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Effect of pressure on spin-gap behavior in Pr-doped and oxygen-deficient $YBa_2Cu_3O_7$ thin films

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The influence of an external pressure on the behavior of the spin-gap temperature observed in the dc resistivity is measured on $Y_{1-x}Pr_xBa_2Cu_3O_7$ films with $x=0.2$, 0.3, and 0.4 and an oxygen-deficient $YBa_xCu_3O_y$ film with $y=6.6$. It is found that the spin-gap temperature shifts with pressure in a sense opposite to the shift of the critical temperature, consistent with the resonating valence bond mean-field phase diagram. The modified carrier density induced by the pressure is the major effect, while no significant effect of enhanced interplane-coupling of the Cu^{2+} spins due to a decrease in the distance between the CuO_2 planes is observed. [S0163-1829(96)05930-9]

In many experiments on high-T_c superconductors a strong pressure dependence of the critical temperature (T_c) is observed. It is generally believed that the applied pressure causes extra holes to be pushed into the CuO_2 planes. This is experimentally confirmed by, e.g., Furrer *et al.*[1] with inelastic neutron-scattering measurements. These extra holes lead to an increase of T_c with pressure. In the case of YBaCuO, the charge transfer from the CuO chains to the CuO_2 planes is influenced by a rearrangement of the ions in the chains resulting from the modified pressure.

For oxygen-depleted films ($YBa_2Cu_3O_{7-d}$) the effect of increasing pressure on the critical temperature is always positive ($dT_c/dP>0$) up to a certain pressure.[2] The influence of pressure on the $Y_{1-x}Pr_xBa_2Cu_3O_7$ compound is however more complicated, i.e., the measured dT_c/dP value is positive or negative dependent on the Pr concentration:[3,4] a positive dT_c/dP is observed for small x, but becomes negative when $x>0.2$. This doping dependent effect can be understood as resulting from the interplay between charge transfer and enhanced Pr $4f$/O $2p$ hybridization leading to hole localization.[5] The charge-transfer dominates at small x, explaining the enhancement of T_c, whereas the hole localization is the main effect at higher Pr concentrations, resulting in a decrease of T_c.

In this paper we study the effect of pressure on the so-called spin-gap behavior, observed in transport[6] and magnetic[7] measurements. The first question is whether we can see a variation of the (transport) spin-gap temperature with pressure, similar to the effect we observe when we

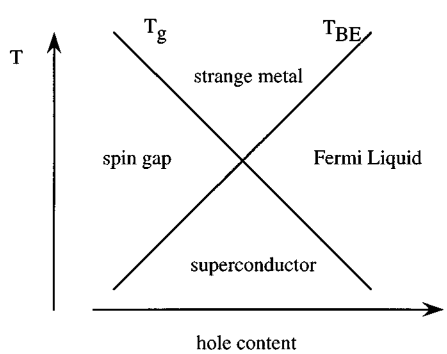

FIG. 1. Phase diagram of the mean-field RVB theory.

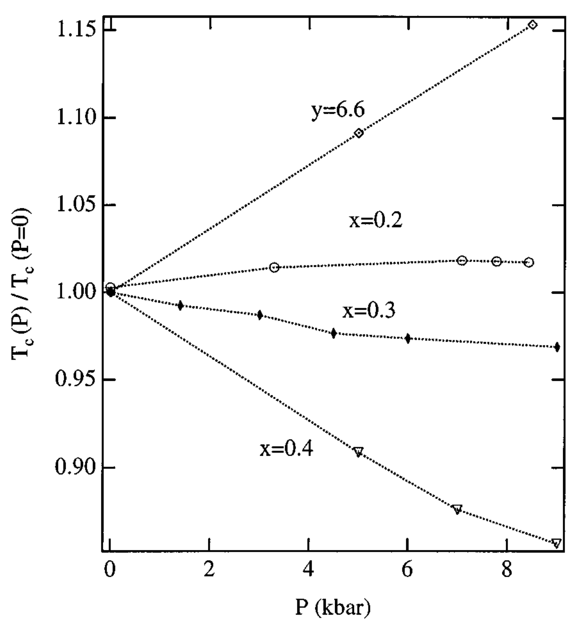

FIG. 2. Variation of the reduced critical temperature $[T_c(P)/T_c(P=0)]$ with pressure for the Pr-doped and the oxygen-deficient samples.

