Distinction between the normal-state gap and superconducting gap of electron-doped cuprates

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We report the study of temperature- and magnetic-field dependent in-plane tunneling conductance on the electron-doped $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ and $Pr_{1-x}LaCe_xCuO_{4-y}$ single crystals. The previously reported normal state gap (NSG) Δ_{ps} and superconducting gap Δ_{sc} were observed simultaneously in superconducting state. Combined with an investigation on the evolution of low-energy spectral weight with temperature, our data present evidence for electron-doped cuprates that Δ_{ps} and Δ_{sc} are most possibly two different energy scales with very different field dependencies, and that superconductivity is condensed from the residual quasiparticle density of states (DOS) of the NSG state.

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I. INTRODUCTION

In the past two decades of extensive study of high-temperature cuprate superconductivity, an anomalous pseudogap has attracted tremendous attention because it is closely related to the state out of which the superconductivity arises.[1–3] Up to now, there is still no consensus on the relationship between the pseudogap and the superconducting gap which has become a crucial issue for understanding the physical origin of the high-temperature superconductivity. For hole-doped cuprates, even the most recent experiments seem to support two different viewpoints, namely, the "two gap" scenario[4–7] and "one gap" (or preformed pairing) scenario.[8,9] Similar confusion was encountered for electron-doped cuprates, in which a normal state gap (NSG) or "small pseudogap" (Δ_{ps}) comparable to the superconducting gap (Δ_{sc}) was noticed several years ago[10,11] and was demonstrated subsequently by various tunneling experiments[12–15] and interlayer transport measurements.[16,17] Alff *et al.*[13] showed that Δ_{ps} vanishes at a doping dependent temperature (T^*) lower than the superconducting critical temperature (T_c) for doping levels close to the optimum. This suggested that the NSG is related to a new order competing with the superconducting one. However, a very different behavior was revealed by a later experiment in which T^* was found to be greater than T_c for the underdoped region while $T^* \approx T_c$ for higher doping levels.[14] This appeared to confirm that the NSG is identical to the superconducting one. This discrepancy, to a large extent, is due to the gradual filling up of the NSG with increasing temperature without any distinct character around T^*. If Δ_{ps} and Δ_{sc} are indeed two different energy scales, they should be distinguished directly in the tunneling spectra with a sufficiently high resolution. However, all the previous tunneling experiments prior to this work cannot distinguish between Δ_{sc} and Δ_{ps},[10,12,13,15] which may be either an evidence for the one gap scenario or just a consequence of the insufficient resolution of these measurements. Thus the relationship between Δ_{sc} and Δ_{ps} is still an open issue for electron-doped cuprates as well as the hole-doped ones.

In this paper, we report a systematic in-plane tunneling study on the electron-doped $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ (NCCO) and $Pr_{1-x}LaCe_xCuO_{4-y}$ (PLCCO) single crystals as a function of field and temperature for various doping levels. We present direct evidence, for the first time, that the superconducting gap Δ_{sc} and the NSG Δ_{ps} are most possibly two different energy scales with distinct field dependencies. According to the analysis of low-energy spectral weight, we can conclude that superconductivity in electron-doped cuprates is condensed from the residual density of states (DOS) of the NSG state.

II. EXPERIMENT

High-quality single crystals of NCCO and PLCCO used in this work were grown by the traveling-solvent floating-zone technique with a subsequent annealing at different temperatures in pure Ar or vacuum.[18] Four superconducting single crystals with various doping levels were studied, including two underdoped PLCCO samples (plcco-un19 and plcco-un23), a nearly optimally doped NCCO sample (ncco-op25) and an overdoped PLCCO sample (plcco-ov17), with $T_c \approx 19$ K, 23 K, 25 K and 17 K, respectively. In order to detect the intrinsic properties of the superconducting planes in the crystals, the in-plane tunneling configuration was designed instead of the *c*-axis tunneling as usually done by scanning tunneling microscopy (STM). For this purpose, the in-plane point-contact junctions were made by carefully driving a Pt/Ir alloy or Au tip toward the crystal along a direction normal to the *c* axis.[19] For the measurements under an external magnetic field, the direction of the applied field is parallel to the *c* axis of the crystals. To obtain high-quality junctions with the least degradation of the sample surface, the crystals were carefully processed by nonaqueous chemical etching before being mounted on the point-contact device.[20] The good reproducibility was demonstrated by the very similar shapes of the spectra measured at various locations on the sample surface.

