

## Semiconducting gap of $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ revealed by break-junction tunnelling spectroscopy

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**Abstract.** The quasiparticle energy spectrum of the semiconducting  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$  was probed by a break-junction technique. The tunnelling conductance exhibits well-defined gap-edge peaks at 4 K with peak-to-peak bias separations 2-2.8 eV, showing the V-shaped gap feature. The gap value  $2\Delta(4\text{K}) = 1-1.4$  eV is inferred in view of the very break-junction nature. As temperature,  $T$ , increases, the gap gradually disappears to merge with the background at  $T \sim 200$  K. Possible origins of the gap are discussed on the basis of the break-junction and complementary data obtained using scanning-tunnelling spectroscopy.

### 1. Introduction

The mechanism of superconductivity in high- $T_c$  cuprates is still in question, although its unequivocal identification was declared a number of times. In particular, it is not known whether the symmetry of order parameter is the same for all studied compounds [1]. Even the main pillar of the Bardeen-Cooper-Schrieffer (BCS) theory, namely the Cooper pairing, has been refuted by the concept of the real-space bipolaron pairs [2]. Further, we do not know in detail the physics of the normal state for these strongly correlated materials and the role of the competitive phenomena, such as charge- and spin-density waves (CDWs and SDWs), charge-carrier localization or a nano-scale disorder of electronic properties in the superconducting state [3,4]. The latter features are appropriate to nominally electron-doped sub-family of high- $T_c$  oxides.

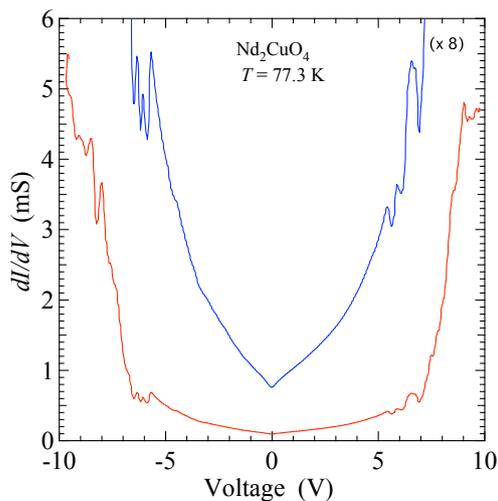
Hence, we have carried out studies of the Ce doped and undoped  $\text{NdCuO}_4$  using both break-junction and scanning-tunnelling spectroscopy (STS) techniques to investigate the electron spectrum of the semiconducting compositions. We have revealed anomalously large normal-state energy gaps, which are similar to their counterparts in the semiconducting  $\text{La}_{1.98}\text{Sr}_{0.02}\text{CuO}_4$  [9].

### 2. Experimental

Polycrystalline semiconducting samples of  $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$  ( $x = 0, 0.15$ ) were studied. They were obtained by annealing in air at  $1000^\circ\text{C}$  for 30 hours and slowly cooled down to room temperature,  $T$ . The superconducting sample used in STS studies was obtained by quenching at  $T = 77\text{ K}$  after annealing in nitrogen atmosphere. The typical linear grain size was  $\sim 10\ \mu\text{m}$ . In break-junction measurements with four-probe geometry, samples were cracked *in situ* below  $77\text{ K}$  to obtain a fresh interface [10]. STS measurements were carried out using Omicron low- $T$  ultra-high-vacuum ( $10^{-8}\text{ Pa}$ ) scanning tunnelling microscope system (LT-UHV-STM) with a field-cleaned Pr-Ir tip. The sample was cleaved there at  $77\text{ K}$  and then transferred to the STM chamber at  $\sim 5\text{ K}$ .

### 3. Results and discussion

Figure 1 shows the break-junction tunnelling conductance for  $\text{Nd}_2\text{CuO}_4$ , which is a semiconducting parent compound for the superconducting  $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ . The tunnelling conductance  $G(V) \equiv dI/dV$  has a V-like shape and is featureless for bias voltages  $|V| < 5\text{ V}$ , where  $I$  stands for the quasiparticle current. For larger  $|V|$  fine structure appears and  $G(V)$  increases steeply. The origin of the structure is not clear. It might be due to Joule heating and/or migrations of surface atoms at the junction interface due to large enough applied biases. Within the range of  $\pm 10\text{ V}$ , no peaks were observed that might be identified with the increase of the electronic density of states (DOS) at gap edges as a result of a DOS depletion inside the semiconducting gap [9].

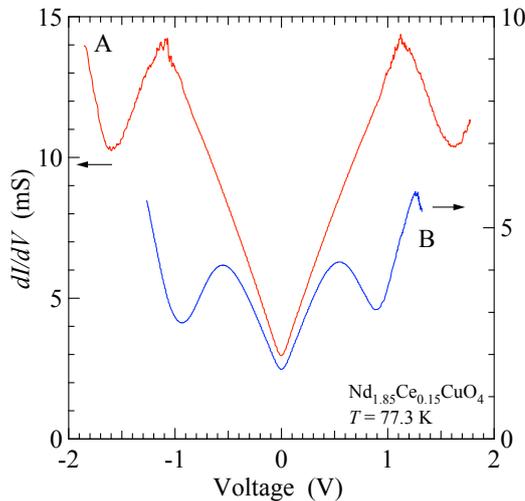


**Figure 1.** Break-junction tunnelling conductance  $G(V) \equiv dI/dV$  for  $\text{Nd}_2\text{CuO}_4$  at temperature  $T = 77.3\text{ K}$ . Here  $I$  and  $V$  are the quasiparticle current and bias voltage, respectively.

