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Muon spin rotation study of field-induced magnetism in heavily overdoped $La_{2-x}Sr_xCuO_4$

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Abstract

In the huge volume of research on the cuprates, there is very little that investigates the properties of the highly overdoped area of the phase diagram, where superconductivity disappears with increased carrier concentration. In the spirit of further exploring this region, we have performed the first known μSR measurements of heavily overdoped single-crystalline $La_{2-x}Sr_xCuO_4$. We discovered an anomalous field-induced broadening of the internal magnetic field distribution. The broadening is linear in applied field and has a Curie–Weiss temperature dependence. Furthermore, the effect increases with doping above $x = 0.12$ and is maximal at our highest doping ($x = 0.3$). Possible relaxation mechanisms are discussed.

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1. Introduction

In the 20 years since the discovery of high-temperature superconductivity, it has become conventional wisdom that the heavily overdoped cuprates are entirely described by canonical Fermi liquid theory. An unfortunate corollary of this wisdom is that the overdoped region of the phase diagram is well understood, and interesting behaviour in the cuprates is confined to optimally or underdoped samples.

In actuality, there is very little experimental data on heavily overdoped samples, and much of the existing data suggests *unconventional* behaviour. Resistivity measurements consistently give power laws which are less than 2 [1–3]. Hall constants are distinctly temperature dependent at all known dopings and temperatures [1,4]. Optical conductivity measurements show unusual frequency dependence [2]. All of these results contradict Fermi liquid theory.

Magnetic response is also unusual. Magnetisation data for $La_{2-x}Sr_xCuO_4$ (LSCO) has long been shown to fall on a universal curve for all dopings [5]. Yet, recent measurements on pure single crystals of overdoped LSCO show that, for $x \geqslant 0.22$, susceptibility acquires an additional Curie term [6]. A similar Curie term has also been observed in $Tl_2Ba_2CuO_{6+\delta}$ [3]. This is again in contrast to Fermi liquid theory, where one would expect temperature-independent Pauli paramagnetism.

These irregularities have led to the term "Anomalous Fermi Liquid" [4]—a name which reflects our lack of understanding of this region of the phase diagram. Part of the problem is the deficiency of high-quality heavily overdoped crystals that has plagued the community for many years. However, this situation has been somewhat rectified recently with the advancement of crystal growth techniques. One excellent example is the set of heavily overdoped samples of LSCO studied by Wakimoto et al.

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with neutron scattering [7], wherein they drew interesting parallels between superconductivity and incommensurate magnetism.

Using these same samples, we have performed what is to our knowledge the first single-crystalline μSR study of the cuprates in the heavily overdoped regime.

2. Experimental results: TF-μSR

Single crystal samples of $La_{2-x}Sr_xCuO_4$ with $x = 0.25$ ($T_c \approx 14\,K$) and $x = 0.30$ ($T_c \approx 0\,K$) were grown at the University of Toronto with the travelling solvent floating zone technique and cut into $\sim 6\,mm \times 6\,mm$ crystals with the large face perpendicular to the c-axis. We performed transverse field μSR (TF-μSR) experiments on the HI-TIME spectrometer at TRIUMF in Vancouver, B.C. Fields were chosen between 2 kG and 5 T and oriented along the c-axis. Temperatures were varied between 2 and 180 K.

At high temperatures, experimental asymmetries in both samples are consistent with inhomogeneous relaxation from randomly oriented nuclear dipolar moments. Decay envelopes are Gaussian in shape with weak relaxation that is largely independent of temperature and applied field. As one lowers the temperature, however, relaxation increases monotonically, reaching values as high as $2.2\,\mu s^{-1}$ at 2 K. Envelope shapes also change dramatically, from Gaussian decay at high temperatures to root exponential decay at the lowest temperatures.

