Critical Behaviour in the Spin Fluctuations and Superfluid Density of La$_{2-x}$Sr$_x$CuO$_4$

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We study the doping dependence of low frequency spin fluctuations and the zero-temperature superfluid density of La$_{2-x}$Sr$_x$CuO$_4$ using the muon spin relaxation ($\mu$SR) and ac-susceptibility techniques. Superconductivity is found to coexist with low frequency spin fluctuations over a large region of the superconducting phase diagram. The characteristic temperature of spin fluctuations detected by $\mu$SR decreases with increasing $x$ and vanishes above a critical doping $x_c \sim 0.19$. This value of $x_c$ coincides with the doping at which the normal state pseudogap extrapolates to zero. The superfluid density behaves in the opposite way to the low frequency spin fluctuations. It increases with $x$ and becomes nearly doping-independent for $x > x_c$. These results are consistent with predictions involving quantum criticality at $x_c$.

The importance of quantum criticality and the interplay between magnetism and superconductivity in elucidating the pairing mechanism in high-$T_c$ superconductors (HTS) have been discussed in many theoretical and experimental works since their discovery [1–3]. Spin fluctuations and quantum criticality have been extensively studied in heavy fermion compounds, where the appearance of superconductivity as the Neel temperature is suppressed and the superfluid density becomes nearly doping independent as expected for conventional BCS superconductors. The correlation between these two quantities provides evidence for the presence of a quantum critical point, in support of theoretical predictions made by several groups [1,2].

Zero-field $\mu$SR is a sensitive modern technique for studying low frequency spin dynamics. It is a local probe and can detect very small (1G) internal magnetic fields [21]. The data obtained by $\mu$SR give a direct measure of the low energy spin dynamics in the sample [21]. From the time dependence of the depolarisation of muons, one can obtain important information about the characteristic energy or temperature scale of spin fluctuations and the spin-glass transition temperature.

La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) is one of the simplest HTS systems. It has a simple crystal structure and its carrier concentration can be accurately controlled. Undoped La$_2$CuO$_4$ is an insulator with long range antiferromagnetic order. Substitution of Sr$^{2+}$ for La$^{3+}$ introduces holes into the CuO$_2$ planes and at $x \simeq 0.02$ the system exhibits a short range ordered, antiferromagnetic or spin glass phase [1,13]. Superconductivity emerges near $x = 0.05$ and the superconducting transition temperature approximately follows a parabolic doping dependence and vanishes at $x \sim 0.30$ [22].

The samples we studied were high quality polycrystalline La$_{2-x}$Sr$_x$CuO$_4$ ($x = 0.03 - 0.24$) synthesised in Cambridge. The phase purity of these samples, characterised by powder x-ray diffraction and micro-Raman spectroscopy [23], as well as extensive transport [24] and thermodynamic measurements [25,26] was found to be better than 1%. Their lattice parameters and $T_c$ values were also in good agreement with published data obtained on powders as well as single crystals [27,14].

Zero-field (ZF) and transverse-field (TF) $\mu$SR experiments were performed at the pulsed muon source, ISIS Facility, Rutherford Appleton Laboratory and spectra
were collected over the temperature range from 40 mK to 150 K. In a μSR experiment, 100% spin polarised positive muons are implanted into a specimen and evolve in their local magnetic environment. The muon decays with a life time 2.2 μs, emitting a positron preferentially in the direction of the muon spin at the time of decay. By accumulating time histograms of such positrons one may deduce the muon depolarisation rate as a function of time after implantation. The pulsed muon facility at ISIS allows the muon polarisation function to be measured for up to 6-7 muon life times, and thus small changes in the depolarisation function can be detected. This property of the ISIS muon facility has allowed us to study the temperature dependence of slow spin fluctuations in a HTS. The muon is expected to reside in the most electronegative site of the lattice. In the case of LSCO this is 1.0 Å away from the apical O²⁻ site of the CuO₂ plane [28]. As will be discussed later, this assignment is supported by the strong enhancement in the muon depolarisation rate with Zn doping since Zn is known to substitute in the CuO₂ planes. We therefore believe that the results reported here are dominated by the magnetic correlations in the CuO₂ planes.

