Planar $^{17}$O NMR study of Pr$_y$Y$_{1-y}$Ba$_2$Cu$_3$O$_{6+x}$

W.A. MacFarlane, J. Bobroff, P. Mendels, L. Cyrot, H. Alloul, and N. Blanchard
LPS, Bât. 510, Université Paris-Sud, UMR8502 CNRS, 91405, Orsay Cedex, France

G. Collin
LLB, CEN Saclay, CEA-CNRS, 91191 Gif-sur-Yvette, France

J.-F. Marucco
LCNS, Université Paris-Sud, 91405, Orsay, France
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We report the planar $^{17}$O NMR shift in Pr substituted YBa$_2$Cu$_3$O$_{6+x}$, which at $x=1$ exhibits a characteristic pseudogap temperature dependence, confirming that Pr reduces the concentration of mobile holes in the CuO$_2$ planes. Our estimate of the rate of this counterdoping effect, obtained by comparison with the shift in pure samples with reduced oxygen content, is found insufficient to explain the observed reduction of $T_c$. From the temperature dependent magnetic broadening of the $^{17}$O NMR we conclude that the Pr moment and the local magnetic defect induced in the CuO$_2$ planes produce a long range spin polarization in the planes, which is likely associated with the extra reduction of $T_c$. We find a qualitatively different behaviour in the oxygen depleted Pr$_y$Y$_{1-y}$Ba$_2$Cu$_3$O$_{6.6}$, i.e. the suppression of $T_c$ is nearly the same, but the magnetic broadening of the $^{17}$O NMR appears weaker. This difference may signal a weaker coupling of the Pr to the planes in the underdoped compound, which might be linked with the larger Pr to CuO$_2$ plane distance, and correspondingly weaker hybridization.

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Among the isostructural Rare Earth analogues of the well-studied high-$T_c$ system, YBa$_2$Cu$_3$O$_{6+x}$, only Pr/Y substitution suppresses superconductivity. The most widely accepted explanation of this effect is that it is due to counterdoping of the planes, i.e. the concentration of mobile holes in the planes is depleted, resulting in the insulating behaviour found for $y \approx 0.6$ (e.g. Ref.[1]). Several proposals for the origin of this counterdoping have been advanced; for example, the valence of Pr may be larger than 3+ (e.g. Ref.[2]), the large ionic radius of Pr$^{3+}$ may influence the chain-planes charge transfer (similar to other large trivalent ions[3]), or strong hybridization of Pr with its neighbouring planar oxygen may cause holes to be removed from the conduction band into localized or insulating-band states. X-Ray and Cu NQR experiments[4] apparently rule out the first two possibilities.

Planar $^{17}$O NMR provides a sensitive probe of the spin susceptibility ($\chi_s$) of the CuO$_2$ planes in the cuprate high $T_c$ superconductors. In particular, in YBa$_2$Cu$_3$O$_{6+x}$, $\chi_s(T)$ has been shown to be strongly dependent on the carrier content ($n$) of the planes, especially near $T_c$. Optimal doping[5]. In the present work, we argue that determining $\chi_s(T)$ provides a measure of $n$ in the normal state of Pr$_y$Y$_{1-y}$Ba$_2$Cu$_3$O$_7$ (Pr$_y$O$_x$). Such a local measurement of $n$, which is free of any influence of parasitic phases, quantitatively constrains models of counterdoping. Furthermore, we find a Curie-like line broadening of the $^{17}$O NMR indicating that the Pr ion is a magnetic perturbation which induces a long range oscillatory spin polarization response in the CuO$_2$ planes, as has been found for Ni and Zn in-plane substitutions[6]. This effect is somewhat analogous to the RKKY oscillations found in conventional metals containing magnetic impurities. Such a perturbation may be responsible for the extra reduction of $T_c$, which is obtained after removing the net counterdoping effect.

Finally, motivated by results from underdoped YBa$_2$Cu$_3$O$_{6+x}$ with Zn and Ni as in-plane Cu substitutions, which exhibit enhanced local magnetic effects and $T_c$ reduction relative to optimal doping[7] we have also studied deoxygenated samples (Pr$_y$O$_{6.6}$). The results indicate that Pr induces a weaker magnetic perturbation and is therefore less coupled to the CuO$_2$ planes in Pr$_y$O$_{6.6}$.

After detailing the samples and technical aspects of the measurement, we present the results for Pr$_y$O$_x$ then Pr$_y$O$_{6.6}$. Then we discuss the data in relation to the results of other experiments.