Spectra were fit assuming a temperature- and field-independent Gaussian relaxation with an additional exponential relaxation that was allowed to vary. Fig. 1a shows the exponential relaxation rates in $La_{0.7}Sr_{0.3}CuO_4$ for a variety of fields and temperatures. The temperature dependence of the relaxations at a given field are well-described by a Curie–Weiss (C–W) function for the entire temperature range. The scaling of the relaxation, as given by the Curie constant (Fig. 1b), is linear in the applied magnetic field and passes through the origin. This indicates that the anomalous relaxation is a *field-induced* effect. The central role of applied field is further revealed when one notes that the Weiss constant (Fig. 1c) is also roughly proportional to field.

Our measurements on $La_{1.75}Sr_{0.25}CuO_4$ also show an anomalous increase in relaxation with decreasing temperature. However, deviation from the C–W temperature dependence is seen at temperatures below T_c, where superconductivity is a complicating factor. Additional data are provided by Savici et al. [8], whose previous work has shown that relaxation rates in $La_{1.88}Sr_{0.12}CuO_4$ follow the

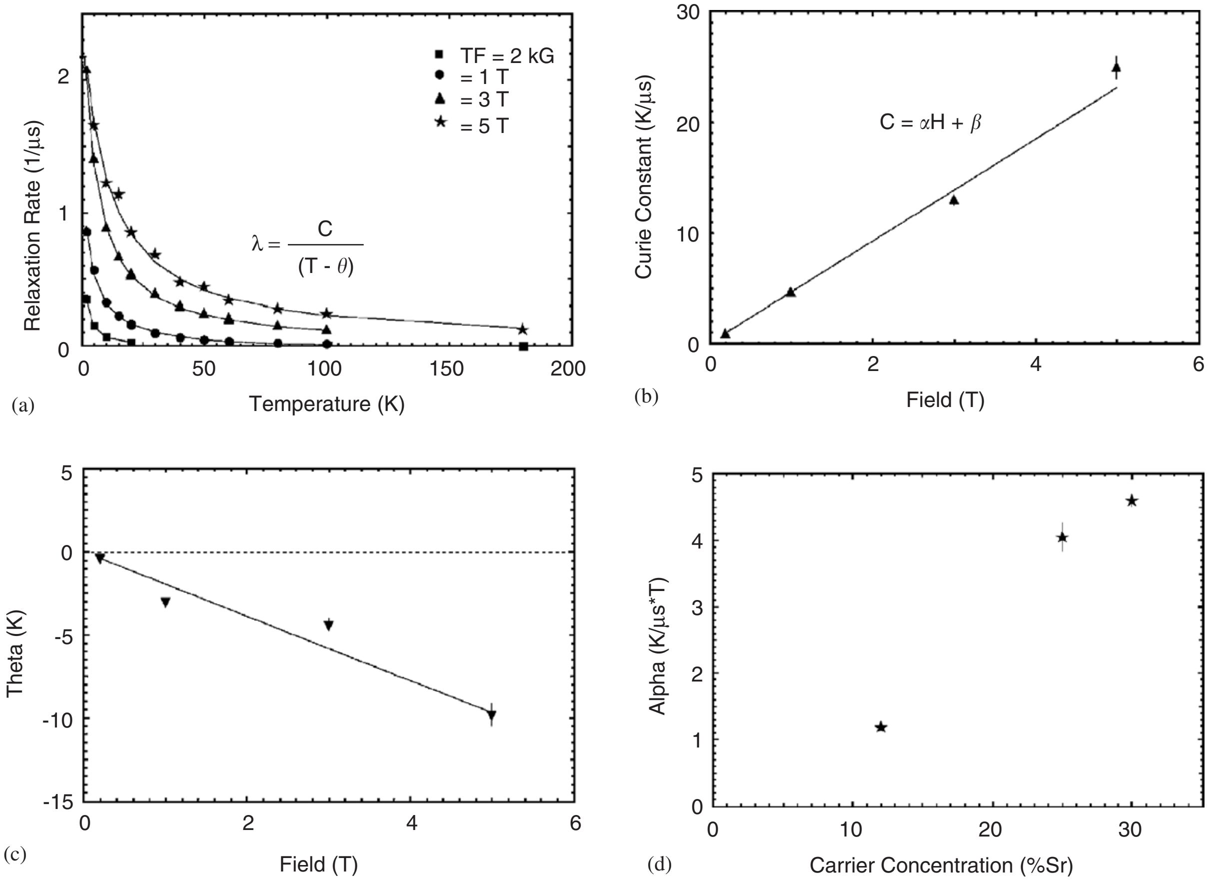

Fig. 1. (a) Relaxation rates for the 30% Sr sample at various fields. Solid lines represent fits to a Curie–Weiss function. (b) Curie constant as a function of field. (c) Weiss constant as a function of field. (d) Overall scaling of the anomalous relaxation as a function of Sr content.

same phenomenology for temperatures above the superconducting and magnetic transition temperatures. For the purpose of comparison with the 30% Sr sample, we will concentrate only on the high-temperature C–W behaviour in this paper.

Fig. 1d displays the slope of the Curie constant versus magnetic field line, α, for each sample as a function of Sr content. One can think of these values as the overall scaling of the relaxation phenomenon, for which temperature and field dependences have been taken into account. The scaling seems to vary smoothly with carrier concentration, and is largest for our largest doping. Notably, though, it does not scale with Sr content, as one might expect if the broadening were merely an impurity effect associated with cation substitution.

3. Discussion