The zero temperature superfluid density, ρs(0), was determined from measurements of the magnetic penetration depth λab for current flow in the CuO₂ planes, (ρs∼λ⁻²). λab was obtained for all samples using the low-field ac-susceptibility technique (typically at 1 GHz and 333 Hz) for grain-aligned powders [29]. Details of the technique can be found in refs [29,31]. For each doping concentration of LSCO, the result presented here is typical of those obtained for 2-4 samples prepared independently by aligning powders from the same polycrystalline pellet. Four pieces were cut out from each aligned sample and measured. In other words 8-16 samples were investigated for each doping content. The values of λ⁻²(0) for samples with x≥0.15 were also confirmed by standard TF-cooled μSR experiments performed on unaligned powders at 400 Gauss. In a TF-cooled μSR experiment, the field distribution of a flux-line lattice produced by an external field is probed by muons. The depolarisation rate of the initial muon spin is proportional to λ⁻²(0) [29,31]. For x<0.15, λ⁻²(0) obtained from TF-μSR is less accurate than that obtained from the ac-susceptibility measurements because of a strong enhancement in the muon depolarisation rate below 0.3 Tc [31] probably due to the rapid increase in slow spin fluctuations with underdoping as discussed below.

Figure 1 shows the typical time dependence of the ZF muon asymmetry for x=0.08 (Tc=21 K) for two characteristic temperatures. In all samples the high temperature form of the depolarisation is Gaussian, consistent with dipolar interactions between the muons and their near neighbour nuclear moments. This was verified by applying a 50 G longitudinal field, which completely suppressed the depolarisation. The onset of dynamical relaxation processes becomes apparent at low temperatures by a deviation of the depolarisation from a Gaussian behaviour.

The depolarisation rate can be well described by a stretched exponential function G(t) = A₁ exp(-at) + A₂, where γ is the dynamic muon spin depolarisation rate and A₂ accounts for a small time independent back-

![FIG. 1. Typical zero-field μSR spectra of La₂₋ₓSrₓCuO₄ for x=0.08 measured at (a) 1.3 K and (b) 9 K. The solid line in (b) is a fit to a stretched exponential, discussed in the text. In (a) the solid line is a fit to a Lorentzian, describing the initial rapid drop of asymmetry, plus a stretched exponential, describing the long-time tail.](image)

![FIG. 2. Temperature dependence of the exponent β of La₂₋ₓSrₓCuO₄ for x=0.06, 0.08, 0.125, 0.15. The left hand panel shows linear plots with the arrows indicating Tc whereas in the right hand panel we show semi-log plots with the arrows indicating Tg. The error in β is ±0.06.](image)
ground arising from muons stopping in the silver backing plate. As can be seen in Fig. 2 the exponent $\beta$ starts from the value 2 at high temperatures, corresponding to Gaussian relaxation, but decreases steadily at lower temperatures, suggesting the development of a low frequency component in the spectral weight of the spin dynamics. We take the temperature at which $\beta$ first drops below the value 2 as one indicator of the energy scale of the magnetic correlations in a given sample. This temperature is denoted $T_f$. At low temperatures (Fig. 2) the values of $\beta$ decrease towards the value 0.5. At lower temperatures still the form of the relaxation function changes (Fig. 1a): there is an initial rapid drop of asymmetry, followed by a long-time tail with a slower relaxation. This is very characteristic of the behaviour found in spin glass samples below $T_g$ [2], where the initial rapid drop is ascribed to the static distribution of random local fields while the long-time behaviour results from dynamical processes. In this regime the data were fitted to the form $G_z(t) = A_1e^{-(\gamma_1 t)} + A_2e^{-(\gamma_2 t)^2} + A_3$. We identify the spin glass temperature $T_g$ as the temperature at which the value of $\beta$ (Fig. 2) reaches the value 0.5. This "root exponential" form for the relaxation function is a common feature of spin glasses [3,4], and in the present samples this temperature coincided with the cross-over in behaviour (as shown in Fig. 1) and with a peak in the longitudinal relaxation rate. All the samples with $x = 0.03 - 0.125$ followed the same behaviour.

Our data indicate that the spin glass phase persists beyond $x = 0.125$. In fact the onset of the spin glass phase for $x = 0.125$ occurs at a higher temperature than that for $x = 0.10$. This may be due to the formation of strongly correlated antiferromagnetic stripe domains in this range of doping [11,13,19]. For $x = 0.15$ and 0.17, $T_g$ becomes very small (< 45 mK) and $T_f$ is approximately equal to 8K and 2K, respectively. For $x \geq 0.2$, there are no changes in the depolarisation function to the lowest temperature measured (40 mK). The presence of a finite $T_g$ for $x > 0.125$ is verified by the Zn doping studies discussed below.

