

Gapped tunneling spectra in the normal state of $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$

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We present tunneling data in the normal state of the electron doped cuprate superconductor $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$ for three different values of the doping x . The normal state is obtained by applying a magnetic field greater than the upper critical field H_{c2} for $T < T_c$. We observe an anomalous normal state gap near the Fermi level. From our analysis of the tunneling data we conclude that this is a feature of the normal state density of states. We discuss possible reasons for the formation of this gap and its implications for the nature of the charge carriers in the normal and the superconducting states of cuprate superconductors.

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I. INTRODUCTION

The nature of the normal state of high- T_c superconductors is a matter of great interest because it gives information about the nature of the charge carriers and the pairing mechanism leading to superconductivity. It is now widely accepted that, in cuprate superconductors there is a depletion of the density of states (DOS) at the Fermi level (E_F) at a temperature T^* , which is much higher than the superconducting transition temperature (T_c). This gap is called the pseudogap (PG). The evidence for this PG has come from various experiments which include angle resolved photoemission spectroscopy (ARPES), optical conductivity and tunneling spectroscopy.^{1,2} For the hole-doped high- T_c superconductors (e.g., BSCCO) these measurements show that the superconducting gap (Δ_{SC}) has a similar width as the PG. This leads many to believe that the PG and the Δ_{SC} have a similar origin. Theories predicting pair correlations above T_c have been suggested, which are supported by these experiments. The other important property of the PG is its doping dependence. The PG appears at higher temperatures for the underdoped compounds and is not observed in overdoped compounds.² The situation is less clear for other cuprates such as the hole-doped system $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO). The width of the PG in LSCO from ARPES (Ref. 3) and optical conductivity⁴ measurements comes out to be much larger than the width of Δ_{SC} .⁵ These results raise questions about the origin of the PG and more experiments on the normal state of the cuprates are necessary.

A lot of interest has been generated recently by the electron doped counterparts of the high- T_c cuprates. These materials have the general formula $R_{2-x}\text{Ce}_x\text{CuO}_4$ ($R = \text{Nd, Pr, Sm}$). These materials were believed to have an s -wave superconducting gap symmetry but recent results have shown that the gap has a d -wave symmetry similar to the hole-doped cuprates.⁶⁻⁸ A comparison between the properties of the hole-doped and electron doped systems is needed as input for theories of high- T_c superconductivity. In this context it is important to investigate the normal state of the electron-doped materials and check if results similar to the hole-doped cuprates are obtained, e.g., the formation of a PG in the normal state. Transport measurements have shown that

there is a metal-insulator crossover as a function of Ce doping in the normal state of the electron-doped cuprate $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$ (PCCO).⁹ Recently tunneling spectroscopy measurements have been reported in the normal state of PCCO and NCCO.^{10,11} These reports give evidence for a normal state gap for fields above the bulk H_{c2} and $T < T_c$. In Ref. 11 the authors suggest that this normal state gap could be due to the presence of a PG in the normal state of the electron doped superconductors. However, the width of this gap is not consistent with other recent measurements using ARPES and optical conductivity which show normal state gaps of 60 meV and wider.^{12,13} Further work is therefore necessary to explain the origin of this normal state gap and the differences between the properties of the hole and electron-doped systems.

In this paper we report our tunneling studies of the normal state of the electron doped cuprate superconductor $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$ for different values of the doping x . Preliminary work was reported in Ref. 10. For PCCO H_{c2} is of the order of 10 tesla for $x = 0.15$ at 1.5 K. This makes it possible to obtain the normal state by applying a field greater than H_{c2} and to then perform the tunneling studies. In hole-doped cuprates the H_{c2} is much higher (≥ 50 T for optimally doped samples) and the tunneling studies which have been performed in the normal state are for $T > T_c$. We find that in the normal state, there is a depletion of the density of states (DOS) near the Fermi level (E_F). Further investigation reveals that this normal state gap (NSG) closely resembles the correlation gap formed in disordered metals due to enhanced electron-electron interactions. We present data which show that this NSG is not due to residual superconductivity at the surface for fields above the bulk H_{c2} . The formation of this NSG gives us new information about the nature of the charge carriers in the normal state and hence about the pairing mechanism.