I. SAMPLES

The powder samples were synthesized by conventional high temperature solid state reaction of Pr$_6$O$_{11}$, Y$_2$O$_3$, BaO$_2$ and CuO as detailed in Ref.[8]. This synthesis procedure reproducibly results in a family Pr$_y$Y$_{1-y}$Ba$_2$Cu$_3$O$_7$, of compounds with monotonically decreasing $T_c(y)$[9]. Pr is not expected to affect the maximal oxygen uptake of the sample significantly[10], and this was confirmed thermogravimetrically, i.e. $\delta x \leq 0.008$ for $y=0.2$ relative to $y=0$. The presence of Pr substituted on the Ba site can be detected by Nuclear Quadrupolar Resonance (NQR) of the chain Cu as has been established by inten-
tional Pr/Ba substitution. The chain NQR spectrum for samples synthesized by us with the same procedure, but with much larger Pr content is the same as that of pure YBa$_2$Cu$_3$O$_7$, indicating that for our samples Pr/Ba substitution is negligible. The samples were isotopically enriched by annealing in a $^{17}$O$_2$ enriched atmosphere. Subsequent deoxidization was accomplished by vacuum annealing under thermogravimetric control. The samples were aligned by suspension in Stycast 1266 epoxy cured in a 7.5 T magnetic field. Alignment was checked using $^{89}$Y NMR at 300 K. It was found that for $y \leq 0.1$, the samples were well aligned, but for $y = 0.2$, the powder did not completely align. However, systematic differences in the results due to imperfect alignment are negligible in comparison to the $n$ dependent contribution of $\chi_s(T)$.

II. NMR TECHNICAL ASPECTS

A. Experimental Method

Conventional spin echo NMR spectra of the central transition of the plane oxygen (O(2,3)) were taken in the temperature range 80 to 350 K with the field aligned parallel to the c-axis (except as noted above). The typical $\pi/2-\tau/\pi$ sequence consisted of $\sim 1\mu s \pi/2$ pulse and a $\tau$ of $\sim 100\mu s \ll T_2$. The contrast in spin lattice relaxation rates ($T_1^{-1}$) between the plane and apical O(4) oxygen sites was exploited (using fast repetition $\sim 50$ ms) to eliminate contamination from O(4). Typical Fourier transformed spectra are illustrated in Fig. 1. From the raw spectra, the effect of Pr substitution is clearly to cause temperature dependent shift and broadening of the resonance. As shown below, the temperature dependent shift is very similar to deoxidized pure YBCO$_7$. We analyze the frequency spectra by extracting the peak position and the width $\Delta \nu$ defined as twice the upper frequency halfwidth at half maximum. This definition of $\Delta \nu$ excludes the contribution to the linewidth of a slight inhomogeneity of $n$. This inhomogeneity of about $\pm 5\%$, which is also found in pure compounds, induces a larger broadening of the low frequency side of the line.

The average Knight shift $^{17}K_c$ was extracted from the peak position as follows. We correct for the small second order quadrupolar shift of the central line ($\nu_1^{(2)}$) assuming the same values of the quadrupolar interaction as in pure YBa$_2$Cu$_3$O$_{6+x}$. For the field along c, $\nu_1^{(2)}$ varies from 12.1 kHz ($x = 1$) to 10.6 kHz ($x = 0.6$) and is independent of temperature. These corrections are not large (less than 300 ppm at the experimental field), and are very weakly doping dependent. Thus, although one does expect changes in the average quadrupolar interaction with $y$ between the extremes of $y = 0$ and $y = 1$, such effects are small compared to the $T$ dependent part of the shift.

B. Calibration of $^{17}K_c(2,3)$

In order to compare the shift effect to the doping effect of changing the oxygen content in pure YBCO$_{6+x}$, a set of curves was developed for $0.6 \leq x \leq 1$ to summarize the changes of the shift in terms of the single parameter $x$, see Fig. 2. The function, based on a well-known phenomenological pseudogap $T$-dependence, was of the form:

$$^{17}K_c(T, x) = a(x) - bT + c(1 - \tanh^2(T^+(x)/T)),$$

where $T^+$ is a measure of the pseudogap. The $T$ profile of the shift varies strongly with $x$ for $x < 0.85$ (Fig. 2). The dependence of the shift is weaker for $x < 0.85$. In particular for $0.6 < x < 0.85$, the main effect is a $T$ independent downward offset of $^{17}K_c$, while for $0.45 < x < 0.6$ there is additionally a compression of the variation of $^{17}K_c$, e.g. in the difference between 300 K and 100 K. Thus measurement of the $T$ profile of the shift constitutes a measure of the doping level of the CuO$_2$ planes with high sensitivity near optimal doping and less sensitivity at lower doping.

