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PHYSICAL PROPERTIES OF $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ AT HIGH PRESSURE AND WITH DIFFERENT OXYGEN CONTENTS

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We have measured initial susceptibility on $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ with y varing 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 anambigiously 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 composititon of $Nd_{1.85}Ce_{0.15}CuO_4$. 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 sampels had reached the desired oxygen content they were quenched with a cooling rate of 10° C/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(± 0.001)Å and c = 12.08(± 0.002)Å.

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 temperatur. A too large loos of oxygen (y > 0.22) results in a transformation to a red–brown phase. We believe that the outgoing oxygen comes from the (Nd,Ce)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 $Ce^{4+} \rightarrow Ce^{3+}$. 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 $(La,Ba)_2CuO_4$ family with two apical O–atoms and the $YBa_2Cu_4O_8$ and $YBa_2Cu_{3.5}O_7$ [5] compounds with two chains resp. alternativly two and one chain as intercalation exhibit a strong pressure dependence. The $YBa_2Cu_3O_7$ 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 Cu(2), it is not surprising that we even have detected a negativ dT_c/dp.

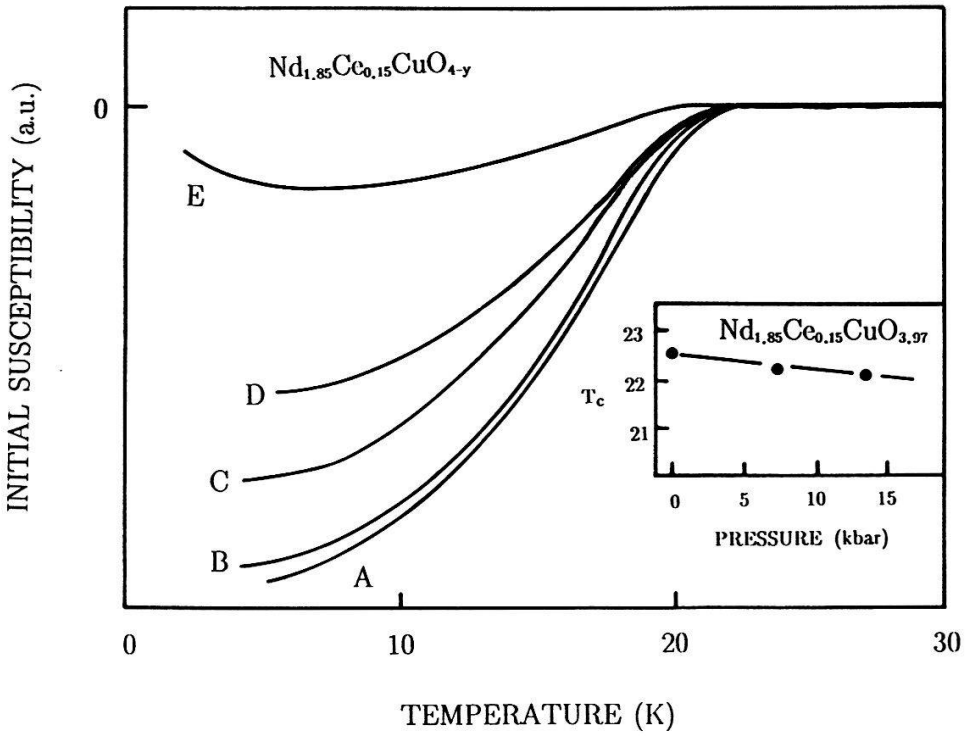

FIG.1 Initial susceptibility of $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ with the different oxygen contents y : A: 0.03, B: 0.07, C: 0.02, D: 0.21, E: 0.32.
The inset shows the pressure dependence of T_c of $Nd_{1.85}Ce_{0.15}CuO_{3.97}$.

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