III. RESULTS AND DISCUSSIONS

Figure 1(a) shows the temperature dependence of the in-plane tunneling spectra measured on the sample of ncco-

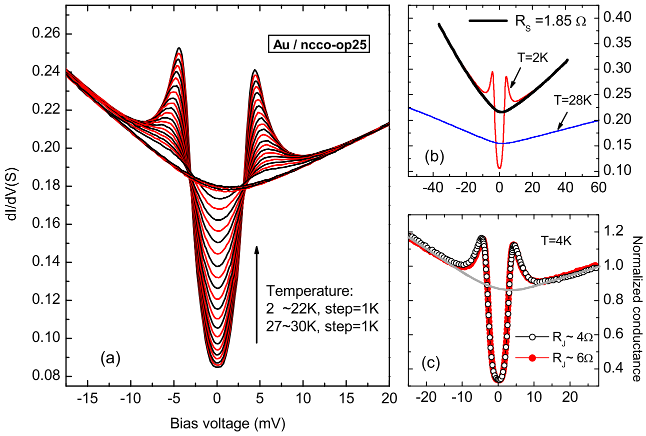

FIG. 1. (Color online) (a) Conductance vs voltage for ncco-op25 measured at various temperatures. (b) Spectra measured in superconducting state ($T=2$ K) and normal state ($T=28$ K), respectively. The thick solid line indicates the spectrum of 28 K by subtracting the normal state resistance of the sample ($R_s=1.85\ \Omega$). (c) Comparison of two spectra measured at the same location whereas with different junction resistances, the solid line indicates the background according to the spectra of above T_c (see text).

op25. Similar results were obtained for all the samples studied (refer to Fig. 4). All the spectra measured above T_c have been transformed by replacing dV/dI and V by $dV/dI-R_s$ and $V-IR_s$, respectively, in order to remove the contribution of the normal state resistance of the sample (R_s) which is involved when the sample becomes normal at $T>T_c$ (or $H>H_{c2}$).[19] As shown in Fig. 1(b), by subtracting $R_s=1.85\ \Omega$ from the raw data of 28 K, only the contribution from the barrier is left which has the same higher-energy background as that of the 2 K spectrum. In Fig. 1(a), data between 23 K and 26 K are not shown because the spectral shape is deformed by the "critical current effect"[21] around superconducting transition. Since the normal state spectra (above T_c) have an almost identical shape except the slight variation at low energy due to thermal excitation, the spectrum above T_c can be regarded approximately as the background of the low-temperature ones. In our experiments, in order to avoid artificial errors, a prudential way has been taken to ensure our measurements in an appropriate regime as elaborated in Refs. 19 and 20. In this regime, as illustrated in Fig. 1(c), the spectra with different junction resistances can be scaled into a universal curve, hence the contribution from the nonsuperconducting impurity phases and the significant interfacial dissipation can be ruled out. Furthermore, the measurements were proved to be highly reproducible everywhere on the sample surface.

Unlike the situation in hole-doped cuprates, normal state of electron-doped cuprates can be accessed much more easily even at extremely low temperature due to their much lower H_{c2}. This provides an uncommon chance to study both superconducting state and normal state of cuprates at very low temperature. Figure 2(a) shows the spectra of $T=2$ K in both superconducting state ($H=0$ T) and normal state ($H=9.5$ T) of ncco-op25. In the normal state one observes a remarkable loss of spectral weight (corresponding to a suppression of DOS) around Fermi energy. Similar results were