III. RESULTS FOR PR$_y$Y$_{1-x}$BA$_2$CU$_3$O$_7$

A. Shifts: The Effect on Doping

For the Pr$_y$O$_7$ samples, the measured shifts relative to the $^{17}$O frequency in water are shown in Fig. 3 together with the curves for oxygen depleted pure YBa$_2$Cu$_3$O$_{6+x}$.
the resemblance is so strong (Fig. 3) that it is quite reasonable to assume a common origin for the evolution of the $T_c$ profiles, i.e. a reduction of $n$. Moreover, measurements of quite different properties (e.g. $\chi_s$) also find evidence of a reduction of $n$. Henceforth, we will simply assume this is the case and proceed with an analysis based on the phenomenological model of Eq. (1). From the fits, we can thus extract a value for the equivalent oxygen concentration $x^{eq}$ as a function of Pr content $y$, which is reported in Fig. 4 together with a linear fit.

The decrease of $T_c$ with $y$ is much more rapid than expected from counterdoping alone. This can be seen by comparing $T_c(x)$ for the oxygen depleted materials with $T_c(x^{eq})$ (Fig. 5). The difference between the two curves clearly shows that another mechanism of $T_c$ suppression is present. The excess reduction in $T_c$, $\Delta T_c^{ex}$, has been extracted as shown in Fig. 5, and is plotted as a function of Pr concentration in Fig. 6. The slope of $\Delta T_c^{ex}(y)$ is about 0.51 K/\%.

Let us compare this result with former studies. Previous Cu NMR measurements for Pr substituted samples also found underdoped behavior in both the plane copper shift and spin lattice relaxation temperature dependence. Also using simultaneous Ca/Y and Pr/Y substitution a somewhat larger slope of $\Delta T_c^{ex}(y)$ was inferred. But in both cases there was no direct measurement of the counterdoping effect of Pr. Here the specific advantages of the $^{17}$O NMR, such as its high sensitivity and intrinsically narrow resonance, together with the systematics of our study have allowed us to reach accurate quantitative conclusions.

If we extrapolate from the dilute regime of our measurements, the observed counterdoping effect is already sufficient to explain the metal-insulator transition at...
$y \approx 0.6$, where $x^{eq} \approx 0.4$. But, as the decrease of $T_c$ is even larger than that obtained from the counterdoping alone, we expect a non-superconducting part of the phase diagram whose ground state is likely an insulating magnetically disordered state, similar to Zn substituted samples. But, as the decrease of $T_c$ is even larger than that obtained from the counterdoping alone, we expect a non-superconducting part of the phase diagram whose ground state is likely an insulating magnetically disordered state, similar to Zn substituted samples.

### B. Broadening: Magnetic Effects

We can gain more insight into the effects induced by Pr substitution and more specifically on the origin of $\Delta T_c^\text{ex}$ from the $^{17}$O linewidths, $\Delta \nu$. The observed $\Delta \nu$ are presented in Fig. 7 together with that of the pure system. It is apparent that there is an excess broadening related to the Pr concentration which increases as $T \to 0$. As Pr is a paramagnetic ion with a local susceptibility possessing a Curie $T^{-1}$ component, it is natural to associate the line broadening with such a magnetic contribution. Indeed $\Delta \nu(T)$ in Pr$_y$O$_7$ scales approximately with the Pr susceptibility. To interpret this broadening, we move now to a discussion of its origin.

The spatial distribution of local fields due to randomly placed magnetic impurities is expected to give rise to a broadening of the host nuclear resonance lines which scales with the impurity contribution to the susceptibility, typically a Curie law for dilute weakly interacting impurities. In contrast, dilute nonmagnetic disorder generally produces a temperature independent broadening due, for example, to a distribution of electric field gradients.

