

Tunneling into the Normal State of $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$

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The temperature dependence of the tunneling conductance was measured for various doping levels of $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$ using planar junctions. A normal state gap is seen at all doping levels studied, $x = 0.11$ to $x = 0.19$. We find it to vanish above a certain temperature T^* . T^* is greater than T_c for the underdoped region and it follows T_c on the overdoped side. This behavior suggests finite pairing amplitude above T_c on the underdoped side.

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In conventional superconductors, below a certain temperature T_c , pairs of opposite spins and momenta are formed. At that temperature, these pairs also condense into a state of zero resistivity when an energy gap, Δ , opens up in the density of states. This seems not to be the case in the high- T_c cuprates. For the hole-doped cuprates, Renner *et al.* [1] have shown that a gap in the density of states persists above T_c , especially for underdoped cuprates. This behavior was referred to as the pseudogap state [2]. One of the interpretations put forth to explain this phenomenon was in terms of incipient superconductivity above T_c with a lack of phase coherence [3]. Another possibility is that the pseudogap is due to some order not necessarily following T_c on the overdoped side [4].

On the electron-doped side of the phase diagram, tunneling experiments showed a gap in the density of states when the normal state was accessed by applying magnetic fields higher than the minimal field needed to quench superconductivity, H_{c2} [5,6]. Alff *et al.* [7] later showed that this gap vanishes at a doping dependent temperature (T^*) lower than T_c for doping levels close to the optimum. This appeared to rule out the possibility of preformed pairs in the electron-doped cuprates and suggested that this gap was related to an order parameter that competes with the superconducting one. Recently, we found evidence for a quantum phase transition near $x = 0.165 \pm 0.005$ from transport measurements on $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$ (PCCO) [8]. It is tempting to relate this phase transition with the gap feature observed in the normal state tunneling spectra. However, unlike the hole-doped cuprates, in the electron-doped cuprates a broad region of antiferromagnetism in the phase diagram extends up to very high dopings, possibly into the superconducting dome [9,10]. This suggests that the quantum phase transition we have observed is likely to be an antiferromagnetic (AFM) to a paramagnetic transition. On the other hand, we are going to show here that the normal state tunneling gap (NSTG) persists even in the highly overdoped $x = 0.19$ sample, i.e., way beyond the AFM region. This suggests that the NSTG is unrelated to the quantum phase transition we previously reported and probably not directly related to the AFM phase. The NSTG

has a typical amplitude of about 5 meV. It is quite distinct from the large gap (of the order of 100–200 meV) seen in angle-resolved photoemission spectroscopy (ARPES) [11]. The large gap has also been observed in optical reflectivity [12] and correlated with the AFM phase, i.e., a spin density wave gap.

In this Letter, we report an extensive and systematic tunneling study into PCCO as a function of field and temperature for many Ce doping levels: $x = 0.11, 0.13, 0.15, 0.16, 0.17, 0.18, \text{ and } 0.19$. We find the NSTG temperature, T^* , to be greater than T_c for $x < 0.17$ and $T^* \approx T_c$ for $x \geq 0.17$. This is a very different behavior than found previously [7]. Our data suggest that pairs are formed and condensed at the same temperature for $x \geq 0.17$, as in conventional superconductors.

Lead (Pb) contacts, approximately 5000 Å thick, were evaporated on fresh PCCO films and on faces parallel to the c axis (ab faces) of freshly cleaved $\text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ single crystals. The junctions' area was approximately $0.5 \times 0.5 \text{ mm}^2$ for the films and $1 \text{ mm} \times 30 \mu\text{m}$ for the crystals. This results in a true tunneling contact. In both cases, the PCCO surface is exposed to ambient atmosphere for no more than a few minutes. It has been demonstrated in Al/YBa₂Cu₃O_{7- δ} contacts that the aluminum counter-electrode takes oxygen from the cuprate, thus creating an oxide barrier [13]. A similar process occurs in PCCO/Pb contacts [14]. To prevent overreduction of oxygen in the vicinity of the junction, we use slightly oxygen rich films. In such a case, there could be a slight difference of oxygen concentration between the bulk film and the junctions' area. For that reason the T_c report here is measured at the junction using the tunneling characteristics itself. The bulk T_c measured in resistivity and the T_c measured by the tunneling conductance differ by less than 2 K. $I(V)$ characteristics were measured using a current source and a voltmeter and were differentiated digitally. The conductance characteristics of junctions made on films and on crystals of similar doping level show no difference. This indicates that tunneling into the films, although nominally c axis oriented, is predominantly in-plane tunneling due to