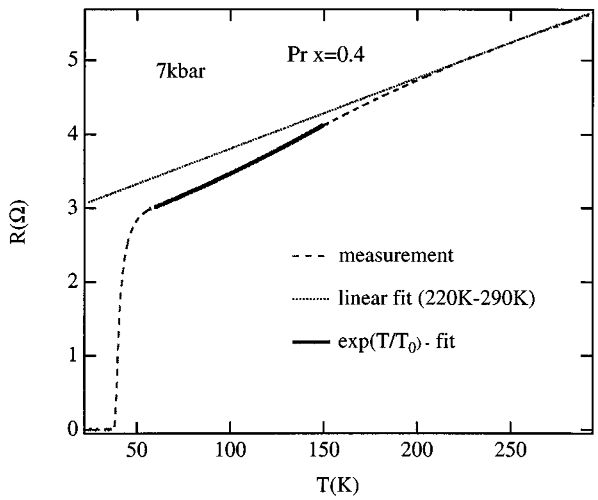

FIG. 3. Fit through the resistance data using an exponential and a linear curve for temperatures below and above the spin-gap temperature, respectively.

TABLE I. Critical temperatures and T_0 values deduced from the exponential fit for the $x=0.3$ and $x=0.4$ Pr-doped YBaCuO layers.

Pr doping	P (kbars)	T_c (K)	T_0 (K)
$x=0.3$	0	59.2±0.2	186.9±1.6
	1.4±0.3	58.8±0.2	192.4±0.7
	3.0±0.3	58.5±0.2	194.1±0.7
	4.5±0.3	57.8±0.2	197.1±0.7
	6.0±0.3	57.7±0.2	198.9±0.7
	9.0±0.3	57.4±0.2	204.0±0.7
$x=0.4$	0	43.1±0.2	248±5.2
	5.0±0.3	39.2±0.2	271.3±0.5
	7.0±0.3	37.8±0.2	282.8±0.6
	9.0±0.3	36.9±0.2	286.1±0.5

change the number of carriers by varying the Pr content or removing oxygen. In that case we expect the spin-gap temperature to shift in a sense opposite to the shift of the critical temperature with pressure, following the phase diagram of the mean-field resonating valence bond (RVB) theory[8] shown in Fig. 1.

Secondly, decreasing the distance between the CuO_2 planes, we can check the influence of interplane spin coupling, which is thought to be crucial for the spin-gap formation.[9] A strongly enhanced coupling between the planes would lead to an increase of the spin-gap temperature with pressure for all doping concentrations, independent of the shift of T_c.

High-pressure resistivity measurements are performed in a liquid clamp cell from the High Pressure Institute, Warsaw, Poland (0–11 kbars). The pressure is measured *in situ* by means of a calibrated InSb pressure gauge. The pressure is applied at room temperature using a hydraulic press. The pressure is clamped into the cell by means of a set screw and the cell is cooled to low temperatures for the measurements.

The $Y_{1-x}Pr_xBa_2Cu_3O_7$ films are fabricated with laser ablation on $LaAlO_3$ substrates with Pr concentrations $0.2 \leqslant x \leqslant 0.4$. An oxygen-deficient $YBa_2Cu_3O_y$ sample with $y=6.6$ is prepared performing an anneal and subsequent cooling in a controlled oxygen pressure, following a constant oxygen line in the oxygen pressure-temperature phase diagram.[10] The films are covered with a polyimide layer in order to prevent degradation of the YBaCuO in the fuel-alcohol mixture with which the pressure cell is filled. The polyimide is opened at four gold-covered contact points. Four leads are attached with silverpaste to these gold contacts.

The shift of the critical temperature with pressure for the different samples is shown in Fig. 2. For the different Pr concentrations, we find a $T_c(P)$ dependence which is similar to the results reported by Neumann and Kretschmer.[3] The pressures mentioned are measured at 4 K.

