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Pseudogap-less high-$T_c$ superconductivity in BaCo$_x$Fe$_{2-x}$As$_2$

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Abstract – The pseudogap state is one of the peculiarities of the cuprate high-temperature superconductors. Here we investigate its presence in BaCo$_x$Fe$_{2-x}$As$_2$, a member of the pnictide family, with temperature-dependent scanning tunneling spectroscopy. We observe that for under-, optimally and overdoped systems the gap in the tunneling spectra always closes at the bulk $T_c$, ruling out the presence of a pseudogap state.

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With the discovery of the pnictide family of high-temperature superconductors [1], a new window to the physics behind high-temperature superconductivity (HTSC) has been opened. At first glance, the phase diagrams of the cuprates and the pnictides are strikingly similar. In both systems, electron or hole doping suppresses the magnetic ground state of the parent compound and produces a superconducting dome, see, for instance, ref. [2]. However, whereas the cuprates are Mott insulators at low dopings and upon doping first completely lose their long-range magnetic order before superconductivity emerges, the pnictides have a metallic ground state and upon doping directly cross from a magnetic to a superconducting phase. The nature of this transition is still under debate, as some studies find that the two regions can coexist [3–5] while others indicate a phase separation scenario [6,7].

At low doping concentrations, the cuprates furthermore display a second energy scale, characterised by a depression of spectral weight at the Fermi level, which is observed both in the superconducting and normal state, the so-called pseudogap [8,9]. Since the role of the pseudogap phase in the cuprates is still under heavy debate, the establishment of the presence or absence of such a phase in the iron pnictides is of particular importance. There are several indications that at least some of the pnictides have a pseudogapped region at the underdoped side of the phase diagram. For instance, anomalous resistivity characteristics in LaFeAsO$_{1-x}$F$_x$ and SmFeAsO$_{1-x}$F$_x$ have been interpreted in a pseudogap-like scenario [10]. Nuclear magnetic resonance (NMR) investigations [11,12] and angle-resolved photoemission (ARPES) measurements [13] have found signs of a pseudogap in the former system as well. It should be noted, however, that Andreev spectroscopy measurements argue against a pseudogap in the latter system [14]. In Ba$_{1-x}$K$_x$Fe$_2$As$_2$ ARPES [15], optical conductivity [16] and pump-probe spectroscopy [17] measurements showed pseudogap-like behaviour, and claims of possible pseudogap behaviour have been made in the FeSe$_{1-x}$Te$_x$ system [18]. In the case of BaCo$_x$Fe$_{2-x}$As$_2$, ARPES measurements have reported a slight depression above the superconducting transition temperature [19], possibly caused by a weak pseudogap.

Another observation that could be linked to a pseudogap phase is the large spread in the separation between the quasi-particle peaks ($2\Delta_{p-p}$), observed in tunneling spectra of the pnictides [20,21], which is difficult to reconcile with the sharpness of the superconducting transition. In underdoped cuprates, these spectra are dominated by the pseudogap features, and the true superconducting quasi-particle spectrum has been resolved only very recently [22]. The question is therefore whether also in the pnictides tunneling spectra show a mix of pseudogap and superconducting gap features.

In this letter we report temperature- and doping-dependent scanning tunneling spectroscopy measurements on under-, optimally and overdoped BaCo$_x$Fe$_{2-x}$As$_2$ crystals ($x = 0.08$, 0.14 and 0.21) to determine whether tunneling experiments indeed observe the superconducting gap, which closes at $T_c$, or a pseudogap, which does not.
Fig. 1: (Colour on-line) Optimally doped BaCo$_{x}$Fe$_{2-x}$As$_{2}$ ($x = 0.14$, $T_c = 22 \pm 0.5$ K): (a)–(d) Differential conductance spectra for four different superconducting gap sizes on the optimally doped compound, as a function of temperature. The spectra are offset in discrete steps with respect to each other for clarity. $V = 45$ mV, $I = 40$ pA. (e)–(h) The same spectra as shown in (a)–(d), but normalised to the spectrum for each location taken at 24 K. (i) Color legend for (a)–(h). (j) Comparison between different normalisation temperatures, excluding an influence of the normalisation on the results.

