

Common Energy Scale for Magnetism and Superconductivity in Underdoped Cuprates: A Muon Spin Resonance Investigation of $(\text{Ca}_x\text{La}_{1-x})(\text{Ba}_{1.75-x}\text{La}_{0.25+x})\text{Cu}_3\text{O}_y$

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We characterize the spontaneous magnetic field, and determine the associated temperature T_g , in the superconducting state of $(\text{Ca}_x\text{La}_{1-x})(\text{Ba}_{1.75-x}\text{La}_{0.25+x})\text{Cu}_3\text{O}_y$ using zero and longitudinal field muon spin resonance measurements for various values of x and y . Our major findings are (i) T_g and T_c are controlled by the same energy scale, (ii) the phase separation between hole poor and hole rich regions is a *microscopic* one, and (iii) spontaneous magnetic fields appear gradually with no moment size evolution.

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There is growing evidence that at low temperatures (T), cuprates phase separate into regions that are hole “poor” and hole “rich” [1]. While hole rich regions become superconducting below T_c , the behavior of hole poor regions at these temperatures is not quite clear. Some data support the existence of magnetic moments in these regions. In impure cases, such as Zn or Li doped $\text{YBa}_2\text{Cu}_3\text{O}_7$ YBCO, the impurity creates both the hole poor regions [2] and the magnetic moments [3,4]. In pure cases, such as $(\text{LaSr})_2\text{CuO}_4$ LSCO, these magnetic moments are created spontaneously and undergo a spin-glass-like freezing at T_g [5]. However, there are still many open questions regarding these moments and the spontaneous magnetic fields associated with them. For example, Is there a true phase transition at T_g ? What is the field profile and how is it different from, or similar to, a canonical spin glass? Is the field confined solely to the hole poor regions or does it penetrate the hole rich regions? Also, the interplay between magnetism and superconductivity is not clear. Is strong magnetic background beneficial or detrimental to superconductivity?

In this Letter, we address these questions by performing zero field (ZF) and longitudinal field muon spin relaxation experiments on a series of polycrystalline $(\text{Ca}_x\text{La}_{1-x})(\text{Ba}_{1.75-x}\text{La}_{0.25+x})\text{Cu}_3\text{O}_y$ (CLBLCO) samples. This superconductor belongs to the 1:2:3 family and has several properties that make it ideal for our purpose. It is tetragonal throughout its range of existence $0 \leq x \leq 0.5$, so there is no ordering of CuO chains. Simple valence sums [6], more sophisticated bond-valance calculations [7], and thermoelectric power measurements [8] show that the hole concentration is x independent. As shown in Fig. 1, by changing y , for a constant value of x , the full superconductivity curve, from the underdoped to the overdoped, can be obtained. Finally, for different Ca contents, parallel curves of T_c vs y are generated. Therefore, with CLBLCO one can move continuously, and with minimal structural changes, from a superconductor resembling YBCO to one similar to LSCO.

The preparation of the samples is described elsewhere [9]. T_c is obtained from resistivity measurements. We also

verified using transverse field muon spin resonance (μSR) that CLBLCO respects the Uemura relations and that it is a bulk superconductor.

The μSR experiments were done at two facilities: ISIS, Rutherford Appleton Laboratory, UK, and Paul Scherrer Institute (PSI), Switzerland. Most of the data were taken with a ^4He cryostat. However, in order to study the internal field profile we had to avoid dynamical fluctuations by freezing the moments completely. For this purpose we used the ^3He cryostat at ISIS with a base temperature of 350 mK.

Typical muon asymmetry [$A(t)$] depolarization curves, proportional to the muon polarization $P_z(t)$, are shown in Fig. 2(a) for different temperatures in the $x = 0.1$ and $y = 7.012$ ($T_c = 33.1$ K) sample. The change of the polarization shape with temperature indicates a freezing process, and the data can be divided into three temperature regions. In region I, given by $T \gtrsim 8$ K, the muon relaxes according to the well-known Kubo-Toyabe (KT) function,

