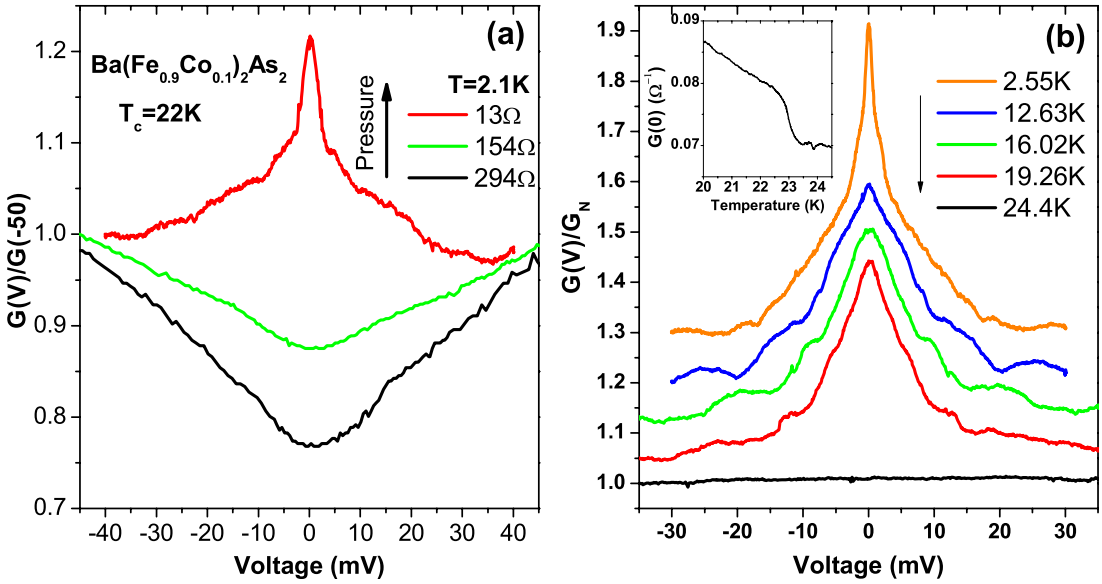

Figure 5. (a) The point-contact conductance curve, $G(V)$, evolving from V-shape to ZBCP with the increase of tip pressure at $T = 2.1$ K; (b) the temperature dependence of $G(V)$ with ZBCP. The curves are vertically shifted for clarity. Inset shows the temperature evolution of the zero-bias conductance around the bulk T_c.

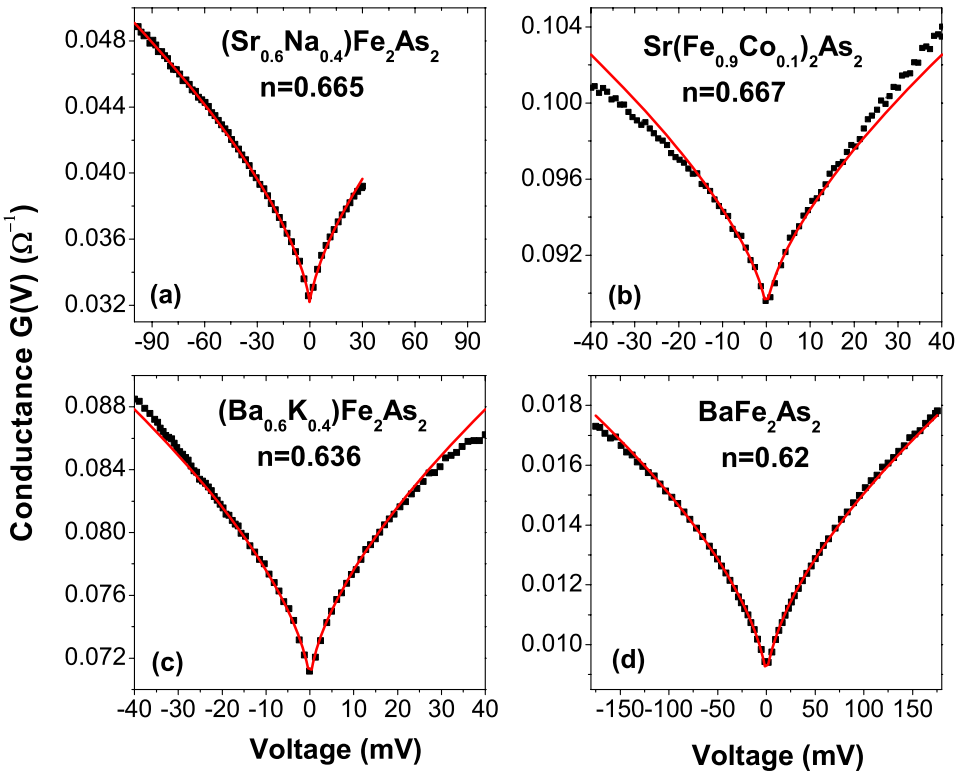

Figure 6. V-shape conductance valley (VCV): $G(V)$ curves observed (black) and power law fit (red) with n as the fitted power coefficient for the point-contact junctions on (a) $(Sr_{0.6}Na_{0.4})Fe_2As_2$ at $T = 2.33$ K; (b) $Sr(Fe_{0.9}Co_{0.1})_2As_2$ at $T = 2.20$ K; (c) $(Ba_{0.6}K_{0.4})Fe_2As_2$ at $T = 2.17$ K; (d) $BaFe_2As_2$ at $T = 1.95$ K.