There is some debate in the literature on the nature of the cleavage surface [20,21,23–27]. We follow the view that this system, in order to avoid a polar surface, cleaves through the Ba layer, exposing half a Ba layer on either side of the cleave [26–28]. Although a comparison of regular and hard X-ray photoemission data suggests that the cleavage surface does not have a strong influence on the global near-surface electronic environment [29], LDA calculations report electronic properties differing from the bulk on $\sqrt{2} \times \sqrt{2}$ reconstructed Ba surfaces [27]. However, as we observe identical superconducting gaps on both $\sqrt{2} \times \sqrt{2}$ as well as $2 \times 1$ or other surface terminations, we present data from spectroscopic surveys recorded on the modal termination topography. We do note that areas with a relatively large $z$-corrugation ($> 2$ Å) show anomalous spectroscopic signatures, likely caused by a local Ba surplus or deficiency, which were therefore not used for spectroscopic surveys. The experimental setup, which enables us to track the surface with atomic precision as a function of temperature, has been described elsewhere [21].

First, optimally doped BaCo$_{x}$Fe$_{2-x}$As$_{2}$ ($x = 0.14$) samples were studied ($T_c = 22 \pm 0.5$ K). Figures 1(a)–(d) show the evolution of the gap for four different locations on the surface for temperatures ranging from 4.5 K to 24 K. As can be seen in the figure, the clear coherence peaks at 4.5 K are smoothly depressed and are completely gone at temperatures above $T_c$. Due to the U-shaped background, it is difficult to follow the temperature dependence in great detail. To overcome this problem, an often used procedure is to divide the differential conductance at a temperature, $T$, by that taken in the normal state, i.e. $(dI/dV)_T / (dI/dV)_{T_{\text{normal}}}$. The result of this analysis is plotted in fig. 1(e)–(h). Here, unlike

Fig. 2: (Colour on-line) Underdoped BaCo$_{x}$Fe$_{2-x}$As$_{2}$ ($x = 0.08$, $T_c = 14 \pm 1$ K): (a) Average spectrum of ~4000 spectra taken both at 5 K and 20 K on a square grid on the same 100 $\times$ 100 Å$^2$ field of view. (b) Spectrum obtained by normalising the 5 K spectrum shown in (a) to the 20 K spectrum. (c) Several single pixel spectra taken along a random line from the conduction map of which the average is shown in (a) illustrating the variation in the spectra, the gap edges are indicated by circles.
in the cuprates, the $2\Delta_{p-p}$ values extracted from the normalised spectra are identical to those extracted from the raw data for all four shown sizes of superconducting gap. The exact temperature at which the gap closes or fills is difficult to estimate, but for all gap sizes it is within 1 K of the bulk $T_c$. The fact that the peak-to-peak separation is seemingly temperature independent can be well understood by taking into account temperature broadening effects, and its behaviour tracks the closing of a temperature broadened BCS-like gap rather well [30].

To ensure that the normalisation temperature used in the procedure described above does not influence the normalised spectra, normalisation of a spectrum taken at 17.5 K both to the spectra taken at 22 K, i.e. very close to $T_c$, and that taken at 24 K are compared in fig. 1(j). Both normalised spectra are clearly very similar, whereas the spectrum taken at 22 K normalised to the one taken at 24 K is basically a straight line. From this we can conclude that spectra taken above $T_c$ do not show an observable variation and the choice of normalisation temperature is not critical, as long as it is $> T_c$. It is thus safe to say that the gaps observed are indeed the superconducting gaps.

