Point contact spectroscopy of Pr$_{2-x}$Ce$_x$CuO$_4$ in high magnetic fields

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We have studied the density of states of the normal state of the electron-doped superconductor Pr$_{2-x}$Ce$_x$CuO$_4$ at low temperatures and in magnetic fields up to 31 Tesla using point contact spectroscopy on single crystals. Our data clearly reveal an anomalous gap in the normal state density of states. This normal state gap survives even in the highest applied field of 31 T. Our results cast doubt over whether this gap found in electron-doped cuprates is the analog of the pseudogap in hole-doped cuprates. We have suggested an alternate origin of the normal state gap, which involves the effect of disorder and electron correlations at the surface of cuprates.

The normal state of copper-oxide (cuprate) superconductors has been the subject of intense research because it may hold the key to understanding the mechanism leading to the observed high-critical temperatures ($T_c$) in these materials. In conventional superconductors both the formation and condensation of paired carriers into a zero resistance state occurs at the critical temperature $T_c$, which is accompanied by the formation of the superconducting gap. A striking feature of hole-doped cuprates is the formation of a gap in the density of states well above $T_c$, viz. the pseudogap (PG) [1, 2, 3]. Several models have been suggested to explain the origin of the pseudogap, which can be sorted into two main categories viz. the pre-formed pairing theory [4, 5] and the competing order parameter theory [6, 7]. Preformed pairing theory considers the pseudogap state to be a phase fluctuation regime which is a precursor to the superconducting state whereas, the competing order parameter models suggest that the pseudogap is due to another competing phase.

The pseudogap has been studied extensively in hole-doped (p-doped) cuprates mainly by probing the region outside the superconducting dome in the temperature - hole-doping ($T - x$) phase diagram [3]. The presence of a pseudogap in electron-doped (n-doped) cuprates is more controversial. One example is the difference in the energy scales of the gaps measured in the normal state of n-doped cuprates using different techniques. While tunneling and point-contact spectroscopy show a normal state gap in n-doped cuprates with an energy scale similar to the superconducting gap (∼ 10 meV) [8, 9, 10], angle-resolved photoemission spectroscopy and optical reflectivity reveal a much larger gap (∼ 100 - 200 meV) which is attributed to a spin-density wave gap [12, 13]. In the normal state of p-doped cuprates there is reasonable agreement among the gaps measured using different experimental techniques [3]. [Note: We refer here to the higher energy gap found in the normal state of p-doped cuprates which increases with decreasing doping and not the lower energy gap which decreases with decreasing doping as recently reported in ref. 14]. For the rest of this manuscript we will refer to the higher energy gap in p-doped cuprates, as the pseudogap.] Furthermore, the phase fluctuation region above $T_c$ observed by Nernst effect measurements in p-doped cuprates is either absent [15] or is smaller in n-doped cuprates [16].

A unique feature of the n-doped cuprates is that it is possible to probe the normal state inside the superconducting dome for the entire doping range by driving the superconductor into the normal state with a field $H$ greater than the upper critical field $H_{c2}$, due to the relatively low values of $H_{c2}$ (∼ 10 T at low temperatures for the optimally doped compounds). Tunneling across grain boundary junctions and point-contact spectroscopy results in magnetic fields $H > H_{c2}$ have clearly shown the presence of a normal state gap (NSG) in the density of states, which is comparable in energy scale to the superconducting gap and which increases in energy scale as the doping is decreased [8, 9, 10]. Alff et al. have mapped the region in the phase diagram where this NSG is observed [11]. The NSG observed in n-doped cuprates shows a similar behavior with doping as the high energy pseudogap in p-doped cuprates [3, 11, 14]. If the NSG in n-doped cuprates is indeed analogous to the pseudogap in p-doped cuprates, then Alff et al. result suggests that the pseudogap in n-doped cuprates is present inside the superconducting dome and vanishes at a doping level close to the optimum value ($x \sim 0.16$), which supports the competing order parameter scenario [11]. However, Dagan et al. have performed tunneling spectroscopy measurements on thin films of Pr$_{2-x}$Ce$_x$CuO$_4$ for the doping range 0.11 < x < 0.19 and have shown that the NSG is observed well into the overdoped region [17]. In fact, in the overdoped region ($x > 0.17$) the temperature $T^*$ above which the NSG vanishes is approximately equal to $T_c$. This result provides strong evidence that the NSG is related to the superconductivity in PCCO and if the NSG is the pseudogap, supports the pre-formed pairing scenario. Recently, Kawakami et al. have reported the observation of a pseudogap in the n-doped cuprate Sm$_{2-x}$Ce$_x$CuO$_4$-4 using interlayer tunneling transport [18]. By studying this pseudogap’s behavior in magnetic fields up to 45 T, Kawakami et al.
concluded that the pseudogap in both $n$-doped and $p$-doped cuprates is formed due to spin-singlet (pair) formation above $T_c$ \cite{18}. By comparing the behavior of the NSG in high magnetic fields to the pseudogap reported in ref. \cite{13}, we expect to obtain clues to explain the origin of the NSG. In this paper we report the first point contact spectroscopy (PCS) measurements on single crystals of optimally doped Pr$_{2-x}$Ce$_x$CuO$_4$ ($x = 0.15$, PCCO) in magnetic fields up to 31 T. We show that the NSG survives even in a field of 31 T, which casts doubt over the validity of the assumption that the NSG is the pseudogap in $n$-doped cuprates. We suggest alternate origins for the formation of the NSG.

