Andreev Reflection Like Enhancement Above Bulk $T_c$ in Electron Underdoped Iron Arsenides

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We use point contact spectroscopy (PCS) to probe the superconducting properties of electron doped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ ($x = 0.05, 0.055, 0.07, 0.08$) and hole doped Ba$_{0.8}$K$_{0.2}$Fe$_2$As$_2$. PCS directly probes the low energy density of states via Andreev reflection, revealing two distinct superconducting gaps in both compound families. Apart from the electron underdoped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$, the excess current due to Andreev reflection for the compounds follows the typical BCS temperature dependence. For underdoped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$, the temperature dependence of the excess current deviates from that of BCS, developing a tail at higher temperatures and surviving above bulk $T_c$. Possible explanations for this anomalous behavior are explored.

Point contact spectroscopy (PCS) [1] proves to be an extremely useful spectroscopic technique for studying conventional and unconventional superconductors since it is sensitive to the magnitude and symmetry of the superconducting order parameter. A point contact junction consists of a nanoscale metallic contact with a superconductor, with transport across the junction dominated by Andreev reflection [2]. The density of states may be directly extracted from the conductivity using the Blonder-Tinkham-Klapwijk (BTK) model [3]. PCS was instrumental in determining the precise location of the line nodes for the heavy fermion compound CeCoIn$_5$ [4], and in providing direct evidence for the multi-gap nature of the superconductor MgB$_2$ [5].

A number of research groups have utilized PCS to study the iron based superconductors. The results are well summarized in a recent review article by Daghero et al [6]. For the Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ and Ba$_{1-x}$K$_x$Fe$_2$As$_2$ families, measurements on near optimal and overdoped samples have revealed the existence of multiple gaps consistent with s-wave pairing [7, 8]. To our knowledge, no results have been reported for underdoped compounds, which constitute the most fascinating region of the 122 family phase diagram. Underdoped compounds exhibit a coexistence of magnetism and superconductivity at low temperatures [9] as well as nematic fluctuations in their normal state [10].

In this paper we present Andreev reflection data indicating multiple s-wave superconducting gaps for electron underdoped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ ($x = 0.05, 0.055$) and hole underdoped Ba$_{0.8}$K$_{0.2}$Fe$_2$As$_2$. We also present data on near optimal electron doped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ ($x = 0.07, 0.08$) that is in agreement with the published PCS literature.

We fit our lowest temperature data using the extended BTK model with two s-wave superconducting gaps [11]. All the point contact junctions show split Andreev peaks and none of the compounds have a maximum at zero bias voltage. This indicates that the superconducting order parameter does not have any nodes and the Fermi surfaces are fully gapped.

We define the superconducting transition by two temperatures: $T_c^{\text{onset}}$, for when the resistive transition starts, and $T_c^{\text{bulk}}$, for when it falls by 90% of the value at $T_c^{\text{onset}}$. The electron underdoped compounds show an Andreev reflection-like conductance enhancement between $T_c^{\text{onset}}$ and $T_c^{\text{bulk}}$ which we argue most likely arises from inhomogenous doping effects. For the rest of the compounds, the temperature evolution of the excess current due to Andreev reflection appears to follow the standard BCS like behavior and disappears at $T_c^{\text{bulk}}$.

Single crystals of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ are grown out of FeAs flux [12, 13] ($x = 0.05, 0.055, 0.07, 0.08$). Ba$_{0.8}$K$_{0.2}$Fe$_2$As$_2$ crystals are grown in Sn flux [14]. Metallic junctions are formed on freshly cleaved c-axis crystal surfaces and $dI/dV$ across each junction is measured using a standard four-probe lock-in technique. Junctions are constructed via the soft PCS method [15].

Fig. 1 presents $dI/dV$ curves for Ba(Fe$_{0.95}$Co$_{0.05}$)$_2$As$_2$ (left column, $T_c^{\text{bulk}} = 11.6$ K, $T_c^{\text{onset}} = 17$ K) and Ba(Fe$_{0.945}$Co$_{0.055}$)$_2$As$_2$ (right column, $T_c^{\text{bulk}} = 12.5$ K, $T_c^{\text{onset}} = 17$ K). Fig. 1a and 1b show the $dI/dV$ raw data at the lowest temperatures attained. The Andreev spectra clearly points to the presence of two superconducting gaps, as indicated with arrows. Fig. 1c and 1d show BTK fits to the normalized data from Fig. 1a and 1b, respectively. The dotted blue line is an isotropic s-wave single band BTK fit. While it provides a good fit to the features corresponding to the smaller gap, it cannot account for the features corresponding to the larger gap. A two band BTK approach, solid red line, is required to accurately describe the experimental data. The parameters in the fit are the two superconducting gaps $\Delta_1$ and $\Delta_2$, the Dynes lifetime broadening parameter for these
The Fermi surfaces of these compounds are quasi two dimensional sheets with elliptical electron pockets centered barrier strengths. The ratio $\Gamma$ for the different Fermi surfaces resulting in unequal effective. This might be due to the different Fermi velocities $\Delta$ for the 5% Co sample are $\Delta_1 = 3.0\text{meV}$ and $\Delta_2 = 6.6\text{meV}$ while those for the 5.5% Co sample are $\Delta_1 = 4.0\text{meV}$ and $\Delta_2 = 7.9\text{meV}$. (e, f) Temperature evolution of the Andreev reflection spectra.

