Magnetic and superconducting phase diagram of $Bi_2Sr_{3-x}Y_xCu_2O_8$ as determined by muon-spin rotation

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Muon-spin-rotation (μ^+SR) studies of $Bi_2Sr_{3-x}Y_xCu_2O_8$ ($x=0.3$, 0.5, 0.6, 0.8, and 1.0) show that the magnetic ordering temperature T_N falls rapidly with increasing hole concentration from $T_N(x=1.0)=210$ K to $T_N(x=0.6)=10$ K. In field-cooled high-field measurements $Bi_2Sr_{2.7}Y_{0.3}Cu_2O_8$ was found to be superconducting with $T_c \approx 65$ K. The resulting phase diagram reflects a transition from magnetic to superconducting phases with increasing hole concentration similar to that found in studies of $YBa_2Cu_3O_{7-\delta}$ and $La_{2-x}Sr_xCuO_{4+y}$.

Many studies of the superconductors $La_{2-x}Sr_xCuO_{4+y}$ and $YBa_2Cu_3O_{7-\delta}$ have shown that the parent compounds of these systems are antiferromagnetic (AFM).[1–7] Previous positive muon-spin-rotation (μ^+SR) studies[1,7–11] have shown that the addition of holes to the antiferromagnetic compounds $YBa_2Cu_3O_6$ and La_2CuO_4, by doping with O^{2-} or through the substitution of Sr^{2+} for La^{3+}, results in a drop in the Néel temperatures (T_N) of both systems. In each compound, further doping eventually leads to a metallic superconducting phase.

The discovery of superconductivity in $Bi_2Sr_2CaCu_2O_8$,[12] which also contains CuO_2 planes, introduced another 2D system in which the dependence of magnetic order and superconductivity on hole concentration could be studied. The carrier concentration of $Bi_2Sr_2CaCu_2O_8$ can be reduced by substituting trivalent Y^{3+} for divalent Ca^{2+} or Sr^{2+}.[13,14] Recent μ^+SR work on the insulators $Bi_2Sr_2YCu_2O_8$ and $Bi_2SrCaYCu_2O_8$ has demonstrated that these systems undergo magnetic transitions at 210 and 15 K, respectively.[15,16] The similarity between the behavior of $Bi_2Sr_2YCu_2O_8$ and the parent compounds YBa_2- Cu_3O_6 and La_2CuO_4 suggested extending this work to samples with a variety of yttrium concentrations. In this paper, we report the results of our muon-spin-rotation studies, performed on the $M15$ and $M20$ muon channels at TRIUMF, on $Bi_2Sr_{3-x}Y_xCu_2O_8$ for $x=0.3$, 0.5, 0.6, 0.8, and 1.0.

Samples were prepared by reacting stoichiometric mixtures of Bi_2O_3, SrO_2, Y_2O_3, and CuO_2 at 900 K for 12–24 h.[14] Subramanian *et al.* performed magnetization and resistivity measurements on the same samples studied in the current paper and found that specimens with $0.2 \leq x \leq 0.4$ are superconducting, while those with $0.5 \leq x \leq 1.0$ are insulators.[14] Recent anomalous scattering synchrotron crystallographic studies on $Bi_2Sr_2CaCu_2O_8$ have demonstrated that cation deficiencies exist on Bi and Sr sites.[17] These vacancies reduce the number of positive ions, thus increasing the effective number of hole carriers in the CuO_2 planes. It is likely that vacancies play a similar role in increasing the number of hole carriers in the specimens of the $Bi_2Sr_{3-x}Y_xCu_2O_8$ system and that the role of Y in both systems is to consume the carriers created by deficiencies on the Bi and Sr sites. Chemical analysis confirms that the hole concentration in the $Bi_2Sr_{3-x}Y_xCu_2O_8$ samples studied decreases with increasing x.[14] X-ray work indicates a crystallographic phase purity greater than 97% for all samples studied.

μ^+SR is an extremely sensitive method of measuring local magnetic fields at interstitial sites in solids.[18–21] A beam of polarized positive muons (μ^+), produced by the weak decay of pions (π^+), is stopped in a target sample. After a μ^+ comes to a rest at a given site, it precesses in the magnetic field produced by both internal and external sources ($\omega=\gamma|\mathbf{B}_{loc}|$ where $\gamma/2\pi=13.55$ kHz/G and $\mathbf{B}_{loc}=\mathbf{B}_{int}+\mathbf{B}_{ext}$). The muon decay positrons, which are emitted preferentially in the direction of the μ^+ spin, can be used to record the evolution of the μ^+ polarization as a function of time. With positron counters (C_1 and C_2) placed on opposite sides of the sample volume, the Fourier transform of the ratio

$$A(t) \propto \frac{C_1(t)-C_2(t)}{C_1(t)+C_2(t)}$$

reflects the distribution of fields seen by the implanted muons.