There have been several tunneling studies reported on the electron-doped cuprates but the results have been inconsistent *vis a vis* the values of the SC gap, the observation of the zero bias conductance peak formed due to Andreev bound states, etc.¹⁴⁻¹⁶ The standard techniques used for other cuprates such as YBCO, e.g., multilayer junctions, are difficult to use for the electron doped materials because (1) a -axis

oriented superconducting films of PCCO or NCCO have not been grown yet and (2) the extreme sensitivity of these compounds to the oxygen stoichiometry which makes it difficult to fabricate tunnel junctions without changing the superconductor in some way. In this paper we report tunneling data on three different types of junctions and show that they all give consistent results.

II. EXPERIMENTAL DETAILS

The tunneling studies were performed mainly on *c*-axis oriented thin films of PCCO grown using pulsed laser deposition (PLD) on LaAlO_3 (LAO), and yttrium stabilized zirconia (YSZ) substrates. Details of the film growth are given in Refs. 17,18. The films have been optimized for oxygen content by maximizing T_c for each cerium concentration. The films were characterized by x-ray, ac susceptibility, and resistivity measurements.

We have used three techniques to tunnel into the *a-b* plane of *c*-axis oriented thin films. In the first technique, a thin film of *c*-axis oriented PCCO, covered with SiO_2 , is broken in vacuum to expose the *a-b* plane while Ag is being evaporated in the vacuum chamber. A junction is thus formed between the freshly exposed PCCO surface and silver. The size of the junction is reduced to about $200 \mu\text{m}$. This is a new method of forming junctions to tunnel into the *a-b* plane of PCCO details of which will be discussed in a future publication. We will call these “break junctions” in the subsequent sections although they are not break junctions in the usual sense. The second technique is very similar to the break junction method. Here the *c*-axis oriented film is covered with SiO_2 and broken in air. Indium was pressed on the freshly exposed side of the film. We used indium to check the quality of the junction by recording the tunneling features of indium. These will be called “pressed junctions.” The third method was the usual point contact junction formed by pressing a gold electrode on the side of a single crystal or a freshly broken thin film such that the direction of current flow is perpendicular to the *c* axis.

III. RESULTS AND DISCUSSIONS

We first show the tunneling data on the optimally doped PCCO sample ($x=0.15$). Figure 1(a) shows the data using the break junction method. The superconducting gap is clearly visible and results in a bunching of states near $\pm\Delta_{\text{SC}}$ (coherence peaks). The value of Δ_{SC} is about 5 meV which is similar to the values reported in earlier tunneling studies.^{14,5} The conductance ($G=dI/dV$) within the gap voltage does not go to zero because of two possible reasons: (1) the junctions have more than one conduction channel, i.e., are not tunnel junctions but ballistic point contact junctions and (2) the likely *d*-wave symmetry of the SC gap. The effect of the first point has been discussed in detail in the classic paper by Blonder, Tinkham, and Klapwijk (BTK) (Ref. 19) for the case of isotropic *s*-wave superconductors. In the BTK analysis the junction was parametrized by a quantity Z which depends on the scattering at the interface between the metal and the superconductor. High transparency junctions have $Z\sim 0$