data shown as opposed to the one band BTK model (dotted blue line). Note here also a conductance enhancement just above $T_{\text{bulk}}^c$. For Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$ ($T_{c}^{\text{bulk}} = 24.5\text{ K}$, $T_{c}^{\text{onset}} = 25\text{ K}$) the two band BTK fit (solid red line Fig. 2b) gives $\Delta_1 = 4.99\text{ meV}$, $\Delta_2 = 9.95\text{ meV}$. Fig. 2d shows the temperature evolution of the Andreev spectra of this junction. For near optimal doped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ our gap values are in good agreement with those reported in the literature for PCS [6, 8], scanning tunneling microscopy [17], and angle resolved photoemission spectroscopy [18].

Fig. 2e and f show $dI/dV$ for two different junctions on Ba$_{0.8}$K$_{0.2}$Fe$_2$As$_2$. The superconducting transition is very broad, with $T_{c}^{\text{onset}} = 26\text{ K}$ and $T_{c}^{\text{bulk}} = 15\text{ K}$. Fig. 2e shows the data can be fit with a single superconducting gap ($\Delta = 2.2\text{ meV}$), while Fig. 2f shows clear features corresponding to two superconducting gaps ($\Delta_1 = 2.7\text{ meV}$, $\Delta_2 = 7.5\text{ meV}$).

The Fermi surfaces of these compounds are quasi two dimensional sheets with elliptical electron pockets cen-
TABLE I.

| Crystal                        | $\Delta_1$ | $\Delta_2$ | $Z_1$ | $Z_2$ | $\Gamma_1/\Delta_1$ | $\Gamma_2/\Delta_2$ | $w$ | $T_{\text{onset}}$ | $T_{\text{bulk}}$ | $T_S$ | $T_N$ |
|-------------------------------|------------|------------|-------|-------|---------------------|---------------------|-----|------------------|-------------------|-------|-------|
| Ba(Fe$_{0.95}$Co$_{0.05}$)$_2$As$_2$ (e UD) | 3.0 meV   | 6.6 meV    | 0.395 | 0.40  | 0.56                | 0.62                | 0.67 | 17 K             | 11.6 K            | 78 K  | 70 K  |
| Ba(Fe$_{0.945}$Co$_{0.055}$)$_2$As$_2$ (e UD) | 4.0 meV   | 7.9 meV    | 0.31  | 0.35  | 0.25                | 0.30                | 0.30 | 17 K             | 12.5 K            | 75 K  | 63 K  |
| Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$ (e OD) | 4.0 meV   | 7.0 meV    | 0.28  | 0.30  | 0.20                | 0.15                | 0.20 | 23 K             | 21.7 K            | -     | -     |
| Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$ (e OD) | 4.99 meV  | 9.95 meV   | 0.39  | 0.435 | 0.36                | 0.23                | 0.50 | 25 K             | 24.5 K            | -     | -     |
| Ba$_{0.8}$K$_{0.2}$Fe$_2$As$_2$ (h UD)       | 2.2 meV   | -          | 0.373 | -     | 0.18                | -                   | 1   | 26 K             | 15 K              | 90 K  | 90 K  |
| Ba$_{0.8}$K$_{0.2}$Fe$_2$As$_2$ (h UD)       | 2.7 meV   | 7.5 meV    | 0.32  | 0.45  | 0.57                | 0.53                | 0.37 | 26 K             | 15 K              | 90 K  | 90 K  |

FIG. 3. (color online) Comparing the zero bias conductance (blue) and bulk resistivity (red) curves. (a, b) For the electron underdoped compound Ba(Fe$_{0.95}$Co$_{0.05}$)$_2$As$_2$ and Ba(Fe$_{0.95}$Co$_{0.05}$)$_2$As$_2$, the superconducting transitions are wide and the zero bias conductance starts to rise close to $T_{\text{onset}}$. (c) For the nearly optimal doped compound Ba(Fe$_{0.92}$Co$_{0.08}$)$_2$As$_2$, the superconducting transition is narrow. (d) For Ba$_{0.8}$K$_{0.2}$Fe$_2$As$_2$, like the electron underdoped compounds the transition is broad but the zero bias conductance only starts to rise close to $T_{\text{bulk}}$.
FIG. 4. (color online) (a) The temperature evolution of the excess current, $I_{\text{exc}}$, for all our samples. The solid black line shows the dependence for weakly coupled $s$-wave BCS superconductors. The temperature has been normalized to bulk $T_c$. Apart from the electron underdoped compounds, reasonable fits are obtained. For them, $I_{\text{exc}}$ initially follows the BCS trend before developing a tail at higher temperatures. The dashed gray rectangle is highlighting this anomaly. (b) The trend before developing a tail at higher temperatures. The fits are obtained. For them, $I_{\text{exc}}$ shows the dependence for weakly coupled $s$-wave BCS superconducting gaps. The values of the two gaps may be extracted by using the independent multiband BTK model. Apart from underdoped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$, the temperature evolution of the excess current for the crystals is well described by the BCS temperature dependence. The excess current for underdoped Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ exhibits excess conductance at higher temperatures that survives above the bulk $T_c$. The shape of $I_{\text{exc}}$ vs. $T$ can be simulated assuming microscopic inhomogeneity in the Co doping in the crystals.

