Magnetic ordering in $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ as evidenced by muon spin relaxation

D. W. Cooke, R. S. Kwok, and M. S. Jahan
Los Alamos National Laboratory, Los Alamos, New Mexico 87545
R. L. Lichti and T. R. Adams
Texas Tech University, Lubbock, Texas 79409
C. Boekema and W. K. Dawson
San Jose State University, San Jose, California 95192
A. Kebede, J. Schwegler, J. E. Crow, and T. Mihalisin
Temple University, Philadelphia, Pennsylvania 19122

Using the zero-field-muon-spin-relaxation (μSR) technique clear evidence has been found for antiferromagnetic ordering of Cu moments within the CuO planes of $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$. The Néel temperatures are approximately 285, 220, 35, 30, and 20 K for $x = 1$, 0.8, 0.6, 0.58, and 0.54, respectively. For $x = 0.50$ we observe a fast-relaxing component of the muon polarization in addition to a long-time tail, reminiscent of spin-glass behavior. This region of the phase diagram ($0.5 \leqslant x \leqslant 0.54$) corresponds to the existence of both superconductivity and magnetism. The fully developed local magnetic field for $x > 0.54$ is found to be ~ 16 mT, but decreases to ~ 12 mT at $T = 17$ K for the $x = 1$ sample, presumably due to the onset of Pr-ion ordering. Magnetic ordering also occurs in $PrBa_2Cu_3O_6$; the Néel temperature is ~ 325 K.

I. INTRODUCTION

The interest in $(Y_{1-x}Pr_x)Ba_2Cu_3O_x$ stems from several factors. First, unlike the other rare-earth-based superconductors, $PrBa_2Cu_3O_7$ (PBCO) is insulating rather than metallic or superconducting. It is not understood why the substitution of Pr quenches superconductivity when other rare-earth element substitutions produce superconductors with $T_c \sim 90$ K. Experimentally it has been found that T_c vs x follows the classic Abrikosov–Gor'kov pair-breaking curve with $T_c \rightarrow 0$ K for $x \cong 0.6$, leading to the suggestion that the depairing mechanism is presumably due to strong *f-spd* hybridization between the nearly tetravalent Pr ions and the charge carriers in the adjacent Cu–O planes.[1]

Second, the system remains orthorhombic for $0 \leqslant x \leqslant 1$; therefore, complete pseudobinary alloys of $YBa_2Cu_3O_7$ (YBCO) and PBCO can be prepared. In addition, the oxygen content remains stable for $0 \leqslant x \leqslant 1$. Because this system exhibits both superconductivity ($x \leqslant 0.6$) and magnetism (Pr-moment ordering for $0.4 \leqslant x \leqslant 1.0$), and because it remains orthorhombic for $0 \leqslant x \leqslant 1$, it is a unique system for investigating the interplay between these two ground states without complications which arise from structural changes or variable oxygen content.

We have used zero-field muon-spin-relaxation (μSR) to examine the magnetism of $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ as a function of concentration and temperature. In addition to the previously reported superconductivity and magnetism (Pr ordering), we find that antiferromagnetic ordering also occurs and that the associated Néel temperatures are concentration dependent. We attribute this magnetism to ordering of Cu moments within the Cu–O planes, i.e., to Cu(2) ordering. Previous Mössbauer studies on $(Y_{1-x}Pr_x)Ba_2(Cu_{3-y}Fe_y)O_z$ ($x = 0,1$; $y = 0$, $z = 6,7$) also revealed magnetic ordering of Cu(2) (Ref. 2), but concluded that there was no overlap of superconductivity and antiferromagnetism. The present results suggest that spin-glass-like magnetism coexists with superconductivity in the $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ system.

II. EXPERIMENTAL ASPECTS

Polycrystalline samples of $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ ($0 \leqslant x \leqslant 1$) were prepared using conventional solid-state reaction techniques.[1] High-resolution x-ray diffraction studies indicated that the samples crystallized in an orthorhombic structure for all values of x. Both x-ray and neutron-diffraction studies indicated that there were less than 2% impurity phases in all samples, and that phase separation[3] of the constituents into YBCO with $T_c = 90$ K and $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ of varying x value with reduced T_c did not occur. The effective moment determined from susceptibility measurements suggests that the Pr ions are close to tetravalent. We note, however, that other researchers using different experimental techniques have concluded that the Pr ions are trivalent.[4]

The μSR experiments were done at the stopped muon channel of the Los Alamos Clinton P. Anderson Meson Physics Facility (LAMPF) using standard zero-field techniques.[5] The essential features of the technique are briefly described. Spin-polarized positive muons are implanted into a sample and the decay positrons are detected by counters located in the forward (direction of initial muon momentum) and backward directions. In zero external field (nulled to $\pm 2\,\mu$T) the muon experiences only the internal magnetic field. Therefore, in an antiferromagnet, one should observe one or more discrete frequencies determined by the magnitude of the local magnetic field at the muon site, i.e., $\omega_\mu = \gamma\mu H_{\rm loc}$ ($\gamma_\mu/2\pi = 1.355$ MHz T^{-1}). In contrast, no discrete frequencies will be observed for randomly oriented internal fields.