Point contact spectroscopy (PCS) is similar to scanning tunneling spectroscopy, in the sense that the current injection occurs between a sharp tip and the sample. However, in PCS the tip is actually in physical contact with the sample. Hence, PCS is less susceptible to mechanical vibrations than scanning tunneling spectroscopy, which is an important factor to consider when choosing a measurement method for the water-cooled magnets in the DC High Field Facility at the National High Magnetic Field Laboratory in Tallahassee (NHMFL). The PCS data were taken using a custom built probe designed for operation in the 32 mm bore DC field magnets at the NHMFL \cite{19}. The maximum magnetic field is 33 tesla at temperatures down to 1.5 K.

To tunnel into the $a - b$ plane and apply a magnetic field perpendicular to the $a - b$ plane (see Fig. 1, right inset) we used bevel gears to suitably change the direction of tip-sample approach (Fig. 1 inset). For finer control over the junction resistance, we used a worm-gear arrangement (1:96 ratio) at the top of the probe. Differential conductance ($dI/dV \equiv G$) vs. $V$ curves were obtained directly by using a modulation technique. Details of the apparatus and electronics are given in ref. \cite{19}. The PCCO crystals were grown by the self-flux technique followed by an oxygen reduction procedure to achieve a $T_c \approx 21 \pm 0.8$ K \cite{20}. The data presented in this paper were taken for different point contact junctions on the same single crystal. In addition, we have confirmed the presence of the pertinent density of states features at high magnetic fields for another single crystal and a thin film of PCCO.

Fig. 1 shows a typical $dI/dV - V$ curve obtained on a single crystal of PCCO for $H = 0$. All point contact spectra shown in this paper were taken at $T = 1.5$ K. The coherence peaks which are a signature of the superconducting gap are clearly observed and are marked in the figure. The large value for $dI/dV$ at zero-bias is due to the high transparency of a point contact junction. In addition to the coherence peaks, the point contact spectrum also shows a linear increase in $dI/dV$ ($G$) with $V$ for sample bias above the superconducting gap value. This asymmetric linear background conductance makes it difficult to quantify the effect of the magnetic field on the NSG and needs to be removed. We will describe the procedure for removing the background later in this report.

Fig. 2 shows the variation of the $G - V$ characteristics with an applied magnetic field. Since the resistance of point-contact junctions is low, the $G - V$ curves shift vertically when the superconductor becomes normal above $H_{c2}$. Such an effect is also observed in grain boundary junctions \cite{11}. We have removed this effect by calculating the change in the resistance of the sample from the vertical displacement of the $G - V$ curves and also adjusted

![FIG. 1: Differential conductance ($dI/dV \equiv G$) vs. $V$ curve of a point contact junction between Pr$_{2-x}$Ce$_x$CuO$_4$ ($x = 0.15$) and a Pt-Rh wire at $T = 1.5$ K. The superconducting gap is marked. The right inset shows the configuration of the point contact junction, which ensures current injection into the $a - b$ plane. A small contact area is achieved due to the small radius of the Pt-Rh wire (0.25 mm) and the 20 $\mu$m thick crystal.](image1)

![FIG. 2: The effect of a magnetic field on the point contact spectrum for Pr$_{2-x}$Ce$_x$CuO$_4$ ($x = 0.15$). The normal state gap is clearly visible in a magnetic field of 22 T. The inset shows the raw $dI/dV$-$V$ curves as a function of the applied magnetic field.](image2)
FIG. 3: (a) Normalized point contact spectra showing the effect of a magnetic field on the normal state gap after a linear background conductance has been removed from the $dI/dV$ data. The inset shows the fit to the linear background conductance (solid line), which is used to normalize the spectra. The arrows indicate the range for the linear fit. (b) A 2D plot of the point contact spectra as a function of junction bias and magnetic field, showing the negligible effect of a magnetic field on the normal state gap.

FIG. 4: (a) Normalized point contact spectra in fields up to 28 T. The inset is a point contact spectrum at 31 T. (b) A 2D plot of the point contact spectra as a function of junction bias and magnetic field.