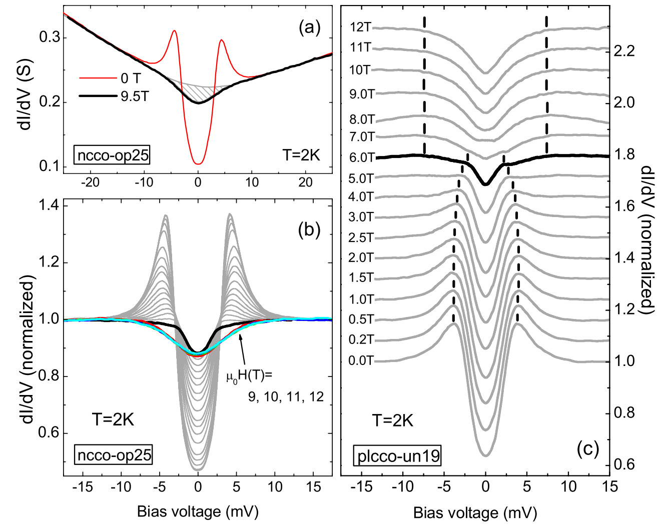

FIG. 2. (Color online) (a) Conductance vs voltage for ncco-op25 in both superconducting state ($H=0$ T) and normal state ($H=9.5$ T). The hatched area indicates the lost low-energy spectral weight in normal state. (b) Field dependence of normalized spectra for ncco-op25. The normal state data with $H=9-12$ T overlap each other. The thin gray lines correspond to the fields from 0 T to 5.5 T, and the thick black line indicates the spectra of $H=6$ T. (c) Field dependence of normalized spectra for plcco-un19. All curves except the lowest one are offset upwards for clarity. Both the superconducting coherence peaks and the NSG edges are indicated by short black bars.

obtained for all the samples studied. This is the first observation of the NSG in NCCO and PLCCO single crystals. Combining all the previous experiments,[10–15] the NSG has been demonstrated in a majority of electron-doped cuprates and seems to be a universal property.

Figure 2(b) shows the field-dependent spectra of ncco-op25 normalized by a high-temperature curve (above T_c) with a transformation eliminating the crystal's resistance as mentioned above. At low fields, the coherence peaks are smeared and move slightly toward higher energy due to field-induced pair breaking. By further increasing field, the energy space between the peaks shrinks continuously until the peak feature can no longer be distinguished. The same phenomenon was observed for all doping levels studied. As another example, Fig. 2(c) shows the result of an underdoped sample (plcco-un19); the distance between the coherence peaks drops rapidly to zero at H_{c2}. These results indicate that the superconducting gap Δ_{sc} decreases monotonically with increasing field and vanishes at H_{c2}. This is one of the main findings in this work, which was not detected in previous measurements most possibly due to the stronger broadening of the measured spectra.[13,14] Recently, we have found that Δ_{sc} decays with increasing field in an optimally doped PLCCO sample using theoretical fitting,[22] while in this work, we present direct evidence from the data themselves for various doping levels.