We now turn our attention to lower doping concentrations, where, in analogy with the cuprates, one is more likely to expect a pseudogap behaviour and where magnetic correlations that could produce a gap are stronger. Figure 2(a) shows the average of $\sim 4000$ spectra taken both at 5 K and 20 K on the same 100 x 100 Å² field of view on a BaCo$_{1-x}$Fe$_2$As$_2$ sample with $x = 0.08$ ($T_c = 14 \pm 1$ K, $T_{SDW} = 70 \pm 3$ K). The low-temperature average, normalised to the high-temperature spectrum is shown in fig. 2(b).

One peculiarity valid for all our measurements performed on underdoped samples is that the superconducting gap signatures are less pronounced than in the optimally doped materials. At this point we note that temperature broadening at 5 K of a pure single band BCS spectrum will already effectively fill the flat bottomed gap [30]. Furthermore, as other experiments have already alluded to, it might very well be possible that there is a non-zero density of states at the Fermi level in the superconducting state due to non-gapped bands [19,31]. Nevertheless, the single pixel spectra, of which several are shown in fig. 2(c), clearly show there are gaps all over the field of view, which after normalisation have the same peak-to-peak separation as is apparent in the raw data and all close at $T_c$. These observations strongly suggest that the gaps observed at low temperature are indeed the superconducting gaps and not pseudogaps.

Since at 20 K the system is still well within the orthorhombic, magnetically ordered state, one might expect gap-like features in the spectra due to a depression of spectral weight as a result of gapping of parts of the Fermi surface [32–34]. Assuming a mean-field gap, such features would be typically at $\sim 10$ mV. However, since these are not observed, they either occur at higher energies, are not picked up due to tunneling matrix element effects, or are simply not present due to phase separation between magnetic and superconducting regions.

Additional measurements, for instance to higher energies, should be performed to address this point. The presence of superconducting gaps over the entire field of view sets a lower limit length scale of tens of nanometers to a possible phase separation between superconducting and non-superconducting magnetically ordered regions.

To complete our survey of the phase diagram of BaCo$_{1-x}$Fe$_2$As$_2$, we turn our attention to the overdoped compound ($x = 0.21$, $T_c = 13 \pm 1$ K). Figures 3(a)–(d) show the detailed temperature dependence of four locations with a different $2\Delta_{p-p}$. Analogous to the analysis...
used for the optimally doped sample, figs. 3(e)–(h) show the spectra normalised to the spectrum taken at the exact same location for $T > T_c$. As was the case for the under- and optimally doped samples, the gaps seen at low temperature in the normalised spectra are identical in magnitude as the raw data gaps and all vanish above $T_c$.

To summarise, for underdoped, optimally doped, and overdoped $\text{Ba}_x\text{Fe}_2-2x\text{As}_2$ ($x = 0.08, 0.14$ and $0.21$), $2\Delta_{p,p}$ observed in tunneling spectroscopy measurements has been tracked as a function of temperature. The gaps for all doping concentrations studied and all gap sizes observed vanish at the bulk $T_c$, excluding a pseudogap scenario in these pnictide superconductors. Since gaps have been seen across the entire field of view of hundreds of square ångstroms, nano-scale phase separation of magnetic and superconducting patches is unlikely, also in the undoped material. The indications of the presence of a pseudogap in several compounds closely related to the one studied here makes the family of pnictide superconductors a theoretical challenge to understand, as the action of specific dopant atoms can lead to completely differing behaviour. An interesting observation is the possible relation between the dopant atom carrier type and the presence or absence of a pseudogap. Signatures of a pseudogap in the electron-doped cuprates have been reported (see ref. [35] and references therein), but these are far less pronounced than in the hole-doped cuprates, where it dominates a large portion of the phase diagram. Similarly, whereas we show here that there is no pseudogap in the electron-doped pnictide $\text{BaCo}_x\text{Fe}_2-3x\text{As}_2$, there are indications of a pseudogap in hole-doped $\text{Ba}_{1-x}\text{K}_{x}\text{Fe}_2\text{As}_2$ [15–17]. Mapping out the presence or absence of a pseudogap in more related compounds, both electron and hole doped, will determine if this is a generic behaviour and should give a better understanding of the role of the pseudogap for high-temperature superconductivity.

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