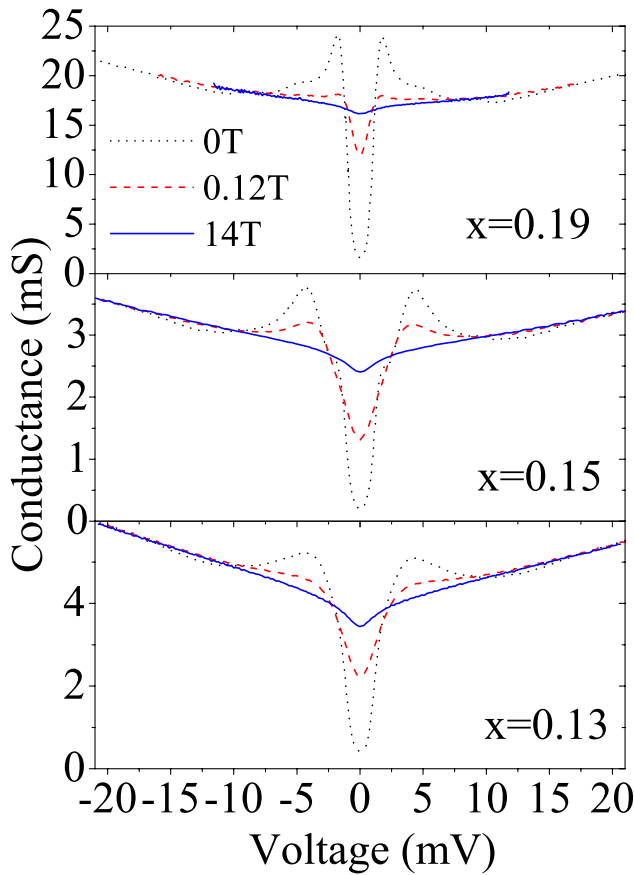


FIG. 1 (color online). The conductance vs voltage for Pb/PCCO junctions for three typical dopings: $x = 0.13$, $x = 0.15$, and $x = 0.19$ at $T = 1.8$ K. The magnetic field is applied parallel to the c axis: dotted black lines, $H = 0$; dashed red lines, $\mu_0 H = 0.12$ T (larger than the Pb critical field); solid blue lines, $\mu_0 H = 14$ T (both electrodes are normal). Note the normal state gap in all three doping levels.

ab plane facets exposed to Pb. The magnetic field is always applied along the c direction.

In Fig. 1 we show conductance vs voltage for three different doping levels at 1.8 K. The dotted black lines are zero field measurements. We note that the low conductance at zero bias indicates good tunneling contacts with negligible leakage currents. The strong Pb and PCCO coherence peaks and the Pb phonons can be clearly seen. The dashed red lines are measurements taken above the Pb critical field (0.1–0.12 T). Only the PCCO coherence peaks can be seen and the Pb phonons are absent. At $H > H_{c2}$ (solid blue lines), superconductivity in the PCCO electrode is quenched, yet a small NSTG feature remains. It is seen as a depression of the conductance at zero bias. It exists in *all* doping levels studied.

Since the $x = 0.11$ sample is not superconducting, we are able to follow the magnetic field dependence of the NSTG from 0.12 T (when the Pb becomes normal) to 14 T, as shown in Fig. 2(a). In Fig. 2(b) we show the conductance

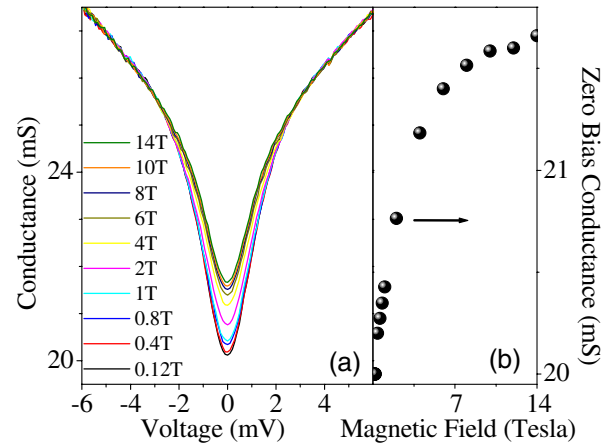


FIG. 2 (color online). Nonsuperconducting $\text{Pr}_{1.89}\text{Ce}_{0.11}\text{CuO}_4$ sample at $T = 1.8$ K: (a) conductance vs voltage at various fields; (b) zero bias conductance vs magnetic field taken from (a). The normal state gap exhibits magnetic field dependence saturating above 8 T.

at zero bias as a function of magnetic field applied parallel to the c direction. The NSTG partially closes at low fields, saturating above ~ 8 T. The low field range where most of the change occurs is hidden by the superconductivity in the other samples. We are not aware of theoretical prediction for such a field dependence. Further experimental and theoretical studies are needed in order to find the origin of this field dependence.