As shown in Figs. 2(b) and 2(c), for the fields higher than H_{c2}, a NSG shows up. In contrary to the behavior of Δ_{sc}, the NSG Δ_{ps} is insensitive to the applied field as demonstrated by the negligible difference between the spectra of various fields from H_{c2} to the highest field of the instrument. This is also consistent with the recent high magnetic-field measurements on another system of $Pr_{2-x}Ce_xCuO_{4-y}$.[15] Around H_{c2}, both the suppressed coherence peaks and the NSG edges can be distinguished simultaneously for the first time [refer to the data of $\mu_0 H=6$ T in both Figs. 2(b) and 2(c)]. In Fig. 2(c), the evolution of the coherence peaks and the NSG edges are indicated explicitly by the short black bars. Here, we defined the NSG edge as the energy at which the conductance decays to 95% of the "NSG depth"—the distance from the bottom of the spectra to the background conductance. However, this may not be the most reasonable definition since the underlying mechanism of the NSG is unknown. Furthermore, to some extent, the NSG edge also depends on the normalization procedure (for example, choosing different backgrounds according to the high-temperature data or a particular function, respectively). Thus we used more conservative criteria to estimate the energy scale of Δ_{ps} and found that, even if a criterion of 70% of NSG depth is taken, the obtained Δ_{ps} is still larger than Δ_{sc}. For the sample of ncco-op25, the lower limit of the criterion for $\Delta_{ps} > \Delta_{sc}$ is about 60%, which corresponds to a spectral weight loss of about 62% (compared to the total spectral weight lost related to the NSG). In other words, only 38% of the total spectral weight is lost between the determined energy scale of Δ_{ps}. Obviously, in this case, Δ_{ps} may be underestimated though it is still bigger than Δ_{sc}. Therefore, it is most possible that the real Δ_{ps} is larger than Δ_{sc}, as can be seen more clearly at higher fields. In the prepairing (or one gap) scenario, Δ_{ps} is a prepairing gap and phase coherence is established at H_{c2}, thus Δ_{sc} is equal to Δ_{ps} in values and Δ_{sc} will not decrease with increasing field because Δ_{ps} is insensitive to field. In this sense, although the height of coherence peak will be suppressed remarkably by field, the peak position is not necessary to decay to zero rapidly (the two-band superconductor MgB_2 is a good example, when a small magnetic field is applied, the coherence peaks of π-band gap is suppressed remarkably while its gap value is almost unchanged). Obviously, this is not the case of our measurements in which Δ_{sc} decays continuously to zero with increasing field. Although the energy scale of the NSG indicated in Fig. 2(c) is not a rigorous value, it is most possible that Δ_{sc} and Δ_{ps} are two different energy scales with distinct field dependencies. It should also be mentioned that the coexistence of Δ_{sc} and Δ_{ps} can be distinguished only around H_{c2}, thus the contribution from the vortex cores in mixed state should be considered. It is known that point-contact junctions often probe both the superconducting regions and vortex cores. Therefore, simply according to the field-dependent data, it is not conclusive whether Δ_{sc} coexists with Δ_{ps} or, alternatively, Δ_{ps} only appears when Δ_{sc} is suppressed in a higher field or in a vortex core.

In order to get further insight into the relationship between Δ_{sc} and Δ_{ps}, we apply a new analysis of our data based on the integration of the spectral weight within the energy well above the gap edges. It is well known that for an ideal tunneling spectrum of a superconductor, by setting the background conductance to zero, the integrated spectral weight (I_W) should be zero. This is desired by the conservation of DOS according to the Landau Fermi-liquid theory. For a point-contact junction, the barrier height at the interface is

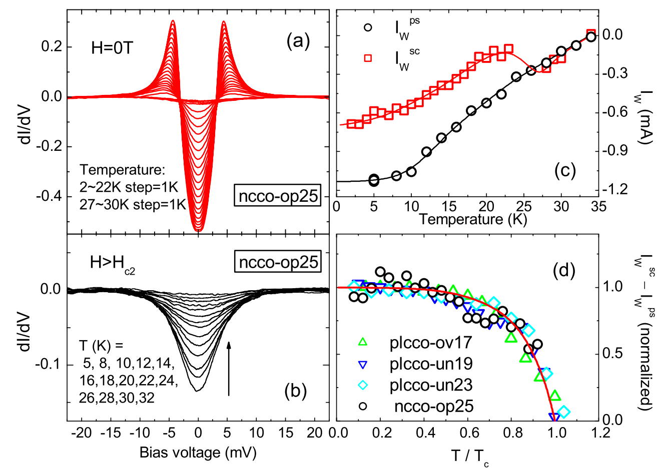

FIG. 3. (Color online) Temperature dependence of normalized spectra for (a) $H=0$ T and (b) $H>H_{c2}$. (c) Temperature dependence of the integrated spectral weight for the spectra shown in (a) and (b), respectively. (d) Difference in the integrated spectral weight between superconducting state and NSG state. The solid line is the theoretical calculation according to the BTK theory.

finite except for an infinite barrier in the ideal tunneling regime. Accordingly, the contribution of the Cooper pairs and/or superfluid is involved in the interface current by the process of Andreev reflection. In this case, I_W should be a positive finite value. With increasing temperature, the contribution of superfluid and hence I_W decays in a particular way according to the form of the pairing potential, and finally drops to zero at T_c. This has been demonstrated extensively by point-contact measurements on various superconductors such as Nb, MgB_2, and Re_3W.[23–25] As shown in Fig. 2(a), the NSG state and superconducting state have the same background, inspiring us to study the evolution of I_W in both states in order to understand the relationship between them.