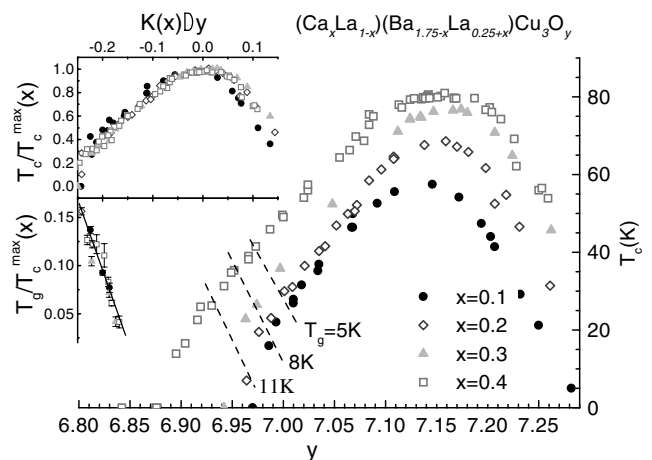


FIG. 1. Phase diagram of $(\text{Ca}_x\text{La}_{1-x})(\text{Ba}_{1.75-x}\text{La}_{0.25+x})\text{Cu}_3\text{O}_y$. The dashed lines indicate samples with equal T_g . Insets: T_c/T_c^{\max} and T_g/T_g^{\max} as a function of $K(x)\Delta y$, where $\Delta y = y - 7.15$, and $K(x)$ is chosen so that all T_c/T_c^{\max} data sets collapse to a single curve.

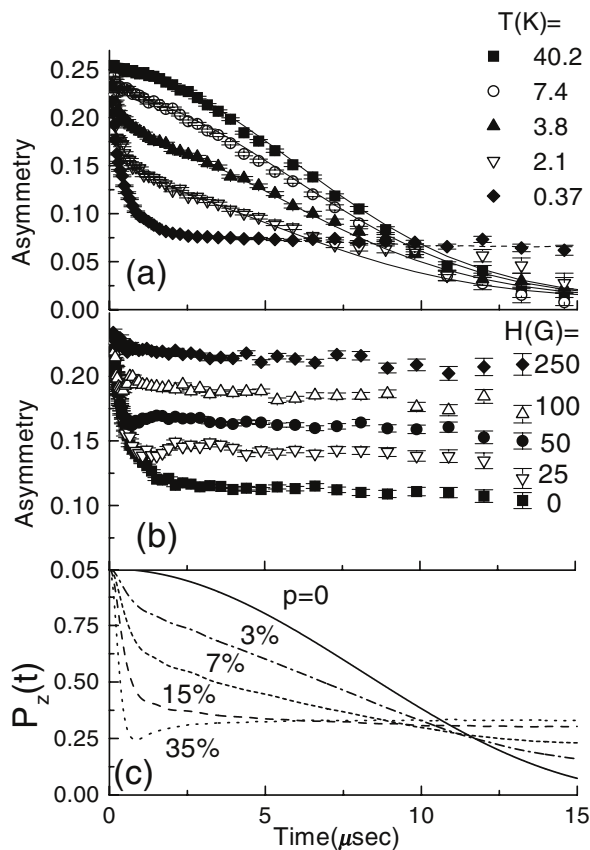


FIG. 2. (a) ZF μ SR spectra obtained in a $x = 0.1$, $y = 7.012$ sample. The solid lines are fit to the data using Eq. (1), the dashed line is a fit using the simulation as described in the text. (b) μ SR spectra obtained in longitudinal fields from the $x = 0.4$, $y = 6.984$ sample at 350 mK. (c) Polarization curves generated by the simulation program as described in the text.

typical of the case where only frozen nuclear moments are present [10]. In region II, bounded by $8 \geq T \geq 3$ K, part of the polarization relaxes fast and the rest as in the first region. As the temperature is lowered the fast portion increases at the expense of the slow portion. Moreover, the relaxation rate in the fast portion seems independent of temperature. Finally, at long time the asymmetry relaxes to zero. In region III, where $3 \text{ K} \geq T$, the asymmetry at long times no longer relaxes to zero, but instead recovers to a finite value. This value is $\approx 1/3$ of the initial asymmetry $A_z(0)$.