The local magnetic field always contains the direct dipolar field of the impurity moments, but in metallic systems the field distribution sampled by the nuclei is usually dominated by (RKKY) spin polarization of the conduction band. Let us first demonstrate that the computed dipolar fields are negligible compared to the measured broadening. Although the precise nature of the Pr moment is still controversial, its contribution to the macroscopic susceptibility is accounted for (in this $T$ range) by a $\approx 2.7 \mu_B$ Curie term plus a constant. In the dilute limit, the dipolar broadening may be estimated as

$$\Delta \nu = \frac{8\pi}{9\sqrt{3}} n D \langle S_z \rangle,$$

where $n$ is the volume concentration of the randomly placed impurities, $D$ is the dipolar coupling to the nuclei with gyromagnetic ratio $\gamma_n$, i.e. $D = 2 \mu_B \gamma_n$, and $\langle S_z \rangle$ is the thermodynamically averaged impurity spin polarization, proportional to the impurity susceptibility. Substituting the values for the Pr moment and the oxy-

![Graph](image-url)
The reported decrease in the effectiveness of Pr in reducing \( T_c \) in \( \text{Pr}_x\text{O}_y\) suggests that the influence of Pr is weaker in the underdoped region of the phase diagram than at optimal doping. In contrast, our value of \( \Delta T_c/y \) (0.94 K%/%) is more than twice this previous report (triangles, Fig. 6), and has about the same magnitude as the \( T_c \) reduction for YBCO-Pr. To investigate the role of Pr substitution in the deoxygenated samples, we studied the \( ^{17}\text{O} \) NMR in two \( \text{Pr}_x\text{O}_y \) samples corresponding to the highest Pr concentrations studied at full oxygenation in the previous section, \( y = 0.1 \) and 0.2.

In the pure samples, the variation of the NMR shifts with oxygen content is much weaker in the underdoped regime than near optimal doping (see Fig. 2). Only a small reduction of the difference of \( ^{17}\text{O} \) between 100 and 300 K is observed, from \( x = 0.65 \) to \( x = 0.45 \) so that one cannot obtain a very accurate determination of the counterdoping from NMR. The data for both the \( y = 0.1 \) and 0.2 samples appear to be simply shifted versions of that for the unsubstituted \( x = 0.6 \) sample (Fig. 8). This observed change in the \( T \) independent shift contribution appears too large to be of quadrupolar origin (see above), especially since no such effect was found in the \( O_7 \) composition. It is also large relative to the \( (T \) independent) orbital shift, about 100 ppm in YBCO-7. Such behaviour precludes a quantitative estimate of the underdoping effect since the calibration of Section III.B does not apply. However, the absence of a reduction of the difference of \( ^{17}\text{O} \) between 100 K and 300 K with increasing Pr content suggests that again counterdoping is insufficient to explain the \( \sim 20\text{K} \) decrease of \( T_c \) observed for \( y = 0.2 \).

Let us now consider the Pr-induced broadening of the \(^{17}\text{O} \) line, which has been linked for YBCO-7 with magnetic effects associated with the Pr. This extra linewidth is quite small for the sample with \( y = 0.1 \). For \( y = 0.2 \) it is larger and increases very rapidly at low \( T \) in the underdoped regime (Fig. 9), much faster than a Curie law. This is reminiscent of the situation encountered in underdoped YBCO-6.6 for substitution of the plane Cu by a magnetic ion, such as Ni, where the rapid \( T \) variation of the width has been shown to be a consequence of the \( T \) dependent enhanced magnetic response \( \chi’(q) \) of the underdoped planes. There is no reason to expect this characteristic of the underdoped planes to be altered by Pr substitution, so a detailed comparison with the case of Ni may be instructive. For Ni/plane Cu substitution, the broadening \( \Delta \nu(100\text{K}) \) of the plane oxygen NMR was found 3 to 4 times larger than near optimal doping, while the magnitude of the susceptibility of Ni does not change significantly with oxygen content. However, here \( \Delta \nu(100\text{K}) \) for the same concentration \( y \) of Pr is smaller for \( \text{Pr}_y\text{O}_6 \) compared to \( \text{Pr}_y\text{O}_7 \). As for the Pr effective moment, its magnitude is not significantly modified from \( x = 1 \) to \( x = 0.6 \) by the changes in crystalline electric field at the Pr site. Therefore the large decrease in linewidth compared to the \( \text{Pr}_y\text{O}_7 \) linewidth data indicate that the coupling of the Pr moment to the planes is reduced in the underdoped state. We discuss this point in the next section together with the data in the optimally doped composition.

V. DISCUSSION

The data presented here demonstrate unambiguously that in \( \text{Pr}_y\text{O}_7 \) Pr acts not only as a counterdopant (reducing \( n \)), but also as a magnetic defect. This is reflected in the long-range oscillatory spin polarisation which gives rise to the broadening of the plane oxygen NMR. The magnetic defect couples strongly to the CuO planes in YBCO-7, while the coupling apparently decreases for deoxygenated samples.