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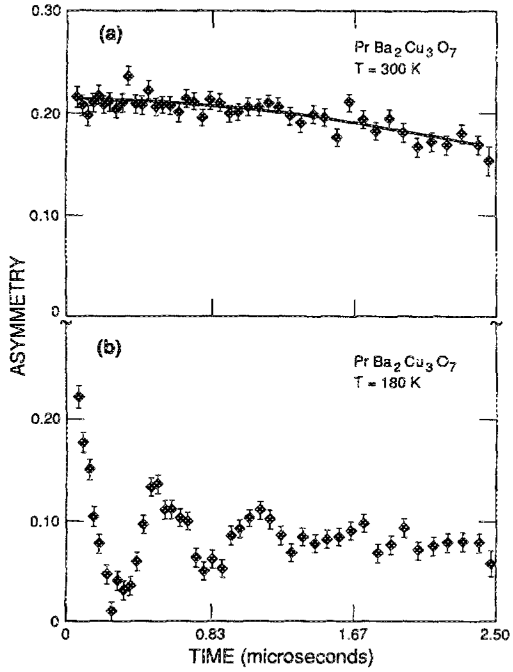

FIG. 1. Zero-field μSR spectra for $PrBa_2Cu_3O_7$ taken at (a) 300 K and (b) 180 K. The oscillatory pattern in (b) is clear evidence for magnetic ordering. No ordering is evident in (a).

III. RESULTS

Representative zero-field μSR spectra for $PrBa_2Cu_3O_7$ taken at 300 and 180 K are shown in Figs. 1(a) and 1(b). At 300 K the internal magnetic field is due to randomly oriented, quasistatic Cu nuclear moments, which depolarize the muon according to $G(t) = \exp[-\frac{1}{2}\sigma^2 t^2]$, where σ is a Gaussian depolarization rate related to the second moment of the local field distribution at the muon site. The solid line of Fig. 1(a) is a Gaussian fit to the data with $\sigma = 0.36\,\mu s^{-1}$. In contrast, the spectrum taken at 180 K shows oscillatory behavior with a well-defined muon frequency. This is clear evidence for the existence of an ordered, local magnetic field. The Fourier transform of Fig. 1(b) shows that the muon precessional frequency is 1.85 MHz. Similar results were obtained for $x = 0.54$, 0.58, 0.6, 0.8, and 1.0. Muon precessional frequencies for $x = 1.0$, 0.8, and 0.6 are shown in Fig. 2. For clarity the $x = 0.58$ and 0.54 results are not plotted. The Néel temperatures are approximately 285, 220, 35, 30, and 20 K for $x = 1$, 0.8, 0.6, 0.58, and 0.54, respectively.

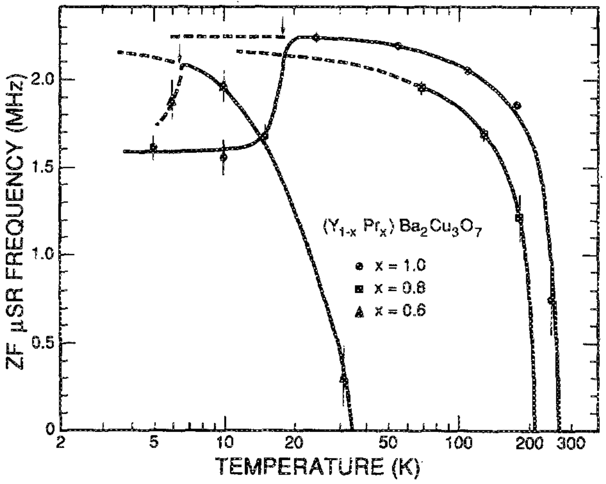

FIG. 2. Zero-field μSR frequencies for $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ ($x = 1.0$, 0.8, and 0.6) as a function of temperature. Néel temperatures are 285, 220, and 35 K, respectively.

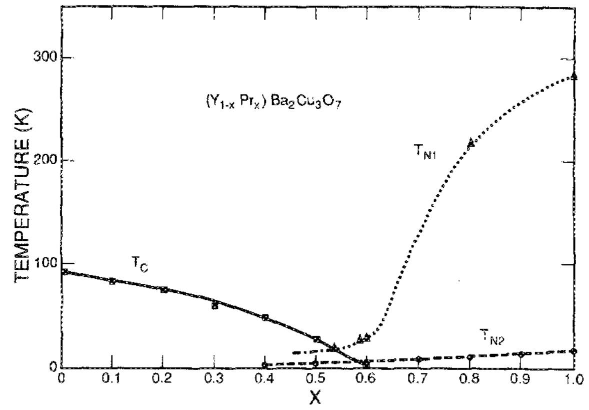

FIG. 3. Phase diagram for $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$. T_{N1} corresponds to antiferromagnetic ordering of Cu moments within the Cu–O planes as determined by zero-field μSR measurements. T_{N2} is attributed to Pr-moment ordering.