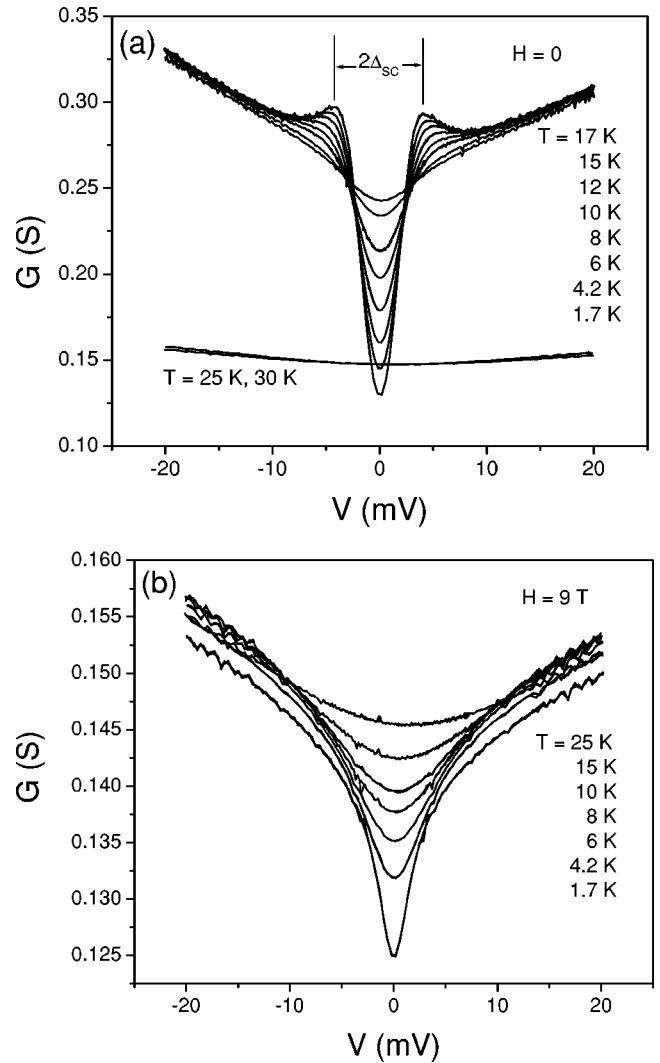


FIG. 1. (a) The G - V curves for the break junction between silver and PCCO ($x=0.15$). The superconducting gap is marked. The curves above T_c are shifted due to the resistance of the film in series with the junction. (b) G - V curves for the same junction in (a) in a magnetic field of 9 T applied parallel to the *c* axis. A prominent gap in the normal state is seen at low temperatures.

and tunnel junctions have $Z\gg 1$. For superconductors with an anisotropic gap a modified BTK model has been described in Ref. 20. A detailed fitting of our data to such models is in progress. However, in the following sections we will use these ideas qualitatively. Figure 1(a) also shows the variation of the tunneling spectra with temperature. This variation is due to a combined effect of the thermal smearing of the spectra and the variation in the density of states (DOS) of the SC with temperature. The Δ_{SC} does not change noticeably with temperature. The gap structure disappears above T_c and the background conductance of the junction is observed for $T=25$ K.

When a magnetic field $H>H_{c2}$ is applied to this tunnel junction at $T=1.7$ K ($T\ll T_c$), the SC gap is no longer observed as expected (i.e., the coherence peaks disappear), *but surprisingly* the gap at the E_F persists. However, the shape of this gap as a function of energy is quite different [Fig. 1(b)]

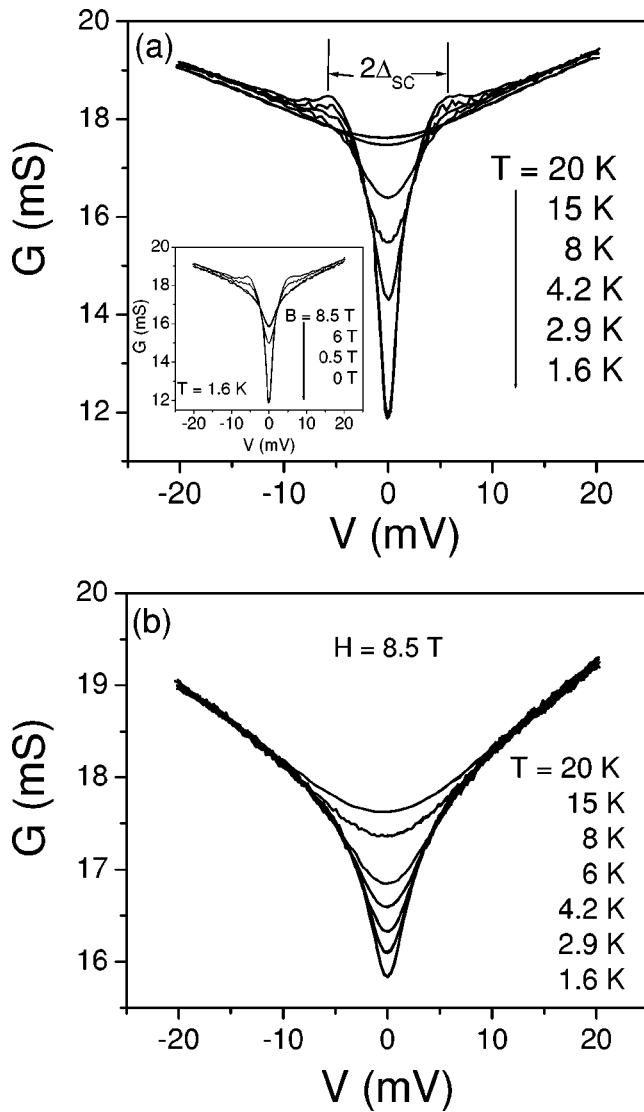


FIG. 2. (a) The G - V curves for the pressed indium junction with PCCO ($x=0.15$). The superconducting gap is marked. The inset shows the effect of the indium gap on the G - V curves. (b) The G - V curves for the same junction in (a) in a magnetic field of 8.5 T applied parallel to the c axis. The normal state gap is clearly visible.