Figure 3 shows the doping dependences of $T_g$ and $T_f$ together with $T_c$. Although the freezing of spins occurs at very low temperatures, low frequency spin fluctuations appear at significantly higher temperatures. Both $T_g$ and $T_f$ are found to decrease with increasing doping and tend to zero at $x_c \simeq 0.19$. Their behaviour resembles that of the normal state gap [19,26] and the magnetic effects determined from measurements above $T_c$, which also tends to vanish at the same $x_c$. This is better demonstrated in the semi-log plot of $T_g$ and $T_f$ shown in the inset of the figure. The similarity in the doping dependence of $T_g$ and $T_f$ suggests that both quantities may have the same scaling behaviour. The exponential doping dependence of $T_g$ has also been found in $Y_{1-x}Ca_xBa_2Cu_3O_{6+\delta}$ for doping of up to 0.09 holes per planar Cu atom [15]. This suggests that the trend of $T_g$ shown in Fig. 3 is common to all high-$T_c$ materials.

The present $\mu$SR data strongly suggest that there is a certain carrier concentration at which two magnetic energy scales associated with the superconducting state vanish. Although they are both much smaller than the normal state gap, there is a similarity in that they all decrease with doping and extrapolate to zero at $x_c \simeq 0.19$ suggestive of a quantum critical point [3].

To further understand the physical properties near $x_c$ we have performed detail measurements of the absolute values of the in-plane penetration depth for samples prepared from the same batches as those measured by ZF-$\mu$SR. The lower panel of Fig. 3 shows the doping dependence of $\lambda_{ab}^{-2}(0)$. In contrast to $T_f$ and $T_g$, $\lambda_{ab}^{-2}(0)$ increases with doping and becomes nearly doping independent above $x_c$. We note the inverse relationship between $\lambda_{ab}^{-2}(0)$ and the magnetic effects determined by $\mu$SR. The suppression of $\lambda_{ab}^{-2}$ in the underdoped region has been reported from the early days of HTS [30]. Recent penetration depth measurements in LSCO and HgBa$_2$CuO$_{4+x}$ showed that not only the in-plane but also c-axis zero temperature superfluid density tend to saturate above $x_c$ [18,20]. The present results allow us to identify a correlation between $\lambda_{ab}^{-2}(0)$ and the magnetic effects determining $T_g$ and $T_f$.

![Figure 3](image-url) FIG. 3. Doping dependence of $T_g$ (squares), $T_f$ (circles), and $T_c$ (dotted circles) of La$_{2-x}$Sr$_x$CuO$_4$. $T_g$ data from Ref. [15] (triangles) are also shown for comparison. The inset is a semi-log plot of $T_g$ (multiplied by 14) and $T_f$ as a function of doping. The lower panel shows the doping dependence of the inverse square of the zero temperature in-plane penetration depth measured by the ac-susceptibility technique.
Distinct changes at $x_c$ are not limited to the properties presented here, but have also been found in many other quantities of HTS. As mentioned already, the superconducting condensation energy and specific heat jump are maximum at $x_c$ and drop quickly with underdoping [21,22] furthermore the temperature dependence of $\lambda_{ab}$ only obeys the $d$-wave weak-coupling BCS formula for $x > x_c$ [19].

As a further check of the validity of $x_c$ in the context of the magnetic scales discussed here we performed ZF- $\mu$SR measurements for La$_{2-x}$Sr$_x$Cu$_{1-y}$Zn$_y$O$_4$ with $x = 0.15, 0.20$, and $y = 0.01, 0.02$. Substitution with Zn slows down the spin fluctuations and greatly enhances the muon depolarisation rate at low temperatures [31,34,35]. A consequence of this, both $T_g$ and $T_f$, are significantly enhanced. For $x(Sr) = 0.15$ and $y = 0.01$ and $0.02$, we find that $T_g = 2.5K$ and $3.5K$ and $T_f = 15K$ and $25K$, respectively. However, for $x = 0.20$ the muon depolarization rate is unchanged by the Zn substitution. This result also suggests that there is indeed a critical point in the slightly overdoped region. Furthermore, the enhancement of the depolarisation rate by Zn doping confirms that it is the spin fluctuations on the CuO$_2$ planes that are probed by $\mu$SR.

In conclusion, our results confirm that spin glass freezing is present in the superconducting state of underdoped LSCO [1][13], but show more clearly that it extends slightly above optimal doping. We have also provided evidence for very slow spin fluctuations in the same doping region. The doping level at which these characteristic features disappear is very close to the special point $x = 0.19$ [20] suggesting a connection between the magnetic energy scales identified in this work, the normal state energy gap and the anomalous properties of underdoped HTS. Our comparison with the measured superfluid density implies that low frequency spin fluctuations compete with superconductivity in the cuprates. The special doping level $x = 0.19$ could therefore mark the point at which these effects disappear and the remaining high frequency magnetic excitations are favourable for superconductivity in the cuprates. The competition between quasi-static magnetic correlations with superconductivity is reminiscent of the way the Neel temperature of heavy fermion compounds goes to zero at a quantum critical point where the magnetic excitations are then most favourable for superconductivity.

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