From Fig. 2 we see that the zero-field μSR frequencies appear to saturate at a value near 2.2 MHz, which corresponds to a local magnetic field of 16 mT. However, near 17 K ($x = 1.0$) the local field is reduced to ~ 12 mT. This reduction is attributed to the onset of Pr-moment ordering, as observed by magnetic susceptibility,[1] specific heat,[1] and neutron scattering.[6]

The experimentally determined phase diagram for $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ is shown in Fig. 3. T_{N1} is derived from zero-field μSR data and is attributed to antiferromagnetic ordering of Cu(2). T_c is based on susceptibility and resistivity measurements, and T_{N2} (associated with Pr ordering) is deduced from μSR, susceptibility, specific heat, and neutron scattering measurements. μSR data taken in a 100 mT transverse field confirm the T_c vs x curve.

The region $0.4 \leqslant x \leqslant 0.6$ in the phase diagram is of special interest because it represents a crossover from magnetism to superconductivity. In Fig. 4 we show the muon depolarization for $x = 0.6$, 0.58, and 0.50 at a fixed temperature of 5 K. It is clear that a well-defined frequency exists for $x = 0.6$ and 0.58, but not for $x = 0.5$. For the latter concentration it appears that the antiferromagnetic signature, i.e., a well-defined frequency, is absent. For $x = 0.5$ there is a very fast-relaxing component of the muon signal in addition to a long-time tail. We attempted to fit the data of Fig. 4(c) with various known muon relaxation functions. The best fit [shown as the solid line of Fig. 4(c)] was obtained with a spin-glass function, which assumed slow fluctuations of the time-varying local magnetic field[7];

$$g_z(t) = \tfrac{2}{3}(1 - a_0 t)\exp(-a_0 t) + \tfrac{1}{3}\exp(-2t/3\tau). \qquad (1)$$

In Eq. (1) a_0 is the width of the muon precession frequency distribution and τ is the correlation time. From the fitted data we obtained a fluctuation rate of $10^5\ s^{-1}$. However, based upon this fit we cannot conclude that the underlying

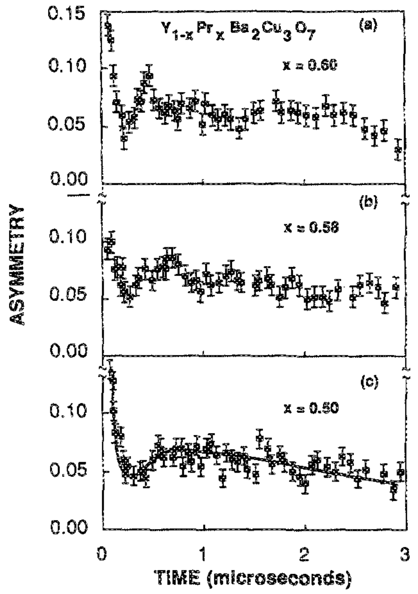

FIG. 4. Zero-field μSR spectra for $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ taken at 5 K: (a) $x = 0.6$, (b) $x = 0.58$, and (c) $x = 0.50$.

magnetism is that of a spin glass, although it would not be unreasonable to assume that the crossover from antiferromagnetism to superconductivity produced a complex magnetic state that resembled a spin glass. This is especially true near $T = 5$ K where the Pr-moment ordering may also be contributing to the formation of this complex ground state, although the *extrapolated* Néel temperature for $x = 0.5$ is only 3.4 K (Ref. 1). Nevertheless, this region of the phase diagram presents a rich arena for investigating the interplay between magnetism and superconductivity in oxide superconductors. This is especially true for the $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ system because it exhibits both magnetism and superconductivity, and retains the orthorhombic structure for $0 \leqslant x \leqslant 1$.

Based on our μSR data we conclude that no clear signature of antiferromagnetism exists in $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ for a concentration of $x = 0.5$. We show a dot-dashed line in the phase diagram (Fig. 3) to represent this fact. Thus the conclusion that *antiferromagnetism* and superconductivity do not coexist in this compound, as offered by Felner *et al.* (Ref. 2), is in agreement with our observations. On the other hand, we suggest that *spin-glass-like magnetism* and superconductivity do indeed coexist.

Zero-field μSR data taken on $PrBa_2Cu_3O_6$ show that magnetic ordering occurs above room temperature, which is the highest operating temperature of our spectrometer. By extrapolating the available data we estimate $T_N \sim 325$ K. Interestingly, two distinct frequencies are observed in this system, which we attribute to the existence of two magnetically nonequivalent muon stopping sites. At 250 and 15 K the measured muon precessional frequencies are $\nu_1 = 1.5$ MHz and $\nu_2 = 3.5$ MHz; $\nu_1 = 2.2$ MHz and $\nu_2 = 4.7$ MHz, respectively.

In summary, zero-field μSR experiments have been conducted on $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ as a function of concentration and temperature. Clear evidence for antiferromagnetic ordering of the Cu moments within the Cu–O planes is found. Néel temperatures vary from 285 to 20 K for $x = 1.0$ and 0.54, respectively. Coexistent spin-glasslike magnetism and superconductivity (in addition to the Pr magnetism) occur for $x = 0.5$. $PrBa_2Cu_3O_6$ is antiferromagnetic with an estimated Néel temperature of 325 K.

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