from the SC gap. We call this unexpected gap the normal state gap (NSG), since resistivity measurements at this magnetic field show that the PCCO film is in the normal state. Before we further analyze this NSG we present data which show that the observation of this NSG does not depend on the type of tunnel junction. Figure 2(a) shows data for a pressed junction using an indium counterelectrode. The indium was used to check the quality of the junction. The contribution of the indium density of states is visible in the zero field data, since when a field of about 5000 G was applied to make the indium electrode normal there is a large increase in the zero bias conductance which indicates that the indium has become a normal metal with no superconducting gap [inset Fig. 2(a)]. In the superconducting state of indium the G at zero bias is not zero which shows that this junction is not in the tunneling limit. We find $Z \approx 0.7$ according to a rough estimate made from the ratio of the conductance at 20

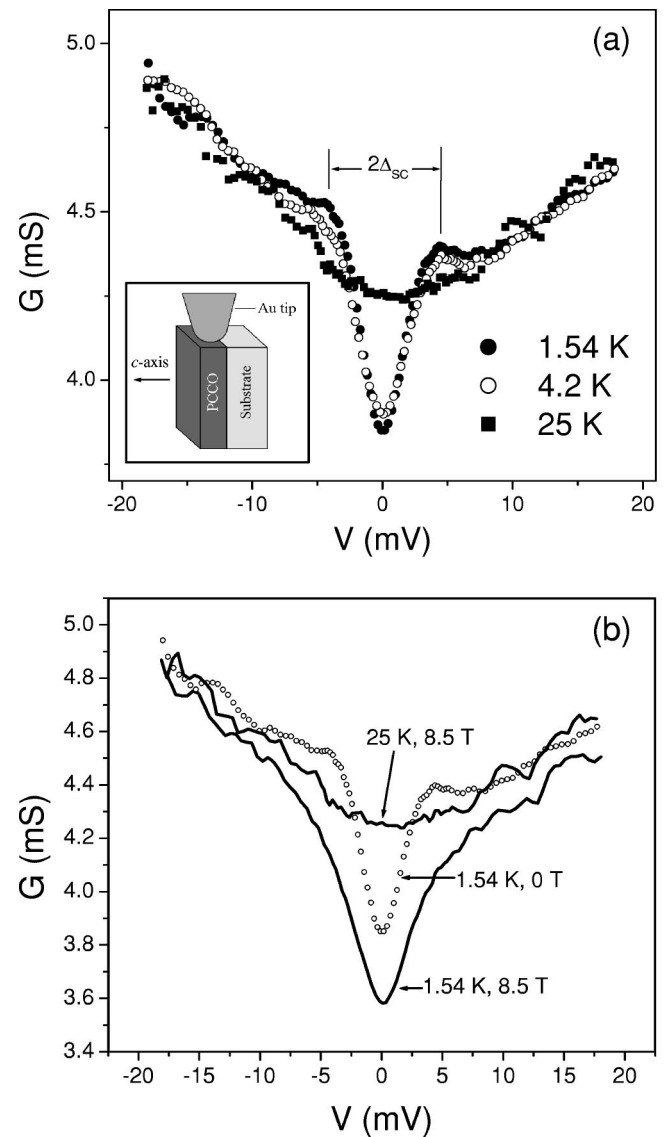


FIG. 3. (a) The G - V curves for the point contact junction between PCCO ($x=0.15$) and gold. The superconducting gap is marked for the curve taken at 1.54 K. The inset shows the setup for the formation of the point contact junction between PCCO and gold for tunneling into the a - b plane. (b) The normal state gap is seen when a field of 8.5 T is applied parallel to the c axis.

mV (G_{20}) and at 0 V (G_0).¹⁹ For fields above 5000 G the contributions to the tunneling spectra will be dominated by the DOS features of PCCO. For an applied field of 8.5 T the NSG is clearly seen for this junction too [Fig. 2(b)]. In Fig. 3(a) we show the tunneling data obtained from point contact tunnel junctions formed between a gold tip and a PCCO thin film. The zero field data clearly shows the gap feature for superconducting PCCO. The temperature dependence of the tunneling spectra is similar to that observed for the other two types of junctions. On the application of a magnetic field of 8.5 T we again observe the NSG [Fig. 3(b)]. From these data we conclude that this anomalous NSG is present irrespective of the kind of tunnel junction.

We now discuss the nature of this NSG in more detail.