To demonstrate that the internal field is static at base temperature, the muon polarization was measured with an external field applied parallel to the initial muon spin polarization. This geometry allows one to distinguish between dynamic and static internal fields. In the dynamic case the asymmetry is field independent [11]. In contrast, in the static case the total field experienced by the muon is a vector sum of H and the internal fields, which are of order $\langle B^2 \rangle^{1/2}$. For $H \gg \langle B^2 \rangle^{1/2}$ the total field is nearly parallel to the polarization. Therefore, in the static case, as H increases, the depolarization decreases, and the asymmetry recovers to its initial value. Because we are dealing with

a superconductor this field sweep was done in field-cool conditions. Every time the field was changed the sample was warmed up above T_c and cooled down in a new field. The results are shown in Fig. 2(b). At an external field of 250 G, the total asymmetry is nearly recovered. Considering the fact that the internal field is smaller than the external field due to the Meissner effect, this recovery indicates that the internal field is static and of the order of tens of Gauss.

We divide the data analysis into two parts: high temperatures (region II) and base temperature. We now discuss region II. Here we focus on the determination of T_g . For that purpose we fit a combination of a fast relaxing function and a KT function to the data [12],

$$A_z(t) = A_m \exp(-\sqrt{\lambda}t) + A_n KT(t), \quad (1)$$

where A_m denotes the amplitude of the magnetic part, λ is the relaxation rate of the magnetic part, and A_n is the amplitude of the nuclear part. The relaxation rate of the KT part was determined at high temperatures and is assumed to be temperature independent. The sum $A_m + A_n$ is constrained to be equal to the total initial asymmetry at high temperatures. The relaxation rate λ is common to all temperatures. The solid lines in Fig. 2 are the fits to the data using Eq. (1).

The success of this fit indicates the simultaneous presence of two phases in the sample; part of the muons probe the magnetic phase while others probe only nuclear moments. As the temperature decreases, A_m (which is presented in the inset of Fig. 3) grows at the expense of A_n . At low temperatures A_m saturates to the full muon asymmetry. A similar temperature dependence of A_m is found in all samples. The origin of the magnetic phase is electronic moments that slow down and freeze in a random orientation. The fact that λ is temperature independent means that, in the magnetic phase, $\gamma_\mu \langle B^2 \rangle^{1/2}$ (where γ_μ is the muon gyromagnetic ratio) is temperature independent. In other words, as the temperature is lowered, more and more parts of the sample become magnetic, but the moments in these parts saturate upon freezing.

Our criterion for T_g is the temperature at which A_m is half of the total muon polarization as demonstrated by the vertical line in the inset of Fig. 3. The phase diagram that is shown in Fig. 3 represents T_g for various samples differing in Ca and O contents. As expected, for constant x , higher doping gives lower T_g .

We have singled out three groups of samples with a common $T_g = 11, 8, \text{ and } 5$ K, as shown in Fig. 3 by the horizontal solid lines. These samples are represented in the phase diagram in Fig. 1 by the dashed lines. The phase diagram, containing both T_g and T_c , is the first main finding of this work. It provides clear evidence of the important role of the magnetic interactions in high temperature superconductivity. In fact, this phase diagram is consistent with recent theories [13] of hole pair boson motion in an antiferromagnetic background. Those theories

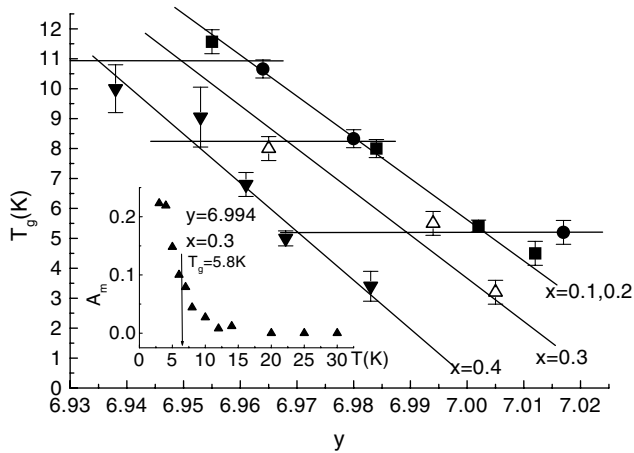


FIG. 3. T_g vs y . The horizontal solid lines are the equal T_g lines appearing in Fig. 1. Inset: magnetic amplitude as a function of temperature for a $x = 0.3$, $y = 6.994$ sample. The arrow indicates T_g of that sample.

