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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 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 Cu^{3+} ion. The $S = 0$ state of Cu^{3+} had been an issue but now from the NdCeCuO -compounds it turns out unambiguously that it is a spin-hole responsible for superconductivity.

Experiment, Results

We have prepared the NdCeCuO compound by mixing appropriate amounts of CeO_2 , Nd_2O_3 and 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°C for 10 hours, pressed into pellets and sintered in air at 1150°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°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 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 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 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 Cu^{3+} resp. Cu^{1+}) is crucial. At high pressure the apical oxygen ion is shifted towards the 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 NdCeCuO -compound with no apical oxygen above $\text{Cu}(2)$, it is not surprising that we even have detected a negativ dT_c/dp .

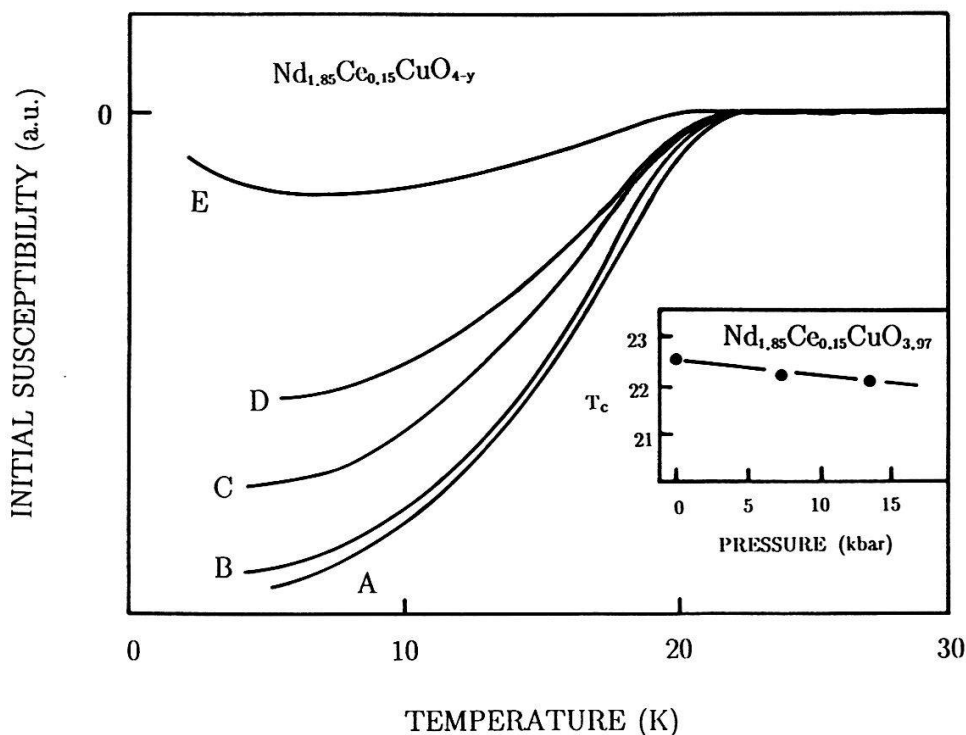


FIG.1 Initial susceptibility of $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{4-y}$ with the different oxygen contents y : A: 0.03, B: 0.07, C: 0.02, D: 0.21, E: 0.32. The inset shows the pressure dependence of T_c of $\text{Nd}_{1.85}\text{Ce}_{0.15}\text{CuO}_{3.97}$.

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- [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)