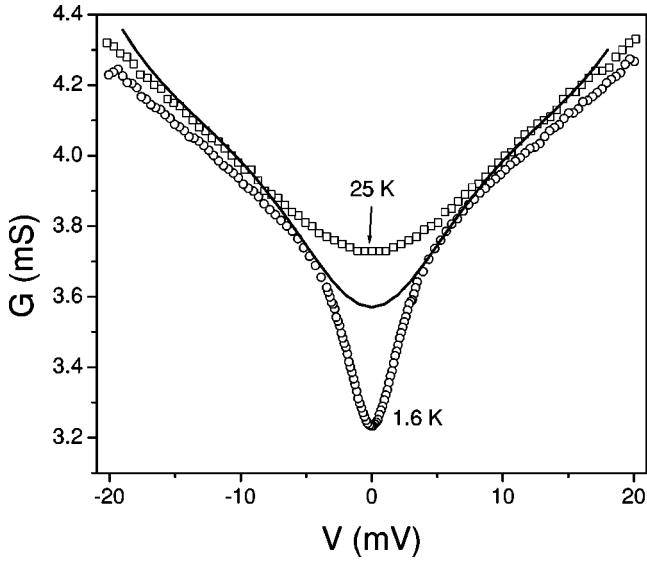


FIG. 4. The effect of thermal smearing on the normal state gap in the tunneling spectra. The normal state gap at 1.6 K (open circles) evolves to the solid line at 25 K due to thermal smearing [calculated using Eq. (1)]. This calculated curve at 25 K does not match the experimental curve at 25 K (open squares). Data shown is for $x=0.15$ composition using the break junction method, in a field of 8.5 T.

First we look at the behavior of this gap as the temperature is increased upto and beyond T_c . The T_c for our optimally doped PCCO films is about 21 K. We see that the NSG becomes indistinguishable from the background for $T > T_c$ [Figs. 1(b), 2(b), and 3(b)]. This could be interpreted as evidence for the NSG being due to some residual superconductivity (even for $H > H_{c2}$) which goes away for $T > T_c$. However we have to consider that the width of the NSG is ~ 7 meV and a temperature of 25 K can smear out features of about 5 meV in the tunneling spectra. To check if the NSG closes around $T = T_c$ just due to thermal smearing we have made a simple estimate of the role of thermal smearing in the temperature evolution of the G vs V curves using the following equation for the tunneling current:²¹

$$I(V, T) = c \int_{-\infty}^{\infty} n\left(E + \frac{eV}{2}\right) \left[f\left(E + \frac{eV}{2}, T\right) - f\left(E - \frac{eV}{2}, T\right) \right] dE, \quad (1)$$

where $n(E)$ is the DOS of the PCCO sample at zero temperature, $f(E, T)$ is the Fermi function at a temperature T , and V is the junction bias. For $n(E)$ we have taken the $G(V)$ vs V curve at 1.6 K and smeared it using Eq. (1) for a temperature T . We have assumed that the density of states of the counterelectrode gold is a constant for the relevant energy range and so is the tunneling barrier for this range of energies both of which are included in the constant c . The results of these calculations are shown in Fig. 4. It shows that thermal smearing alone cannot explain the disappearance of the NSG for $T \sim 20$ K. This means that the NSG has a significant thermal dependence of its own. With this analysis we

have shown that although the thermal smearing of the tunneling spectra has a significant influence on their evolution with temperature the NSG does indeed become smaller with increasing temperature. This thermal dependence of the NSG could be due to residual SC, or it could be that there is no residual SC and the NSG has its own thermal dependence due to the temperature dependence of the interactions which are responsible for the suppression of the DOS at E_F . The possible origin of these interactions will be discussed later in this section.

To study the possible contribution to the NSG of residual SC at the interface and/or the tunnel barrier, we formed a high transparency point contact junction between a gold tip and an $x=0.13$ thin film of PCCO. Figure 5(a) shows the G - V data at various applied fields for such a junction, at 1.6 K. The peak formed at zero bias (for $H < H_{c2}$) is due to Andreev reflection which is observed in high transparency junctions. At $H=0$ the width of the feature is about $2\Delta_{SC}$ (~ 10 meV) and $G(0)/G_N$ is 1.62 [Fig. 5(a) inset], where G_N is the value of $G(V)$ for $eV > \Delta$. These are clear signatures of Andreev reflection (AR) occurring at the interface between gold and PCCO. This also shows that for this junction $Z \sim 0$. We chose the $x=0.13$ composition because it has a low T_c of 12 K and therefore we get a good temperature range to operate in before thermal smearing washes out the features in the $G(V)$ vs V curves. When a magnetic field is applied on this junction, the AR feature reduces in height and also shrinks in width and when the superconductivity is destroyed, disappears completely. This is a clear indication that there is no residual SC at the interface in our G - V curves. However, the NSG is clearly visible. In fact for intermediate values of H the normal state density of states reveals itself at higher V while at the same time there is peak at zero bias due to Andreev reflection, as shown in Fig. 5(b). Since this is a high transparency junction, the junction barrier has negligible effect on the shape of the curve. This is clear from Fig. 5(a) since for $H=0$ the G - V curve is almost flat (independent of V) for $eV > \Delta$.