In Figs. 3(a) and 3(b) we present the temperature-dependent spectra in both superconducting state and NSG state. The spectra are normalized by the high-temperature one ($T=34$ K) with a transformation eliminating the crystal's resistance, then the high-bias conductance is set to zero. It should be pointed out that choosing the data of $T=34$ K as background does not mean that it is the Δ_{ps} closing temperature. Determining the Δ_{ps} closing temperature requires some particular measurements and analysis since the NSG has almost been filled at higher temperature due to thermal effect. This is not the subject of this paper. From Figs. 3(a) and 3(b), it is noted that all spectra merge into each other above a bias voltage around 15 mV. Thus we do the integration of spectral weight between ± 20 mV and the results are presented in Fig. 3(c). It is found that the integrated spectral weight in the NSG state (I_W^{ps}) is a finite minus value reflecting the suppression of DOS around Fermi energy as mentioned above. With increasing temperature, I_W^{ps} increases monotonically toward zero, indicating a gradual recovery of the suppressed DOS. This is consistent with the previous reports on $Pr_{2-x}Ce_xCuO_{4-y}$.[13,14] A surprising finding in this work is the novel evolution of the integrated spectral weight in superconducting state (I_W^{sc}). As expected by a finite-barrier junction mentioned above, I_W^{sc} shown in Fig. 3(c) indeed drops rapidly

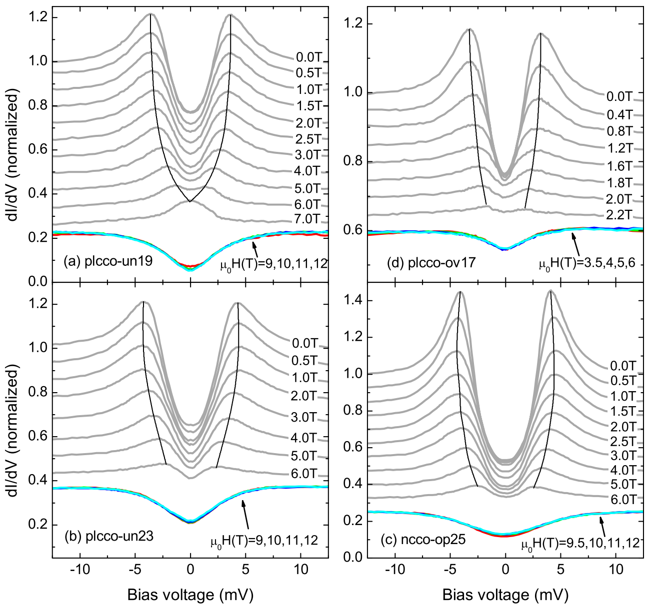

FIG. 4. (Color online) Evolution of spectra at $T=2$ K with increasing field for various doping levels: (a) plcco-un19, (b) plcco-un23, (c) ncco-op25, and (d) plcco-ov17. For each dope, all the data of $H<H_{c2}$ are normalized by the one with the highest field and offset relatively in order to demonstrate the evolution of the coherence peaks as indicated by those two symmetric thin lines.