by other groups [27, 28]. We find the conductance shape does not change up to applied magnetic fields of 9 T. There is no splitting of this V-shape conductance valley (VCV) in the magnetic field, which may rule out the Kondo scattering as an origin. This VCV feature, without Andreev reflection, is probably due to the absence of superconductivity in probed areas, even though the samples are checked to be superconducting. Conductance curves with this VCV, as shown in figure 6(c), are sometimes observed in the point-contact junctions on the same cleaved $(Ba_{0.6}K_{0.4})Fe_2As_2$ crystal as reported in section 3.1 (less than 20% of the time). We have also observed the same VCV feature for

the nonsuperconducting parent compound $BaFe_2As_2$ single crystals even up to 200 meV, as shown in figure 6(d), where SDW magnetic order exists. All the curves can be fit to a power law function $G(V) = G(0) + c * |V|^n$ with a power coefficient, $n \sim 2/3$. This may indicate a universal origin of this VCV observed commonly among different crystals.

In the $BaFe_2As_2$ parent compound, a structure transition occurs at the antiferromagnetic ordering temperature, $T_N \sim 135$ K. A phase-separated coexistence of magnetic order and superconductivity in Fe-122 family is reported in recent muon spin rotation (μSR) studies [40, 41, 43] with a magnetic correlation length >100 Å. Park *et al* demonstrate the mesoscopic phase-separated coexistence of magnetically ordered and non-magnetic states on a lateral scale of ~65 nm in the slightly underdoped $(Ba_{1-x}K_x)Fe_2As_2$ system ($T_c = 26$ K) [40]. Whether such coexistence is an intrinsic electronic property for Fe-122 system or due to some crystalline inhomogeneity remains uncertain.

In investigating the possibility that the VCV originates from magnetic order, we note magnetic order can be detected by PCARS [42]. A point-contact junction made on a superconducting region would exhibit Andreev reflection and one on a nonsuperconducting, magnetically ordered region would not. Instead, a signature due to electron scattering from magnetic order may be detected. This may be the case for $(Sr_{0.6}Na_{0.4})Fe_2As_2$ and $Sr(Fe_{0.9}Co_{0.1})_2As_2$ where no Andreev reflection peak in $G(V)$ has been observed, and $G(V)$ curves with the VCV feature are mostly observed. Goko *et al* apply μSR to investigate $(Ba_{0.6}K_{0.4})Fe_2As_2$ and $(Sr_{0.6}Na_{0.4})Fe_2As_2$ single crystals (same source as ours) and argue that static magnetism sets in at temperatures well above the superconducting T_c. They estimate the magnetic volume fraction to be 50% in $(Ba_{0.6}K_{0.4})Fe_2As_2$ crystals and ~90% in $(Sr_{0.6}Na_{0.4})Fe_2As_2$ crystals at low temperatures [43]. This is consistent with our more frequent observation of the VCV features in $(Sr_{0.6}Na_{0.4})Fe_2As_2$ than $(Ba_{0.6}K_{0.4})Fe_2As_2$ crystals.

Temperature-dependent point-contact measurement for the parent compound $BaFe_2As_2$ reveals the VCV feature is broadened and reduced with increasing temperature. As the Neel temperature ($T_N \sim 135$ K) is crossed, no dramatic change in the spectra is observed but thermal population effects may mask changes in conductance due to the magnetic transition.

4. Summary

In conclusion, our PCARS measurement on the cleaved $(Ba_{0.6}K_{0.4})Fe_2As_2$ single-crystal surfaces shows a superconducting gap energy of ~4 meV when $G(V)$ curves are analyzed by the single-gap BTK model. For point contacts on $Ba(Fe_{0.9}Co_{0.1})_2As_2$, zero-bias peak is frequently observed due to Andreev reflection. A universal power law behavior of the $G(V)$ observed for point-contact junctions on different Fe-122 superconducting samples may be a natural result of the mesoscopic phase-separated coexistence of magnetic and superconducting phases in the Fe-122 system. More careful investigation is needed to understand the origin of the power law VCV behavior. Point-contact measurement in the *ab* plane would be helpful to explore more detailed gap structure of this new superconductor family.

Acknowledgments

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