When the Ce-doped compositions were synthesized, their room- $T$  resistivity,  $\rho$ , drastically decreased by a factor of  $\sim 10^4$  as compared to that for  $x = 0$ , but  $\rho(T)$  still exhibited a semiconducting behavior because the compound was air-annealed. To examine the doping-driven changes of electronic properties, we have measured  $G(V)$  of the doped samples. The results are displayed in Fig. 2. The well-defined gap-edge peaks were observed, which is what one expects for compounds with a semiconducting behavior of the collective origin [11]. At the same time, the spectacular V-shaped behavior of  $G(V)$  inside the gap region indicates a Coulomb-induced charge-carrier localization of low-energy states [3].

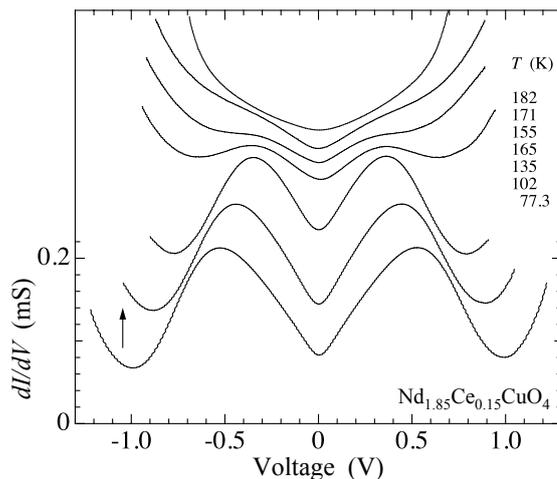
The residual zero-bias conductance  $G(0)$  is down to  $\sim 20\text{-}30\%$  of the peak conductance. It should be noted that in the break-junction experiments one usually observes two kinds of spectra possessing twice different gap-edge energies. These features can be attributed to either anticipated semiconductor-insulator-semiconductor (Sm-I-Sm) junctions (A) or the accidentally formed semiconductor-insulator-normal metal (Sm-I-N) ones (B) in the break-junction set-up. Hence, the peak-to-peak bias distances are either  $V_{p-p} = 4\Delta/e$  (A) or  $2\Delta/e$  (B), respectively, where  $2\Delta$  is the actual dielectric gap (e.g., between the valence and conducting band in the one-particle scheme,  $e > 0$  is the elementary charge). Bearing in mind those possibilities, the curves A and B in Fig. 2 should be considered as appropriate to an Sm-I-

Sm and an Sm-I-N junction, respectively. Therefore,  $2\Delta = 1-1.4\text{eV}$ . This value is approximately twice as large as the pseudogap found in previous optical measurements [7].



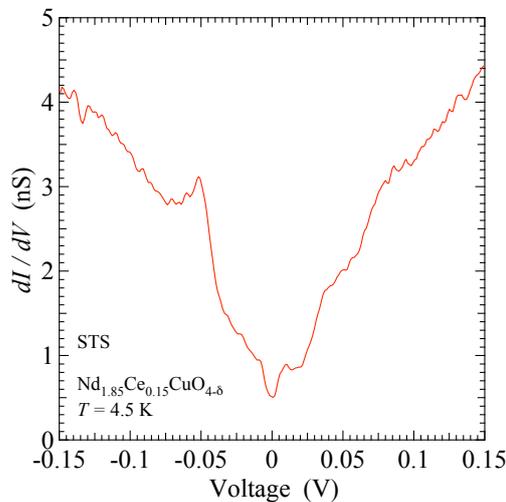
**Figure 2.**  $G(V)$  for  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ . Curves A and B are measured for different junctions.

Figure 3 shows  $G(V)$  for various  $T$  for a break junction showing  $V_{p-p} = 1\text{ V} (= 2\Delta/e)$  at the lowest  $T = 77.3\text{ K}$ . With increasing  $T$ , the  $V_{p-p}$  gradually decreases, the gap structure itself losing its specific form until it completely merges with the background at  $T^* \sim 180 - 200\text{ K}$ . This value is somewhat smaller than the resistively and magnetically determined cusp temperature  $230 \sim 240\text{ K}$  found for *superconducting* samples of the same composition synthesized in the  $\text{N}_2$  flow [12]. The weakly revealed point  $T^*$  most probably indicates the loss of degeneracy by a minority hole band [12] in the two-band electronic structure of  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ . The coexistence between electron- and hole-like charge carriers has been recently noticed [3] for a highly disordered oxide  $\text{SrPbO}_3$ , being on the verge of the metal-insulator transition.



**Figure 3.**  $G(V)$  for  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$  at various  $T$ .

To gain further insight into the intriguing behavior of  $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4$ , we have investigated superconducting  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$  by STS as shown in Fig. 4. We can see the conventional superconducting gap structure at  $V \sim \pm 0.01\text{ V}$ . We emphasize that  $G(V)$  of Fig. 4 demonstrates not only well-defined superconducting peaks, but also conspicuous features at about  $0.05\text{ V}$  and  $0.1\text{ V}$ . These values are consistent with recent STS measurements [8], in which they were attributed to the antiferromagnetic gapping locally coexisting with superconductivity.



**Figure 4.**  $G(V)$  for  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-\delta}$  obtained from scanning tunnelling spectroscopy (STS) measurements at  $T = 4.5$  K.

#### 4. Conclusions

The present break-junction measurements revealed an unexpectedly huge semiconductive gapping, which occurs on the scale  $\sim 1$ -2 V. The very voltage range and the V-like behavior of  $G(V)$  points to the fact that the gap might be severely influenced by the Coulomb localization of charge carriers bearing in mind the intrinsic oxygen ion disorder. To further test the validity of the results, we have carried out reference measurements of the superconducting sample by STS and found gap structures at  $\sim 10$  and  $\sim 50$  meV, which can be attributed to the well-known superconducting gap and, presumably, a gap induced by many-body correlations, respectively.

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