conclude that $T_c \propto Jn_s$ where n_s is the superconducting carrier density and J is the antiferromagnetic (AF) coupling energy [14]. From the measurements at constant x we see that $T_g \propto Jf(n_s)$, where f is some decreasing function of n_s . We assume that $n_s = n_s[K(x)\Delta y]$, where $\Delta y = y - 7.15$ is chemical doping measured from optimum and K is a scaling parameter which relates chemical to mobile-charge doping. Since $T_c^{\max} \propto Jn_s$ (optimum), both T_c/T_c^{\max} and T_g/T_c^{\max} should be functions only of $K(x)\Delta y$. We find $K(x)$ by making all T_c/T_c^{\max} collapse onto one curve. This is demonstrated in the upper inset of Fig. 1. Using these values of $K(x)$ we also plot T_g/T_c^{\max} as a function of $K(x)\Delta y$ in the lower inset of Fig. 1. Again all data sets collapse onto a single curve. This indicates that the same single energy scale J controls both the superconducting and magnetic transitions.

We now discuss the muon depolarization at base temperature. In this case all the muons experience only a static magnetic field, as proven above. This allows one to reconstruct the internal field distribution out of the polarization curve. The polarization of a muon spin experiencing a unique field \mathbf{B} is given by $P_z(t) = \cos^2(\theta) + \sin^2(\theta)\cos(\gamma|\mathbf{B}|t)$, where θ is the angle between the field and the initial spin direction. When there is an isotropic distribution of fields, a 3D powder averaging leads to

$$P_z(t) = \frac{1}{3} + \frac{2}{3} \int_0^\infty \rho(|B|) \cos(\gamma|B|t) B^2 dB, \quad (2)$$

where $\rho(|B|)$ is the distribution of $|\mathbf{B}|$. Therefore, the polarization is given by the Fourier transform of $\rho(|B|)B^2$ and has a 1/3 base line. When the distribution of \mathbf{B} is centered around zero field, $\rho(|B|)B^2$ is a function with a peak at $\langle B \rangle$ and a width Δ , and both these numbers are of the same order of magnitude [e.g., Fig. 4(b)]. Therefore we expect the polarization to have a damped oscillation and to recover to 1/3, a phenomenon known as the dip [e.g., the inset in Fig. 4(b)]. Gaussian, Lorentzian, and even exponential random field distribution [15], and, more importantly, all known canonical spin glasses, produce polarization curves

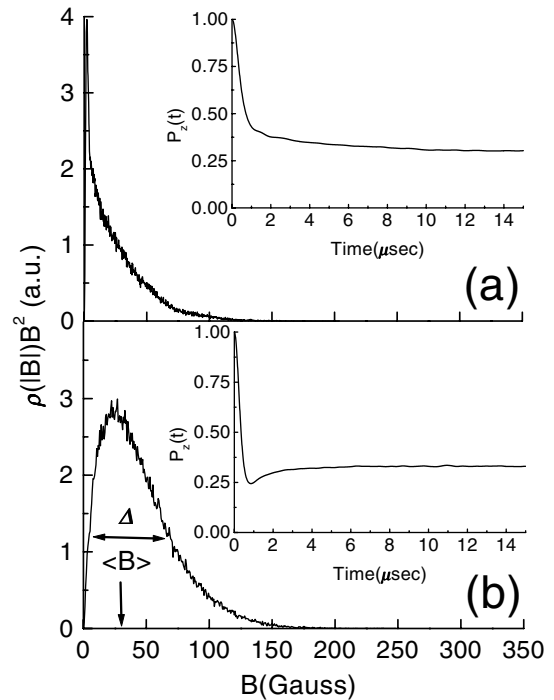


FIG. 4. (a) The internal field distribution extracted from the simulations for the case of correlation length $\xi = 3$ lattice constants, maximum moment size of $0.06\mu_B$ and magnetic moment concentration $p = 15\%$. Inset: the muon spin polarization for that distribution. (b) The same as (a) for the case of $p = 35\%$.

that have a dip before the 1/3 recovery. Furthermore, a dipless polarization curve that saturates to 1/3 cannot be explained using dynamical arguments. Therefore, the most outstanding feature of the muon polarization curve at base temperature is the fact that no dip is present, although there is a 1/3 tail. This behavior was found in all of our samples with $T_c > 7$ K, and also in Ca doped YBCO [16] and Li doped YBCO [17].

