

# Physical properties of Nd<sub>1.85</sub>Ce<sub>0.15</sub>CuO<sub>4-y</sub> at high pressure and with different oxygen contents

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## PHYSICAL PROPERTIES OF $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$ AT HIGH PRESSURE AND WITH DIFFERENT OXYGEN CONTENTS

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We have measured initial susceptibility on  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$  with  $y$  varying from 0.02 to 0.32.  $T_c$  was quite insensitive to the different oxygen contents. The pressure dependence  $dT_c/dp$  for a compound with  $y = 0.03$  was determined to be  $-0.04$  K/kbar.

### Introduction

Since the discovery of electron-doped superconductivity in high- $T_c$  cuprates [1], each model on high- $T_c$  superconductivity has had to suffer a checking whether it is symmetric upon doping of either holes or electrons. A single-band model can be thought of to accomplish this symmetry. The theories based on holes with spin  $S = 1/2$  fail to explain superconductivity with spin-holes on the  $\text{Cu}^{1+}$  ions. However, from the beginning Wachter and Degiorgi [2] and de Jongh [3] have proposed a polaronic model in which they postulated a singlet state of the  $\text{Cu}^{3+}$  ion. The  $S = 0$  state of  $\text{Cu}^{3+}$  had been an issue but now from the  $\text{NdCeCuO}$ -compounds it turns out unambiguously that it is a spin-hole responsible for superconductivity.

### Experiment, Results

We have prepared the  $\text{NdCeCuO}$  compound by mixing appropriate amounts of  $\text{CeO}_2$ ,  $\text{Nd}_2\text{O}_3$  and  $\text{CuO}$  to achieve a composition of  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_4$ . The mixed oxides were calcined in air at  $950^\circ\text{C}$  for 10 hours, pressed into pellets and sintered in air at  $1150^\circ\text{C}$  for 12 hours. The samples were quenched in air to room temperature. The as prepared material was not superconducting. Then the samples were placed into a Perkin-Elmer thermo-analyzer and quantitative amounts of oxygen were removed by heating up to  $1140^\circ\text{C}$  in an atmosphere of pure argon gas for different times. When the samples had reached the desired oxygen content they were quenched with a cooling rate of  $10^\circ\text{C}/\text{min}$ . Beginning from an oxygen deficiency of  $y = 0.02$  per formula unit superconductivity was observed with  $T_c$  near 23 K. Up to  $y = 0.22$  the samples were single phase with a  $T_c$  not changing appreciably. Typical curves of the ac-susceptibility are shown in Fig. 1. The sample with  $y = 0.32$  was investigated by X-ray diffraction to have two phases. Probably one of them (nonsuperconducting) is responsible for the magnetic response at 8 K. The lattice parameters of a specific superconducting compound with  $y = 0.04$  were  $a = 3.947(\pm 0.001)\text{\AA}$  and  $c = 12.08(\pm 0.002)\text{\AA}$ .