The temperature dependence of the NSTG at 12 T for $x = 0.15$ single crystal is shown in Fig. 3. It can be seen that the NSTG closes with temperature. A linear background independent of temperature is observed at high biases. We used this background and its extrapolation to zero bias for the analysis of the temperature dependence of the NSTG as described below. We made conductance vs voltage measurements at various temperatures for all dop-

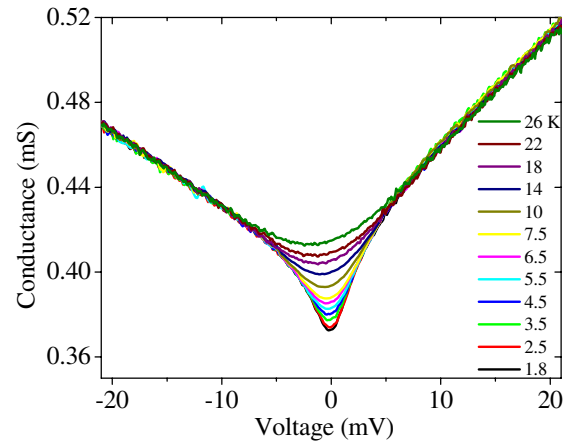


FIG. 3 (color online). Conductance vs voltage for $\text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ crystal at 12 T applied along the c axis and at various temperatures.

ing levels studied. The NSTG temperature T^* is determined by analyzing the temperature dependence as depicted in Fig. 4. The temperature has two effects on the conductance. The first effect is thermal smearing resulting from the broadening of the Fermi function with increasing temperature. The second effect is a possible decrease in the NSTG amplitude. Both effects result in decreasing the gap features. To isolate the latter effect, we do the following: at any temperature (10 K is shown as an example) we calculate, taking into account thermal broadening effects, the expected conductance at, say, 10 K assuming the NSTG is constant with temperature. In this case, the temperature dependence of the conductance can be calculated using $g(eV, T_1) \propto \int g(E, T=0) \frac{\partial f(T_1, E - eV)}{\partial E} dE$ [dashed black line in Fig. 4(a)]. Since we do not have the zero temperature conductance data, we use the 1.75 K data instead. This is a slight overestimation of the thermal smearing since the 1.75 K data are already somewhat smeared. As a background we use the linear approximation (see Fig. 3), which is the simplest possibility for such a background. Plugging this linear background (at $T = 0$) into the thermal smearing integral above, we obtain the background at any temperature needed (the temperature affects only the low bias region). For example, at 10 K for the $x = 0.15$ sample one obtains the solid green line in Fig. 4(a). Now, for example, at 10 K [Fig. 4(a)] we compare the real measurement (dotted red line) with the 1.75 K data smeared to 10 K (dashed black line) and the linear background smeared to 10 K (solid green line). We define the relative depth of the NSTG as the ratio a/b [see Fig. 4(a)] and we follow it as a

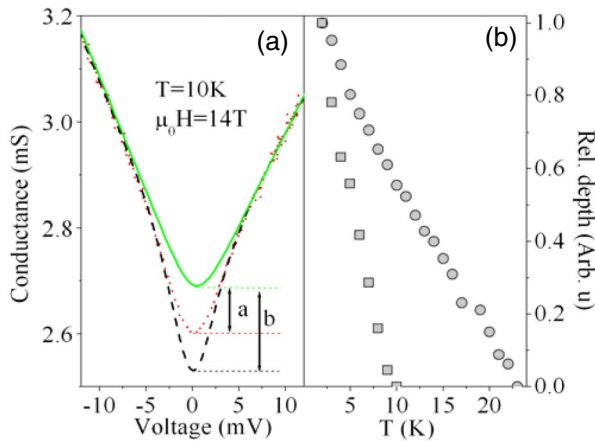


FIG. 4 (color online). (a) $\text{Pr}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ junction conductance. Black dotted line is the 1.75 K data smeared to 10 K assuming that thermal smearing is the only effect of temperature. Red dotted line is the real data at 10 K. Green solid line is the linear background extrapolated from the high bias measurement and thermally smeared to 10 K. The ratio a/b is defined as the relative depth of the gap. (b) The relative gap depth as a function of temperature for the $x = 0.15$ sample (circles) and for the $x = 0.19$ samples (squares). T^* is defined as the temperature at which the relative depth goes to zero.