The effective local magnetic moment of the Pr ion couples directly to the planes, in contrast to the moments of other rare earth ions substituted on the Y site. Indeed the ordering temperature of the Pr moments (17 K) in pure nonmetallic PrBaCuO-7 is much larger than the or-
dering temperature of Gd in GdBaCuO$_7$, which is a high Tc superconductor, indicating stronger superexchange paths through the planes for Pr than for Gd. Such a coupling via, for example, hybridization of Pr with its near neighbor oxygens as suggested by Fehrenbacher and Rice, would also result in a magnetic pair breaking effect.

Additionally such a hybridization should change the superexchange interaction between the Cu hole spins adjacent to the Pr ion. Such a local perturbation in the Cu-Cu interactions may induce an extended magnetic defect in the plane, as does any in-plane perturbation. This magnetic defect would contribute both to the induced spin polarization and the T$_c$ depression, similar to Zn and Ni impurities.

We do not at this stage have enough detailed information regarding the local magnetic properties around the Pr impurities to distinguish between these two contributions, as the NMR line of Fig. 1 is comprised of spins adjacent to oxygen nuclei.

We can, however, qualitatively consider the likely evolution of these effects with decreasing $x$, that is deoxygenation. The established increase of the Pr-O(2,3) distance due to removal of chain oxygen is compatible with a weaker effect of Pr in the underdoped regime. In particular it could cause both a decrease of the exchange coupling of the Pr moment to the planes and a weaker induced magnetic defect in the planes. The former may be responsible for the monotonic decrease of the Pr ordering temperature to about 11 K as $x \to 6$ found in PrBCO.

Another important method of characterizing the Pr defect is through its effect on charge transport, i.e. the residual resistivity $\rho_0$. The anomalous behaviour of in-plane Zn defects and radiation induced disorder have been studied in detail, and the correlation between the reduction in T$_c$ and the increase in $\rho_0$ has been found to depend strongly on $n$. Without resorting to a microscopic formulation of the impurity scattering or its effect on T$_c$, it is of interest to ask whether the Pr defect behaves qualitatively different from the in-plane defects. Until now such a comparison was not possible as the counterdoping effect could not be separated from the scattering. Our measurements provide $\Delta T_c = \Delta T_c^{ex}$ and $n$ at full oxygenation, but transport data on high quality single crystals is lacking. Some measurements on Pr$_{6.6}$O$_{17}$ have been reported and compared to radiation damage. However, we find that $\rho_0$ from Ref. 41 is unreasonably large. Indeed for a 10% reduction of T$_c$, $\rho_0 \approx 5600 \Omega$/plane while for Zn it corresponds to $\Delta \rho_0 < 1000 \Omega$/plane. In order to see if this difference indicates that Pr is quite distinct from Zn, it will be necessary to confirm the reported values of $\rho_0$. Other techniques, such as microwave and optical conductivity, that yield the impurity contribution to the carrier scattering rate are also of interest in a better quantitative analysis of $\Delta T_c^{ex}$.

VI. CONCLUSION

In conclusion, from measurements of planar $^{17}$O NMR, we are able to deduce the net hole doping in the planes accurately in Pr$_y$:O$_7$. Counterdoping by Pr is observed at Pr$_{6.6}$O$_{17}$, but is not sufficient to explain the reduction of T$_c$. The rate of counterdoping (summarized by $x^{eq} \approx 1 - 0.86y$) is important both as a constraint on theories of the T$_c$ suppression and as a means of interpreting the results of other experiments in which the doping level is not directly measured. We suggest that the excess loss of T$_c$ is related to scattering of the remaining mobile holes in the CuO$_2$ planes by the Pr defects. This might be confirmed by obtaining accurate estimates of the residual resistivity due to Pr and reliably obtaining the correlation $\Delta T_c^{ex}(n, \Delta \rho_0)$. It has been demonstrated that the Pr defect induces an in-plane magnetic response which is due to the coupling of the Pr moment itself to the planes and to the local modification of superexchange between copper holes near Pr. In Pr$_{y}$:O$_{6.6}$, the smaller influence of the magnetic perturbation can be attributed to the increase of the Pr-O distance.

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* Current Address: Department of Chemistry, University of British Columbia, Vancouver, Canada V6T 1Z1.

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