around T_c and merges into the trace of I_W^{ps} above T_c. Accordingly, a continuous reduction in I_W^{sc} with increasing temperature is also expected. On the contrary, I_W^{sc} increases monotonically with increasing temperature in a wide temperature range below T_c. The most reasonable explanation for this anomaly is that the NSG state is the background of the superconducting state. This scenario can be further supported by the analysis presented in Fig. 3(d), where I_W^{sc}-I_W^{ps} are calculated and plotted in a reduced scale. It is very clear that the results for all studied doping levels are similar and in good agreement with the calculation of the Blonder-Tinkham-Klapwijk (BTK) model[26] for a finite barrier (as denoted by the solid line). This presents strong evidence that superconductivity in electron-doped cuprates is condensed from the residual DOS of the NSG state, as addressed by specific-heat measurements in hole-doped side.[27] The coexistence of the superconducting gap and NSG in electron-doped cuprates observed here is similar to the recent observations in hole-doped cuprates both by angle-resolved photoemission spectroscopy (ARPES) (Ref. 6) and by STM.[7]

Since superconductivity is contributed from the residual DOS of the NSG state, the more appropriate background for the spectra of superconducting state ($H<H_{c2}$) should be that of the NSG state (i.e., the spectra measured at the same temperature while with $H>H_{c2}$) instead of the higher-temperature one ($T>T_c$). Comparing the data of 6 T of ncco-op25 shown in Fig. 2(b) with that in Fig. 4(c), one can see that the coherence peaks in the former only present themselves as inflexions due to the underlying background of the NSG state, while the peaks become more prominent in the latter by subtracting this background. Similar results were obtained for other doping levels. In this case, as presented in Figs. 4(a)–4(d), it can be seen more clearly that the superconducting gap decreases monotonically with increasing field (the gap value was considered roughly as the positions of the coherence peaks and hence was overestimated due to smearing effect on the measured spectra especially for high fields, therefore, the real gap value should decrease more rapidly than the estimations presented here). On the contrary, as shown in the bottom of each panel of Fig. 4, all spectra measured above H_{c2} are almost identical up to the highest field, indicating very weak field dependence of the NSG. The data for the fields close to H_{c2} are not shown because of the critical current effect mentioned above. In Fig. 5, we sum up the field dependence of Δ_{sc} for all doping levels. It is interesting to find that all data can be scaled into one other very well simply by normalizing the data to their respective upper critical field H_{c2} and maximum gap value $\Delta_{sc}(H=0)$. Furthermore, Fig. 5 explicitly demonstrates that Δ_{sc} is suppressed continuously with increasing field and finally vanishes at H_{c2} for all doping levels studied here, indicating the remarkable field-induced pair breaking and/or suppression of pairing strength. This is consistent with the result of other electron-doped cuprates from magneto-Raman spectroscopy in which the amplitude of Δ_{sc} is strongly suppressed in a finite magnetic field.[28] This behavior is obviously in contrast to that of Δ_{ps} which is insensitive to the field we have applied.

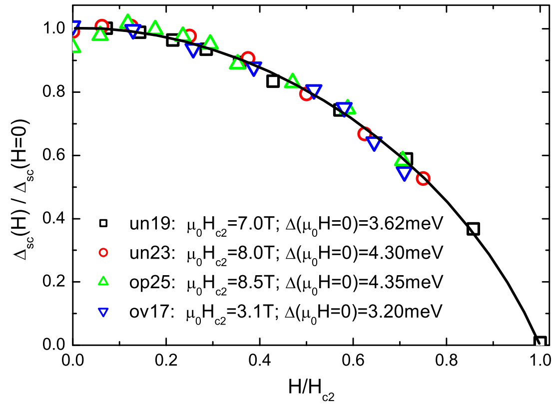

FIG. 5. (Color online) Field dependence of the magnitude of Δ_{sc} which was estimated from the peak positions of the spectra shown in Fig. 4. The solid line is a guide to the eye.

IV. SUMMARY

In summary, we have studied in-plane tunneling spectra of electron-doped cuprates as a function of both temperature and magnetic field. A superconducting gap and a NSG were observed for all studied samples spanning both underdoped and overdoped regimes. Our data present possible evidence that the two gaps have different energy scales with distinct field dependencies while they coexist with each other. By an analysis of low-energy spectral weight, we can conclude that superconductivity in electron-doped cuprates is formed upon the background of the NSG state.

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