function of temperature. It decreases monotonically to zero [see Fig. 4(b)] with increasing temperatures. The temperature at which the ratio a/b goes to zero is defined as T^* . In Fig. 4(b) we show the temperature dependence of the relative depth of the NSTG for two doping levels: $x = 0.15$ and $x = 0.19$. We did a similar analysis for all the doping levels under study. The results for T^* are summarized in Fig. 5 (black circles).

We also show in Fig. 5 (red squares) the critical temperature, T_c . A slight oxygen variation between the bulk film and the vicinity of the junction may result in a small difference between the bulk transition temperature and the local T_c at the junction. This variation is due to oxygen taken from the PCCO film to create the oxide barrier. Therefore, the superconducting transition temperature, T_c , was measured at the junction by comparing, at each temperature, the zero field tunneling measurement to the 14 T spectrum. T_c was defined as the temperature at which these two spectra overlap. We note that at low doping levels $T^* > T_c$, while these two temperature scales become closer around optimum doping and follow each other on the overdoped side. This behavior is totally different than that reported by Alff *et al.* [7]. Their T^* was found to be lower than T_c in the vicinity of optimum doping extrapolating to zero at $x = 0.17$.

We shall now discuss the possible origin of the NSTG. Biswas *et al.* [6] suggested that the NSTG results from electron-electron interactions. This should manifest itself as suppression of the tunneling density of states at zero bias, with a conductance which is logarithmic in voltage at low biases [15]. However, if electron correlations are responsible for the NSTG, it is rather surprising that T^* follows T_c on the overdoped side. Moreover, such correlations also result in $\ln(T)$ conductivity [16]. By contrast, in our overdoped films a metalliclike conductivity is observed [8] where the NSTG is still seen.

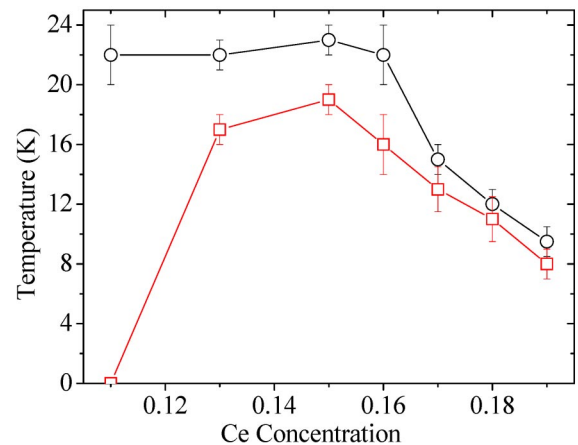


FIG. 5 (color online). Black circles: T^* , the temperature at which the normal state tunneling gap vanishes, determined using the analysis described in the text. Red squares: T_c of the junction (see text for detail).

A second possibility is that the NSTG is due to a partial gapping of the Fermi surface in the AFM normal state characterizing PCCO [9,12]. Tunneling into the AFM state of chromium showed conductance with weak features at zero bias [17] or sometimes even featureless conductance [18]. If the NSTG we observe is due to antiferromagnetism, then it is puzzling why we still observe it at doping levels higher than $x = 0.17$ where neutron scattering experiments show no antiferromagnetism [10].

A third possible explanation for the NSTG is nonequilibrium tunneling due to nonzero electron relaxation times [19]. However, the depression of the zero bias conductance in that case is typically of the order of 0.3%, much smaller than the feature we report here.

Alff *et al.* [7] suggested that the pseudogap in the electron-doped cuprates is due to a hidden order under the superconducting dome. Their data suggested that T^* goes to zero around $x = 0.17$. Recently, we showed evidence for a quantum phase transition in PCCO at a critical doping $x = 0.165 \pm 0.005$ [8]. The data presented by Alff *et al.* appear to be consistent with such a transition. However, the extensive doping study and the simpler data analysis we present here suggest that the NSTG is not a signature of the order parameter that vanishes at $x_c = 0.165$. This vanishing order is more likely to be antiferromagnetism as suggested by other experiments [9,10,12].