To determine the spin-gap temperature, we apply the same procedure we used to describe the high-frequency conductivity.[11] In this reference it is shown that the real part of the conductivity below the spin-gap temperature can be

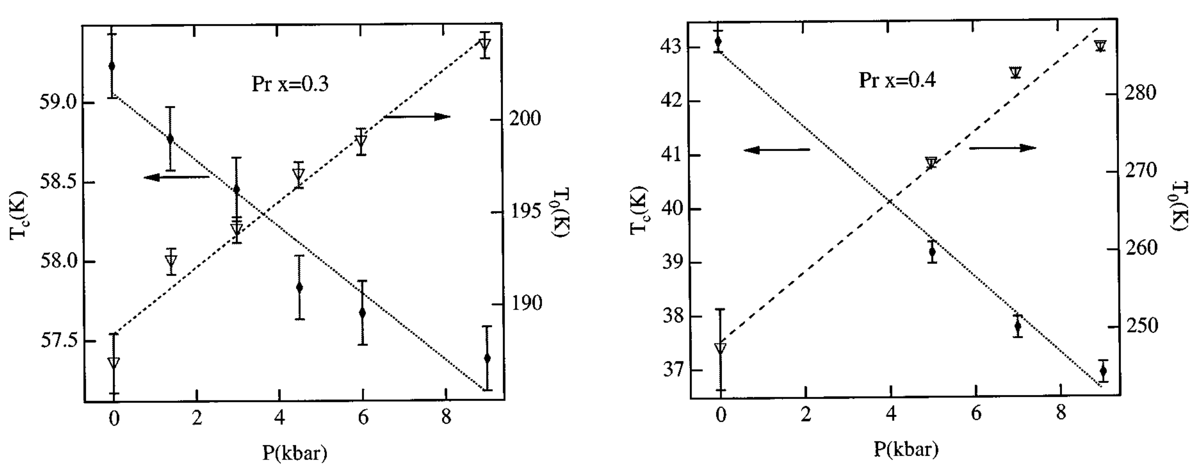

FIG. 4. Evolution of the critical temperature and the T_0 temperature deduced from the exponential fit for the $x=0.3$ and $x=0.4$ Pr-doped YBaCuO layers.

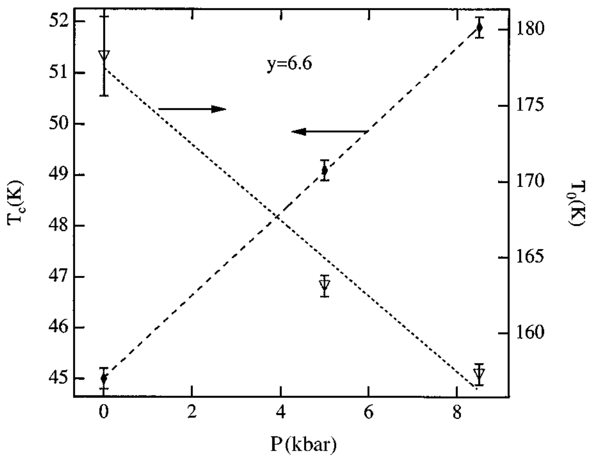

FIG. 5. Evolution of the critical temperature and the T_0 temperature deduced from the exponential fit for an oxygen-deficient YBaCuO layer ($y=6.6$).

fitted for all Pr contents with the expression

$$\sigma_1(T)=C\times\tau(T)\times x_n(T), \tag{1}$$

with $1/\tau(T)\sim\exp(T/T_0)$ representing the decreasing scattering rate below T_0 due to spinon pairing; C is a temperature-independent constant and x_n is the normal fluid fraction. This fit procedure yields a value T_0 which is nearly equal to the observed spin-gap temperature for each doping level. However, the accuracy with which T_0 is obtained is at least one order of magnitude better than the case where T_0 is obtained from the analysis of the deviation from the linear behavior. A detailed error analysis of the latter procedure resulted in an estimated error of about 3 K. An example of a fit using Eq. (1) through the data of the dc-resistance under pressure is shown in Fig. 3 for the $x=0.4$ Pr-doped sample at 7 kbars. The exponential function is fitted through the resistance data in a temperature range between 60 and 150 K and a linear curve is fitted through the high-temperature data between 220 and 290 K. In between those temperature domains and in the temperature range just below T_c we have transition regimes which are difficult to describe.