The magnetically ordered volume fraction of a sample can be measured by applying a weak transverse field (WTF) ($|\mathbf{B}_{ext}| \approx 50\text{G} \leq \mathbf{B}_{int}$) normal to the incident muon polarization (WTF-μ^+SR). In paramagnetic regions of the sample, the magnetic field $\mathbf{B}_{loc}$ at each muon site is approximately equal to the applied external field $\mathbf{B}_{ext}$, and a coherent precession signal is observed. In magnetically ordered regions, $\mathbf{B}_{loc}$ at a given muon site is the vector sum of $\mathbf{B}_{ext}$ and the internal field $\mathbf{B}_{int}$, due to the nearby moments. As the orientation and magnitude of $\mathbf{B}_{int}$ varies from site to site, the distribution of precession frequencies is very broad, resulting in an incoherent signal which is rapidly damped. Hence, neglecting other contributions, the signal remaining after a very short time is proportional to the nonmagnetic volume fraction.[1]

Figure 1 shows the muon precession signals measured in $Bi_2Sr_{2.5}Y_{0.5}Cu_2O_8$ at (a) $T=50$ K and (b) $T=3.0$ K. At 50 K, all the muons precess at a frequency determined by the external field. The very gradual depolarization can be attributed to small inhomogeneities in the local field due to nuclear dipolar broadening. At $T=3.0$ K, the precession signal is the sum of a signal that is rapidly damped and a long-lived signal that reflects that portion of the sample that is not ordered, the background due to muons stopped outside the sample volume, and possibly muon sites in the ordered material at which the internal ordered fields cancel or are very small.

All WTF-μ^+SR data were fitted with the sum of signals:

$$A(t) \propto g(T)e^{-\lambda_\alpha t} + f(T)e^{-\lambda_\beta t}\cos(\omega t+\phi)\ ,$$
$$\lambda_\alpha \gg \lambda_\beta\ .$$

The first term accounts for the rapid depolarization due to

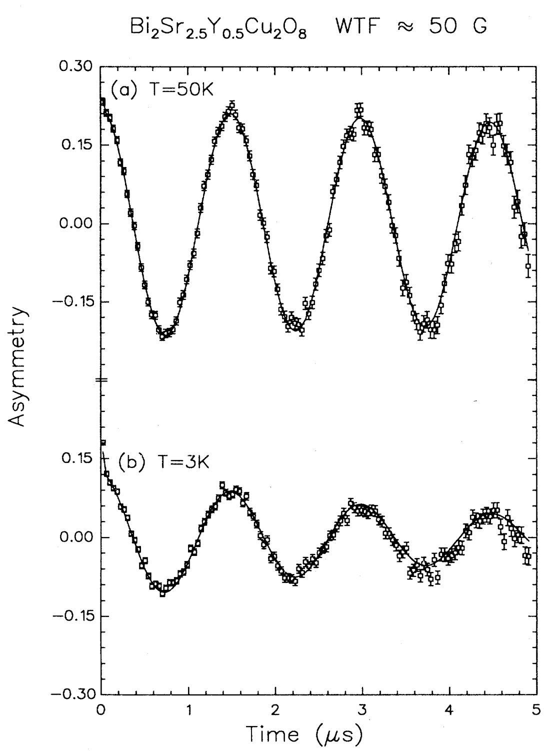

FIG. 1. Muon-spin precession signals seen in $Bi_2Sr_{2.5}Y_{0.5}Cu_2O_{8+y}$ at (a) 50 K and (b) 3.0 K in the presence of a weak transverse field ($\mathbf{B}_{ext} \approx 50$ G). $P(t) \propto [C_1(t)-C_2(t)]/[C_1(t)+C_2(t)]$, where C_1 and C_2 are positron counters positioned on opposite sides of the sample target. The long-lived precession amplitude is roughly proportional to the nonmagnetic volume of the sample.

ordered regions of the target sample, while the second term reflects paramagnetic portions of the sample in which the implanted muons precess at essentially the same rate. The initial amplitude of this latter signal, when normalized by its value when the entire sample in paramagnetic, $f_n(T)=f(T)/f_{para}$, provides an upper limit on the paramagnetic volume fraction. This normalized residual paramagnetic fraction $f_n(T)$ is shown in Fig. 2 as a function of temperature for various values of x in $Bi_2Sr_{3-x}Y_xCu_2O_8$. As can be seen from the figure, $f_n(T)$ decreases over a broad temperature domain for the $x=1.0$ and 0.8 samples, indicating a fairly gradual magnetic transition. For the $x=0.6$ and 0.5 samples, $f_n(t)$ reflects a more sharply defined transition. The inequality of the residual paramagnetic fractions measured at low temperatures may be due to differences in the background caused by different sample mounting geometries. It is also possible that the number of muon sites in the ordered regions of a given sample at which the internal fields cancel may depend on the yttrium concentration. Inhomogeneous distributions of cations within a sample could lead to regions in which no order occurs and, generally, to a broadening of the observed magnetic transition.