The other test to check if the NSG is truly a feature of the normal state is to increase the temperature beyond T_c and observe the evolution of the $G(V)$ vs V curves. Figure 5(c) shows that removing SC by increasing the temperature has an evolution similar to that of increasing H beyond H_{c2} (for $T < T_c$). However, the presence of the NSG is less clear for $T > T_c$ due to thermal smearing. This strongly suggests that the NSG is indeed a normal state feature. We should add here that the high transparency junctions are not tunnel junctions but point contact junctions with more than one channel for conduction. However we are still in the ballistic region, i.e., the junction size, given by a radius a , is smaller than the elastic mean free path of the electron l_e (the Sharvin limit). We know this must be the case because at $T \sim 20$ K, $G(V)$ has a minimum at $V=0$.²² If the junction were in the diffusive limit ($a > l_e$) then the curvature of the tunneling spectra in the normal state would change and there would be a maximum in the G - V curve at $V=0$. Hence the features observed in the G - V curves in the normal state do reflect the DOS features of the normal state. From this experiment we thus

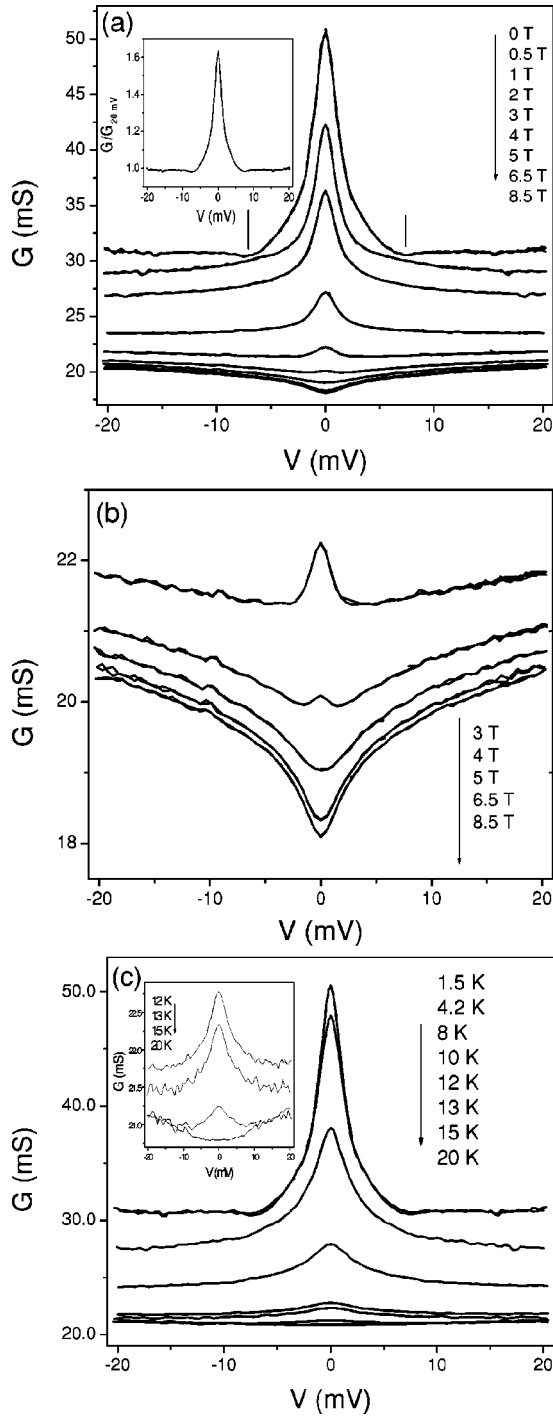


FIG. 5. (a) The G - V curves for a high transparency ($Z \sim 0$) point contact junction between PCCO ($x=0.13$) and gold. A peak due to Andreev reflection (AR) is seen near zero bias. The inset shows the increase in G by a factor of 1.62 at zero bias compared to the G at 20 mV. When a field is applied, the AR peak shrinks both in height and width and disappears for fields ≥ 5 T. (b) An enlarged view of the G - V curves at high magnetic fields. As the AR peak shrinks and disappears the normal state DOS is seen, even when there is a peak at zero bias, for $|V| > \Delta_{SC}$. The normal state gap is again seen for $H \geq 5$ T. (c) The variation of the G - V curves with temperature. The normal state G - V curve is seen at 20 K. The inset shows an enlarged view of the G - V curves at high temperatures.