Similar fits can be performed for the $x=0.3$ Pr-doped sample; for the $x=0.2$ Pr-doped sample, the temperature region where we have a clear exponential behavior is too small to extract reliable T_0 values. The results of this analysis for the $x=0.3$ and $x=0.4$ samples are summarized in Fig. 4 and Table I. We observe an increases of T_0 with applied pressure parallel with the decrease of the critical temperature. Additionally, the deduced T_0 temperatures are in the same range of the values found in the fits of the THz conductivity measurements of Ref. 11.

These measurements clearly show that changing the amount of carriers by pressure has an effect on both the critical temperature and the spin-gap temperature and that we can follow the phase diagram of the mean-field RVB theory in a way similar to changing the carrier concentration by adding Pr or removing oxygen.

For the fact that the spin-gap temperature exhibits a stronger pressure dependence than the critical temperature, a possible explanation can be the shorter distance between the CuO_2 planes resulting in an enhanced interplane coupling of the Cu^{2+} spins. According to the calculations of Ubbens and Lee[9] showing that the spinon pairing is predominantly caused by interaction between the different CuO_2 planes rather than by an intraplane interaction, we expect an enhanced spinon pairing when pressure is applied. We checked the influence of the shorter interplane distance with the measurements on the oxygen-depleted layer, for which the critical temperature *increases* with pressure. If only the modified number of carriers is playing an important role, the spin-gap temperature should decrease with applied pressure, following the RVB phase diagram. If the enhanced spinon pairing is the major effect, the spin-gap temperature should increase. In Fig. 5 and Table II the results of the pressure measurements on a YBaCuO layer with an oxygen content $y=6.6$ are summarized: a distinct increase of the critical temperature with pressure is observed, together with a strong decrease of the temperature T_0. The increase of the number of carriers in the CuO_2 planes induced by the pressure leads to an opposite effect on both transition temperatures, consistent with the RVB model.

TABLE II. Critical temperatures and T_0 values deduced from the exponential fit for the oxygen-deficient YBaCuO layer ($y=6.6$).

O content	P (kbar)	T_c (K)	T_0 (K)
$y=6.6$	0	45.0 ± 0.2	178 ± 2.6
	5.7 ± 0.3	49.1 ± 0.2	163 ± 0.7
	9.1 ± 0.3	51.9 ± 0.2	157 ± 0.7

From this experiment, we can conclude that the charge transfer is the major effect when we apply an external pressure on an underdoped YBaCuO film and that the effect of a decreased interplanar distance is either very small or nonexisting. Concerning this problem, it is also important to note how the different interatomic distances change with pressure. Structural changes in $YBa_2Cu_4O_8$, differing from $YBa_2Cu_3O_7$ in that its unit cell contains four CuO chains instead of two, are measured as a function of pressure.[12] It is observed that the relative change of the in-plane apical oxygen distance Cu(2)-O(1) with pressure is larger than the compression of the c axis, whereas the relative change in the distance between the CuO_2 planes is rather small in measurements up to 6.32 kbars. On the other hand, the enhanced Pr-O hybridization is also an effect of this smaller interplanar distance, demonstrating its importance.

In conclusion, we measured the pressure dependence of both the critical and the spin-gap temperature in Pr-doped and oxygen-deficient YBaCuO. The measured critical and spin-gap temperatures as a function of pressure for the different dopant concentrations fit very well in the phase diagram of Anderson's RVB theory: the increase of the critical temperature with increasing hole content induced by the pressure, is accompanied by a decrease in the spin-gap temperature (for underdoped compounds). No significant effect of an increased interplane coupling of the Cu^{2+} spins is observed.

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