The data shown for the $x=0.3$ sample, which is superconducting with $T_c \approx 65$ K, was taken in a much higher field strength ($|\mathbf{B}_{ext}|=2.2$ kG$\gg H_{c1}$) to ensure the formation of a flux lattice below T_c. The initial asymmetry does not change appreciably in the superconducting state as depolarization due to the magnetic-flux lattice tends to take place over a longer time scale [typically $\sigma(T\rightarrow 0)\approx 3\ \mu\text{sec}^{-1}$]. An abrupt increase in the relaxation rate has been seen near 5 K which suggests the onset of magnetic order. Whether this is due to the presence of both superconducting and magnetic phases, or to superconductivity and magnetic order coexisting in a single phase, has yet to be determined.

Using independent measurements of T_c ($x=0.2$ and 0.4, Ref. 14) and assigning to T_N the temperature at

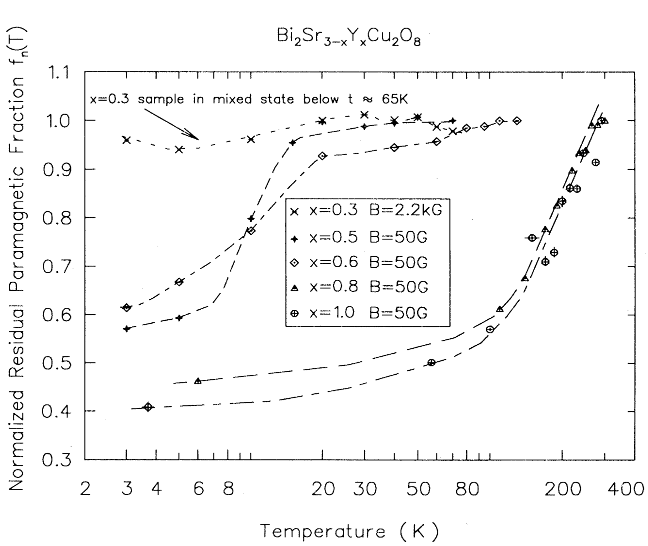

FIG. 2. Normalized paramagnetic volume fraction [$f_n(T)$] vs temperature.

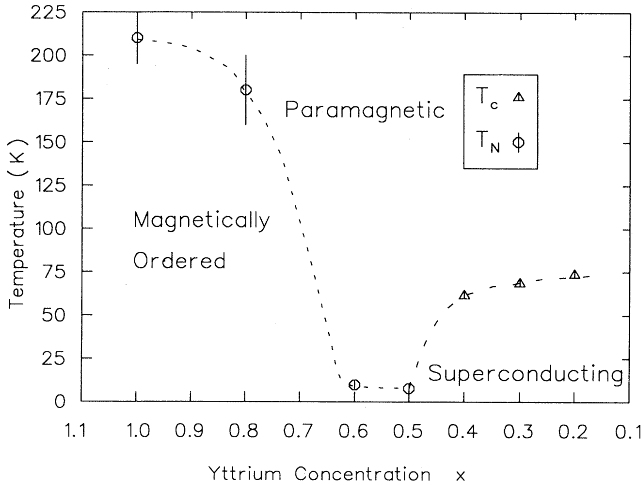

FIG. 3. Néel temperatures (extracted from Fig. 2) and T_c's vs yttrium concentration x. Transverse-field data on superconducting $Bi_2Sr_{2.7}Y_{0.3}Cu_2O_{8+y}$ suggests that magnetic ordering may occur below $T=5$ K.