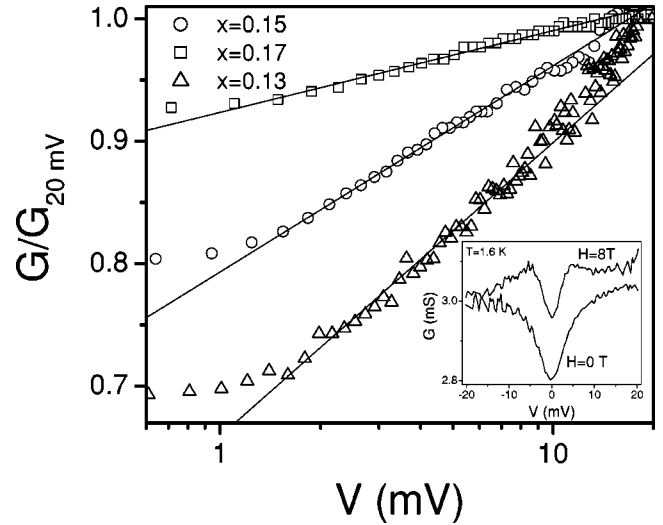


FIG. 6. A plot of G vs $\ln V$ in the normal state of underdoped ($x=0.13, T=1.61$ K), overdoped ($x=0.17, T=1.6$ K), and optimally doped ($x=0.15, T=1.54$ K) compositions of PCCO. Data for positive bias is shown. The solid lines are linear fits to the data for a certain voltage range. The inset shows tunneling spectra for PCCO ($x=0.17$) at 1.6 K, in the superconducting ($H=0$) and normal ($H=8.5$ T) states.

draw two important conclusions: (1) the NSG is definitely a normal state feature and (2) it can be observed for $H > H_{c2}$ as well as for $T > T_c$.

What is the origin of this NSG? Such features in the density of states have been observed before in various systems such as disordered metallic films²³ and perovskite oxides such as $\text{LaNiO}_{3-\delta}$ (Ref. 24) using tunneling spectroscopy. The appearance of these features in the density of states of these materials is due to electron-electron Coulomb interactions which are enhanced due to disorder in the materials. Such effects lead to anomalies in the DOS $n(E)$ near E_F . For three-dimensional systems this anomaly is of the form $n(E) = n(0)[1 + \sqrt{E/\Delta}]$, where Δ is called the correlation gap. For two-dimensional systems the electron-electron interaction leads to decrease in the DOS near E_F given by $\delta n/n \sim -\ln(E/E_0)$.²⁵ These anomalies are the precursors of the insulating phase ‘‘Coulomb gap’’ which is formed at E_F when the disorder is increased and the states near E_F are localized.

The effects mentioned above have been observed in Fermi liquid systems with varying degrees of e - e correlation and disorder. Although the normal state of a cuprate superconductor shows non-Fermi-liquid behavior, in the following we have used the ideas developed for Fermi liquids in our attempt to understand the origin of the NSG. Figure 6 shows a plot of the $G(V)$ as a function of $\ln V$ ($V > 0$) taken at $T = 1.6$ K using a gold and PCCO point contact junction. $G(V)$ is a linear function of $\ln V$ for a large range of V [for $V < 1$ mV, $G(V)$ deviates from the straight line due to thermal smearing]. This behavior follows closely the abovementioned signature of a 2D correlation gap.^{23,26} In Ref. 23 it is shown that as the sheet resistance of the film is increased, the logarithmic anomaly at zero bias becomes deeper. We want