Our data show $T^* \geq T_c$ in contrast with Alff *et al.* [7]. The data we present here suggest a preformed singlet scenario. A nonvanishing pairing amplitude exists up to $T^* > T_c$ in the underdoped region of the phase diagram, while $T^* \approx T_c$ in the overdoped region. This is consistent with previous Nernst effect data, suggesting a broader fluctuation region on the underdoped side [20]. Unlike the hole-doped cuprates, T^* does not increase when the doping is decreased from the optimum level [21]. This can be explained by the proximity of the AFM phase possibly persisting into the superconducting dome. The AFM channel competes with the pairing channel and hence the saturation in T^* [22]. However, we note that, although the NSTG vanishes at $T^* \approx T_c$ on the overdoped side, its field dependence is rather different from the superconducting gap. While the superconducting gap vanishes at $H > H_{c2}$, the NSTG still persists up to fields as high as 14 T. This needs to be understood in the future.

The NSTG has a characteristic energy scale of about 5 meV at 2 K. This energy is much smaller than the “large gap” seen in ARPES or optics. The relation between these two quantities and the superconducting energy gap is yet to

be understood. This is similar to the hole-doped cuprates when both high-energy and low-energy gaps are observed [2].

In summary, we measured in-plane tunneling conductance into $\text{Pr}_{2-x}\text{Ce}_x\text{CuO}_4$ films and crystals. A NSTG is seen in the whole doping range studied, $x = 0.11\text{--}0.19$. We studied its doping, temperature, and field dependences. In the underdoped region the NSTG exists above the superconducting transition temperature, T_c . T^* , the temperature at which the NSTG appears, merges into T_c on the overdoped side. Finite pairing amplitude above T_c on the underdoped side is the most plausible explanation for our data.

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- [1] C. Renner *et al.*, Phys. Rev. Lett. **80**, 149 (1998).
 - [2] T. Timusk and B. Statt, Rep. Prog. Phys. **62**, 61 (1999).
 - [3] V.J. Emery and S. A. Kivelson, Nature (London) **374**, 434 (1995).
 - [4] C. M. Varma, Phys. Rev. Lett. **83**, 3538 (1999).
 - [5] S. Kleefisch *et al.*, Phys. Rev. B **63**, 100507(R) (2001).
 - [6] A. Biswas *et al.*, Phys. Rev. B **64**, 104519 (2001).
 - [7] L. Alff *et al.*, Nature (London) **422**, 698 (2003).
 - [8] Y. Dagan *et al.*, Phys. Rev. Lett. **92**, 167001 (2004).
 - [9] G. M. Luke *et al.*, Phys. Rev. B **42**, 7981 (1990).
 - [10] M. Fujita *et al.*, Phys. Rev. Lett. **93**, 147003 (2004).
 - [11] N.P. Armitage *et al.*, Phys. Rev. Lett. **87**, 147003 (2001).
 - [12] A. Zimmers *et al.*, cond-mat/0406204; Y. Onose *et al.*, Phys. Rev. Lett. **87**, 217001 (2001).
 - [13] D. Racah and G. Deutscher, Physica (Amsterdam) **263C**, 218 (1996).
 - [14] H. Yamamoto, M. Naito, and H. Sato, Phys. Rev. B **56**, 2852 (1997).
 - [15] Y. Imry and Z. Ovadyahu, Phys. Rev. Lett. **49**, 841 (1982).
 - [16] P.A. Lee and T.V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985); V. Yu. Butko, J.F. Ditusa, and P.W. Adams, Phys. Rev. Lett. **84**, 1543 (2000).
 - [17] L. Y. Shen, J. Appl. Phys. **40**, 5171 (1969).
 - [18] G.I. Rochlin and P.K. Hansma, Phys. Rev. B **2**, 1460 (1970).
 - [19] J.G. Adler *et al.*, Phys. Rev. B **11**, 2812 (1975).
 - [20] Hamza Balci *et al.*, Phys. Rev. B **68**, 054520 (2003).
 - [21] N. Miyakawa *et al.*, Phys. Rev. Lett. **80**, 157 (1998).
 - [22] Ar. Abanov, A.V. Chubukov, and A.M. Finkel'stein, Europhys. Lett. **54**, 488 (2001).