The sample bias accordingly [21]. The normal state gap is clearly observed even in fields of up to 22 T.

To analyze the effect of a magnetic field on the NSG, we have to remove the linear background and asymmetry of the $G - V$ curves. The asymmetry of $G - V$ curves has been observed even in scanning tunneling spectra of cuprates [2, 22] and has been explained within the Gutzwiller-Resonating Valence Bond theory [23]. However, for our point-contact junctions and low bias voltages, the origin of the linear background and the associated asymmetry for the positive and negative biases may also be due to multiple inelastic scattering through the tunneling barrier [24]. Fig. 3 shows our method for background subtraction which is based on the method suggested by Shan et al. [25]. Since the background is asymmetric we have used only the data for negative bias voltages rather than use an arbitrary background function near zero-bias to connect the asymmetric linear backgrounds for the positive and negative biases. The linear background was fitted over the range shown by the arrows in the inset of Fig. 3. The normalized G-V curves obtained after the background subtraction is shown in Fig. 3a. The suppression of the density of states near the fermi level in the normal state of PCCO is clearly visible even at fields of 22 T. Since the width of the NSG is difficult to quantify we show the magnetic field dependence of the NSG in the form of a 2D plot (Fig. 3b) and it shows that the magnetic field has a negligible effect on the NSG. Data on a higher resistance point contact junction is shown in Fig. 4a and shows that the NSG is present in fields as high as 28 T. We could not obtain $G - V$ curves at higher magnetic fields without changing the junction characteristics. However, the normal state gap is still observed in a field of 31 T as shown in the inset of Fig. 4a. The 2D plot of the PCS data in Fig. 4a is shown in Fig. 4b and it reveals a small reduction of the NSG in magnetic fields above 20 T.

From the detailed behavior of the NSG in a mag-
To investigate the reason behind the insensitivity of the NSG to magnetic fields, we have plotted the zero bias normalized conductance, $G_0$ as a function of magnetic field in Fig. 5. The initial sharp rise in $G_0$ from 0 to about 3 T is due to the suppression of the superconducting gap. For $H > 3$ T, the gradual rise in $G_0$ indicates the slow reduction of the NSG with field. Assuming the simplest function for the variation of $G_0$ with $B$, we have fitted a line to the data. From the linear function we estimate that $G_0$ will approach 1 (which signifies the complete suppression of the NSG) at a magnetic field of $\approx 90$ T. The energy associated with this value of the magnetic field is $g\mu_B H \approx 10$ meV, which is comparable to the width of the NSG. Although the above estimate is crude, such a correspondence of the energy scales is reminiscent of the effect of a magnetic field on the zero bias anomaly (ZBA) formed due to electron-electron interactions in the density of states of disordered metals [26, 27]. In 3-D the ZBA has the form $n(E) = n(0)(1 + \sqrt{E/\Delta})$, where $\Delta$ is called the correlation gap. The inset of Fig. 5 shows a plot of $G$ as a function of the square-root of the bias voltage in a magnetic field of 26 T. Although the linear behavior (solid line) of $G$ in the bias range $0.8 \text{ mV} < V < 3.5 \text{ mV}$ suggests that the origin of the NSG could be electron-electron interactions, there are two problems with this conclusion. First, the ZBA or the correlation-gap is observed in materials which show a $\ln(T)$ conductivity whereas Dagan et al. have shown that the NSG exists even in overdoped PCCO which shows a metallic conductivity [17]. However, it is possible that the NSG is purely a surface phenomenon since it has been observed only in tunneling and point contact experiments on PCCO [8, 9, 10, 11], which are surface sensitive probes [31] and it is suspected that the surface of cuprates can sometimes show properties different from the bulk [28]. In addition, such features have also been observed in tunneling spectra of other metallic oxides [29]. Secondly, the scaling of the width of the NSG with the superconducting gap (as a function of doping), strongly suggests that the NSG is related to superconductivity although, it is also possible that the evolution of the NSG with doping is related to the insulator-to-metal crossover in the normal state of PCCO near optimal doping [30].

In summary, we have studied the density of states near the fermi level in the normal state of the electron-doped cuprate $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$ ($x = 0.15$) using point contact spectroscopy. We have observed a normal state gap in the density of states, which persists even in a magnetic field of 31 T. The weak magnetic field dependence of the normal state gap leads to two possible explanations: (1) The pseudogap closing field is higher (by about a factor of 3) than expected from a pure Zeeman relation between $T^*$ and $H_{pg}$ and therefore, preformed pairing above $T_c$ is not the origin of the pseudogap [18] or (2) the NSG observed in $n$-doped cuprates is not analogous to the pseudogap in $p$-doped cuprates. Instead it is formed due to electron-electron interactions at the surface of $n$-doped cuprates.

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