The lack of the dip in $P_z(t)$ can tell much about the internal field distribution. It means that $\langle B \rangle$ is much smaller than Δ . In that case the oscillations will be overdamped and the polarization dipless. In Fig. 4 we show, in addition to the $\langle B \rangle \approx \Delta$ case described above [panel (b)], a field distribution that peaks at about zero [panel (a)]. Here $\langle B \rangle$ is smaller than Δ , and, indeed, the associated polarization in the inset is dipless. Thus in order to fit the base temperature polarization curve we should look for $\rho(|B|)B^2$ with most of its weight at about zero field. This means that $\rho(|B|)$ diverges like $1/B^2$ at $|B| \rightarrow 0$, namely, there is an abnormally high number of low field sites.

It also means that the phase separation is not a macroscopic one. If it were, all muons in the field-free part would probe only the nuclear moment and their polarization curve should have a dip or at least its beginning as in the high temperature data. The same would apply for the total polarization curve, in contrast to observation. Thus, the superconducting and magnetic regions are intercalated on a microscopic scale (~ 20 Å) [18]. This is the second main finding of this work.

The special internal field distribution, and the nature of the gradual freezing of the spins, can be explained by the intrinsic inhomogeneity of hole concentration. The part of the sample that is hole poor, and for that reason is “more” antiferromagnetic, will freeze, while the part which is hole rich will not freeze at all. The variation in the freezing temperature of different parts of the sample can be explained by the distribution of sizes and hole concentration in these antiferromagnetic islands [19]. The large number of low field sites is a result of the fact that the magnetic field generated in the magnetic regions will penetrate into the hole rich regions but not completely.

To improve our understanding of the muon polarization, we performed simulations of a toy model aimed at reproducing the results described above. A 2D 100×100 square lattice is filled with two types of moments, nuclear and electronic. All the nuclear moments are of the same size, they are frozen, and they point in random directions. Out of the electronic moments only a small fraction p is assumed to be frozen; they represent magnetic regions with uncompensated antiferromagnetic interactions. Since these regions may vary in size the moments representing them are random, up to a maximum size. The frozen electronic moments induce spin polarization in the other electronic moments surrounding them. Following the work of others [20], we use decaying staggered spin susceptibility which we take to be exponential, namely,

$$\chi'(\mathbf{r}) = (-1)^{n_x+n_y} \exp(-r/\xi), \quad (3)$$

where $\mathbf{r} = n_x a \hat{x} + n_y a \hat{y}$ represents the position of the neighbor Cu sites, \mathbf{a} is the lattice vector, and ξ is the characteristic length scale. Because of this decay, at low frozen spin concentration, large parts of the lattice are practically field-free (except for nuclear moments). However, the important point is that no clear distinction between magnetic and field-free (superconducting) regions exists.

The muon polarization time evolution in this type of field distribution is numerically simulated. The interaction between the muon and all the other moments is taken to be dipolar, and ξ is taken to be three lattice constants [1,2]. The dashed line in Fig. 2(a) is a fit to the $T = 350$ mK data, which yield $p = 15\%$ and maximum moment size $\approx 0.06\mu_B$. As can be seen, the line fits the data very well. However, as expected, the fit is sensitive to $p\xi^2$ only, namely, the effective area of the magnetic islands, so longer ξ would have given smaller p . The field distributions and the polarization curve shown in Fig. 4 were actually generated using the simulation. In (a) the spin density is 15% while in (b) the density is 35%.

In panel (c) of Fig. 2 we show the spin polarization for different hole concentrations, varying from 0% to 35% with the same $\xi = 3$. The resemblance between the simulation results and the muon polarization as a function of temperature in panel (a) leads us to our third conclusion that the freezing process is mostly a growth in the total area of the frozen AF islands.

We are now in a position to address the questions presented in the first paragraph. The appearance of spontaneous magnetic field in CLBLCO is a gradual process. As the temperature is lowered microscopic regions of frozen moments appear in the samples, and their area increases but the moments do not. In the ground state the field profile is very different from that of a canonical spin glass or any other standard magnet. It could only be generated by microscopic intercalation of an abnormal number of zero field regions with magnetic regions without a clear distinction between the two. Finally, and most importantly, the phase diagram containing both T_c and T_g leads us to believe that these temperatures are determined by the same energy scale given by J .

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