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Supplemental Material for
Andreev Reflection Like Enhancement Above Bulk $T_c$ in Electron
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EXCESS CURRENT CALCULATION

Andreev reflection causes an increase in the current transmitted across a normal metal-superconductor point contact junction. \( I_{\text{exc}} \) is defined as the extra current that flows through the junction when compared with its non-superconducting state. To calculate this current we use the equation:

\[
I_{\text{exc}}(T) = \int_{-V < -\Delta}^{V > \Delta} \frac{dI}{dV}(T)\,dV - \int_{-V < -\Delta}^{V > \Delta} \frac{dI}{dV}(T > T_{\text{onset}})\,dV
\]  

FIG. 1. (a) Raw \( dI/dV \) curves taken on Ba(Fe\(_{0.95}\)Co\(_{0.05}\))\(_2\)As\(_2\) at 1.8 K (blue) and 17.2 K (red). (b) The same curves, after they have been symmetrized and normalized to \( dI/dV \) at 17.2 K. \( I_{\text{exc}} \) is represented by the shaded gray area on the plot. It is calculated by integrating the area under the curves and subtracting the value at 17.2 K from the value at 1.8 K. The integration limits are set at ± 20 mV, represented by the short vertical black lines. At biases higher than 20 mV the two \( dI/dV \) curves are nearly identical.

We illustrate how this integration is performed for Ba(Fe\(_{0.95}\)Co\(_{0.05}\))\(_2\)As\(_2\) in Figure 1.

Figure 1a shows the raw \( dI/dV \) curves taken at 1.8 K (blue, lowest temperature attained for this junction) and 17.2 K (red, \( T \sim T_{\text{onset}} \)). Figure 1b shows the same curves after they have been symmetrized and normalized with the curve at 17.2 K. Symmetrization is carried out by taking the average of the \( dI/dV \) values at positive and negative biases.

The next step is to integrate the area under the curves and subtract the current at 17 K from the current at 1.8 K. We choose the integration limit to be from -20mV to +20mV.
since at biases higher than that Andreev reflection dies out and the two $dI/dV$ curves are nearly identical. The gray shaded area in Figure 1b represents the final $I_{exc}$ that we obtain.

This same procedure is repeated for all our crystals at various temperatures. Figure 4a in the main text of our paper is obtained by combining all of the $I_{exc}$ data points.

**INHOMOGENEOUS DOPING MODEL**

![Graphs showing the comparison of $I_{exc}$ with BCS $I_{exc}$](image)

**FIG. 2.** (a) Comparing the $I_{exc}$ calculated from Gaussian and linear distribution functions with the BCS $I_{exc}$. The distribution function $I_{exc}$ develops a tail above $T_{bulk}^{c}$. (b) The number of channels with a given $T_{c}$ for the Gaussian ($\sigma = 5\% T_{bulk}^{c}$) and the Linear distribution functions.

Our basic assumption is that our point contact junction is comprised of multiple channels and there is slight variation in the local $T_{c}$ values of these channels. Most of the channels start showing Andreev reflection at $T_{c}^{bulk}$ but some of them start Andreev reflecting above it while other start below it. We define a variable $T_{c}^{local}$ and set its limits to be 0.8-1.2 $T_{bulk}^{c}$.

We have tried various distribution functions for $T_{c}^{local}$. Figure 2b shows the number of channels with a given $T_{c}$ for a Gaussian ($\sigma = 5\% T_{bulk}^{c}$) and a Linear distribution function. The largest number of channels superconduct at $T_{c}^{bulk}$ and as $T_{c}^{local}$ deviates more and more from $T_{c}^{bulk}$, the number of channels with that $T_{c}$ falls.

$I_{exc}$ is calculated by summing up the excess current due to all the Andreev reflection channels. Figure 4b in the main text of our paper uses the Gaussian distribution function to calculate $I_{exc}$. In Figure 2a, we compare $I_{exc}$ from the Gaussian and linear distribution functions with BCS $I_{exc}$. The general feature is that the $I_{exc}$ for the distribution functions
develops a tail above $T_{c}^{bulk}$. 