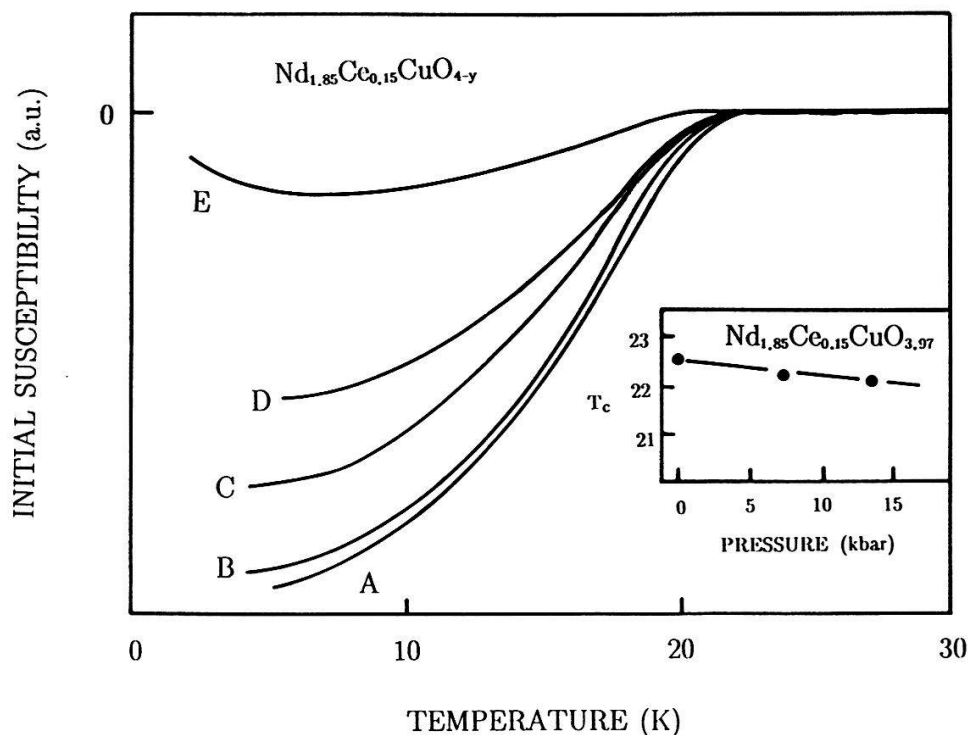
Further we have measured the pressure dependence of  $T_c$  up to 15 kbar as described in a previous paper [4]. There was only a weak pressure dependence of  $dT_c/dp = -0.04$  K/kbar (see inset of Fig. 1).

### Discussion

It is amazing that the variation of the oxygen content does not influence the transition temperature. A too large loss of oxygen ( $y > 0.22$ ) results in a transformation to a red-brown phase. We believe that the outgoing oxygen comes from the  $(\text{Nd,Ce})\text{O}$ -planes between the superconducting  $\text{CuO}_2$  sheets. As Ce can exist in two valence states the charge balance can be achieved by the transition  $\text{Ce}^{4+} \rightarrow \text{Ce}^{3+}$ . Thus, the  $\text{CuO}_2$  plane is stable and even more two-dimensional than in other superconducting cuprates.

Let us look at the pressure dependence. Assuming a band model, pressure causes a broadening of the bandwidth. However, all cuprates have more or less the same superconducting  $\text{CuO}_2$ -planes and thus a similar band structure. Therefore, upon applying pressure, they should behave in a similar way. But the pressure dependence  $dT_c/dp$  of the superconducting cuprates differs within the large range of  $-0.05$  to  $0.6$  K/kbar which is not very consistent with the band-picture.

We suggest a model of exchange coupled bipolarons as described by Wachter and Degiorgi [2]. In this model the concentration of polarons ( i.e. the concentration of  $\text{Cu}^{3+}$  resp.  $\text{Cu}^{1+}$  ) is crucial. At high pressure the apical oxygen ion is shifted towards the  $\text{CuO}_2$ -plane and can serve as a doping reservoir. The more apical O-ions available the more effective will be an applied pressure to adjust the optimal concentration. From this point of view it seems reasonable that the  $(\text{La,Ba})_2\text{CuO}_4$  family with two apical O-atoms and the  $\text{YBa}_2\text{Cu}_4\text{O}_8$  and  $\text{YBa}_2\text{Cu}_{3.5}\text{O}_7$  [5] compounds with two chains resp. alternatively two and one chain as intercalation exhibit a strong pressure dependence. The  $\text{YBa}_2\text{Cu}_3\text{O}_7$  composition with only one apical oxygen is to show a lower  $dT_c/dp$  as is really the fact. For the  $\text{NdCeCuO}$ -compound with no apical oxygen above  $\text{Cu}(2)$ , it is not surprising that we even have detected a negativ  $dT_c/dp$ .



**FIG. 1** Initial susceptibility of  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$  with the different oxygen contents  $y$  : A: 0.03, B: 0.07, C: 0.02, D: 0.21, E: 0.32. The inset shows the pressure dependence of  $T_c$  of  $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{3.97}$ .

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- [4] B. Bucher, J. Karpinski, E. Kaldis and P. Wachter, *Physica C* **157**, 478 (1989)
- [5] The pressure dependence of  $\text{YBa}_2\text{Cu}_{3.5}\text{O}_7$  was determined to be  $dT_c/dp = 0.45$  K/kbar (to be published elsewhere)