to verify if a similar trend is observed in the normal state of PCCO as in the case of disordered metal films. The sheet resistance in the normal state of PCCO increases as x is lowered.⁹ Moreover, the field induced normal state in PCCO has a metallic behavior for overdoped samples ($x > 0.15$) and the resistivity shows an upturn at low temperatures for optimally doped and underdoped samples ($x \leq 0.15$).⁹ A suggested mechanism for this transition was the onset of weak localization below a certain doping. This transition suggests an increase in the electron-electron interactions as x is decreased. If our hypothesis about effect of interactions on the DOS is correct then the tunneling data should reflect this transition. The inset of Fig. 6 shows the point contact tunneling data for an $x = 0.17$ sample of PCCO ($T = 1.6$ K). At zero field the superconducting gap is clearly seen. At 8.5 T the SC gap gives way to the NSG. The NSG in this case is smaller than the one found for $x = 0.15$. Figure 6 also shows the $G_{\text{norm}} - \ln V$ plots for the $x = 0.17$ (at $T = 1.6$ K) and $x = 0.13$ (at $T = 1.61$ K) samples. The deepening of the NSG with lowering of x is clearly indicated by the increase of the slope of the G_{norm} vs $\ln V$ curves and is similar to the trend observed in metallic films with different amounts of disorder.^{23,26} Therefore our tunneling data shows that in the underdoped region there are strong electron-electron interactions which results in a larger NSG. As the doping is increased the effect of the interactions is lowered and a trend towards a more conventional Fermi-liquid-like normal state is obtained as seen from a much smaller NSG. It has been conjectured that for some value of x there is quantum critical transition in the normal state of cuprate superconductors.²⁷ The gradual disappearance of the NSG is an indication that the physical properties of the cuprates are going through a gradual crossover with increasing x , probably due to their proximity to this quantum critical point.

The data discussed above show that there is a NSG in the normal state of PCCO. This gap shows the properties of a 2D correlation gap and is related to the position of the material in the PCCO phase diagram, i.e., it depends on the doping. The pseudogap (PG) is a gap in the normal state of cuprate superconductors and it also depends on the doping.² In fact the other similarity between the NSG observed here and the PG is that the gap becomes smaller for the overdoped compounds. But is our observed NSG the same as the pseudogap (PG)? In Ref. 11 the authors claim that the NSG they observe in PCCO and NCCO is the pseudogap and they also estimate field values at which this NSG is suppressed. However there is serious discrepancy in the energy scale of the pseudogap observed in Ref. 11 and in our experiments with that observed in other experiments. Optical conductivity data on LSCO (a hole-doped cuprate superconductor similar to PCCO) show a PG of about 500 cm^{-1} , i.e., about 62 meV .⁴ New optical conductivity data on e -doped NCCO gives a large pseudogap width of about 600 cm^{-1} which survives

even above room temperature.¹³ Recent ARPES data also shows evidence of a pseudogap larger than 50 meV .¹² In nonsuperconducting NCCO pseudogaps of about 200 meV have been observed from optical conductivity measurements.²⁸ Although the width of our NSG, which is a “soft” correlation gap, cannot be uniquely determined, the energy scale of this gap is much smaller than the gaps observed in optical conductivity and ARPES experiments. Therefore for the e -doped cuprates it is unlikely that these gaps have the same origin. In fact the NSG observed in our tunneling measurements evolves from the Δ_{SC} as the sample is driven normal above H_{c2} . This behavior is similar to that observed in tunneling spectra of the hole doped cuprate BSCCO where a normal state gap evolves smoothly from the Δ_{SC} as T is increased above T_c .² This gap above T_c is believed to be the PG. However, for BSCCO the width of the gap is $\sim 50 \text{ mV}$ which agrees quite well with the ARPES and optical conductivity data.² These facts show that although the NSG we observe displays behavior similar to that of the PG in the normal state of hole-doped cuprate superconductors, the reasons for the discrepancies of the energy scale of the gap between different experiments (for the e -doped case) is still an open question.

There is only a small variation of the NSG with magnetic field in our experiment and for the maximum field values that we applied ($\sim 9 \text{ T}$) the NSG was still prominent. Further experiments are necessary to determine accurately the variation of the NSG with T and H .

IV. SUMMARY AND CONCLUSIONS

In this paper we have shown that the density of states of the normal state of PCCO shows a depletion of states near the Fermi level E_F . This normal state gap can be treated as a “soft” correlation gap which is formed due to electron correlations. Such gaps have been seen in disordered metals using tunneling spectroscopy. We have argued that this gap is not the same gap observed in optical conductivity and ARPES measurements mainly due to a difference in the energy scales. We have shown that the nature of the NSG depends on the doping, i.e., it depends on the position in the phase diagram. We conjecture that this NSG is linked with a quantum phase transition which most probably occurs in the normal state of the cuprates for a certain value of hole or electron doping.

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