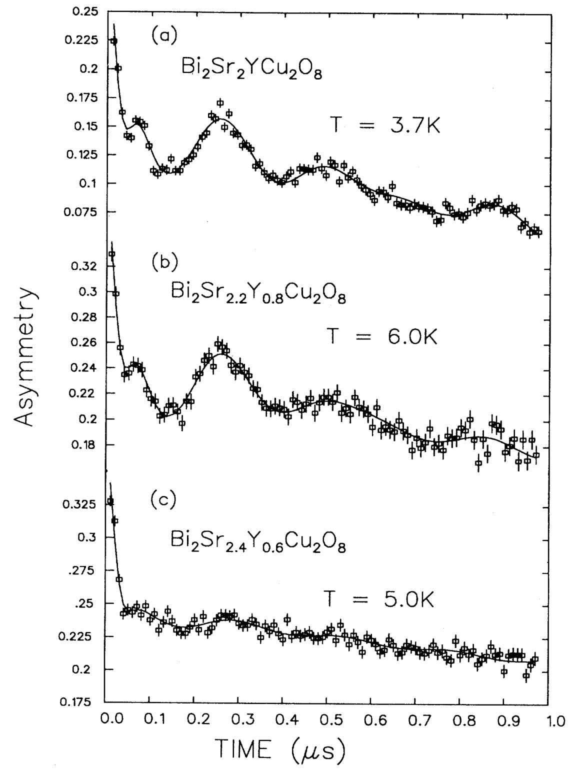

FIG. 4. ZF-μ^+SR spin precession signals seen in (a) $Bi_2Sr_{2.0}YCu_2O_{8+y}$ at $T=3.7$ K, (b) $Bi_2Sr_{2.2}Y_{0.8}Cu_2O_{8+y}$ at $T=6.0$ K, and (c) $Bi_2Sr_{2.4}Y_{0.6}Cu_2O_{8+y}$ at $T=5.0$ K.

which half the participating initial asymmetry in Fig. 2 is gone, a phase diagram relating T_N and T_c to the dopant concentration can be constructed as shown in Fig. 3. This phase diagram is quite similar in form to that found in $YBa_2Cu_3O_{6+x}$ (Ref. 1) and $La_{2-x}Sr_xCuO_{4+y}$.[11] Clearly T_N falls with decreasing Y^{3+} (increasing hole) concentration, dropping sharply around $x=0.7$ and decreasing more gradually as x is reduced. Further substitution of Sr^{2+} for Y^{3+} produces superconducting compounds whose T_c's rise with increasing hole concentration.

The magnetic structure of solids can be characterized by applying the μ^+SR technique in the absence of an external field (ZF-μ^+SR). In ZF-μ^+SR, $\mathbf{B}_{loc}=\mathbf{B}_{int}$, so the resulting muon decay time spectra directly reflect the distribution of internal fields ($\omega=\gamma|\mathbf{B}_{int}|$) seen at the muon site(s). If the field distribution at each muon site is sharply peaked, as is the case in ordered systems, the muons precess coherently and contribute a long-lived oscillatory signal to the total spectrum. In materials with no short-range magnetic order a broad distribution of fields exits at each muon site, resulting in a precession signal that is rapidly damped. In comparatively complex solids like $Bi_2Sr_{3-x}Y_xCu_2O_8$, there may exist a number of different preferred muon sites. Each of these sites will contribute a signal to the total spectrum.[15]

The spectra from ZF-μ^+SR on $Bi_2Sr_{3-x}Y_xCu_2O_8$ for $x=1.0$, 0.8, and 0.6 are shown in Fig. 4. In all three samples, the rapid damping of the muon polarization reflects the broad distribution of fields at the various muon sites in these materials. The oscillating components seen in the $x=1.0$, 0.8, and to a lesser extent, in the $x=0.6$ samples also indicate that at least some short-range order is present. The similarity of the $x=1.0$ and $x=0.8$ spectra demonstrates that the magnitudes of the local moments are comparable in these samples. When the yttrium concentration is reduced to $x=0.6$, the oscillations are damped more quickly, indicating that the distribution of internal fields broadens as the hole concentration is increased. This suggests that the addition of holes reduces the degree of spatial magnetic order present. Additionally, data for $x=0.5$ reflects an even more random field distribution similar to that seen in traditional spin-glass systems.[18]

In contrast to the three-dimensional superconducting and insulating (Ba,K)BiO_3 compounds, in which no evidence of static magnetic order has been seen,[11,22] we have seen magnetic behavior in all insulating samples of $Bi_2Sr_{3-x}Y_xCu_2O_8$ tested. The similarity of the $x=1.0$ and 0.8 spectra suggests that the magnitudes of the ordered moments seen in these two materials are comparable, while the spatial spin correlation length, or the degree of magnetic order, seems to fall as holes are added to the system. The similarity of the magnetic to superconducting transitions found in $La_{2-x}Sr_xCuO_{4+y}$, $YBa_2Cu_3O_{6+x}$, and $Bi_2Sr_{3-x}Y_xCu_2O_8$ strongly suggests that this dependence on the hole concentration is a universal feature of planar CuO_2 high-T_c systems. Recent μ^+SR results on the new electron superconductors[23] indicate that a similar dependence of magnetism and superconductivity on electron concentration exists in electron-doped CuO_2 systems.

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