Among the most surprising features of the present phenomenon is its ubiquity. The observed broadening, easy to identify with its hallmark temperature and field dependences, is observed in a large range of materials with remarkably different properties. The 12%, 25% and 30% Sr samples lie in distinct parts of the cuprate phase diagram and have different magnetic, superconducting and structural properties. Measurements by Savici on $La_{1.75}Sr_{0.15}Eu_{0.1}CuO_4$ and $La_{1.875}Ba_{0.125}CuO_4$, known stripe materials, reveal a similar effect [8].

In light of this, it seems strange that Savici saw almost no field-induced relaxation in crystals of $La_{1.85}Sr_{0.15}CuO_4$ and $La_{1.81}Sr_{0.19}CuO_4$. Illumination may come from Haase et al., who reported similar dependences of the ^{17}O linewidth in NMR experiments on $La_{1.85}Sr_{0.15}CuO_4$ [9], but at notably higher temperatures and fields. This may demonstrate that the present effect is stronger in samples with suppressed superconductivity and is removed to higher energies when superconductivity is robust. Haase also reports having seen similar results in $YBa_2Cu_3O_{7-\delta}$, creating the tantalizing possibility that this is not simply a phenomenon associated with doped La_2CuO_4 systems, but is a more general feature of the cuprates. Faced with such a possibility, it is desirable to find an explanation for this anomalous relaxation.

One can immediately eliminate any relaxation mechanism associated with superconductivity. The temperatures presented in this study extend well above the superconducting transitions of the respective materials. Moreover, the anomalous relaxation is actually strongest in the 30% Sr sample, despite the fact that the sample is non-superconducting. Lastly, the superconductivity in the 25% Sr sample seems to cause deviation from the C–W temperature dependence and not support it.

Further investigation puts more limits on possible relaxation mechanisms. Fig. 2 shows plot of LF-μSR relaxation rates as compared to the TF-μSR rates in the 25% Sr sample at 5 T. LF spectra show rates which are statistically indistinguishable from zero, even for temperatures where the TF-μSR has sizeable relaxation. The situation is similar in every observed field. As LF-μSR asymmetries should be a factor of 2 more susceptible to dynamic processes than TF-μSR ones [10], this observation allows us to state that the anomalous relaxation in these materials is entirely due to a static broadening of the internal magnetic field distribution.

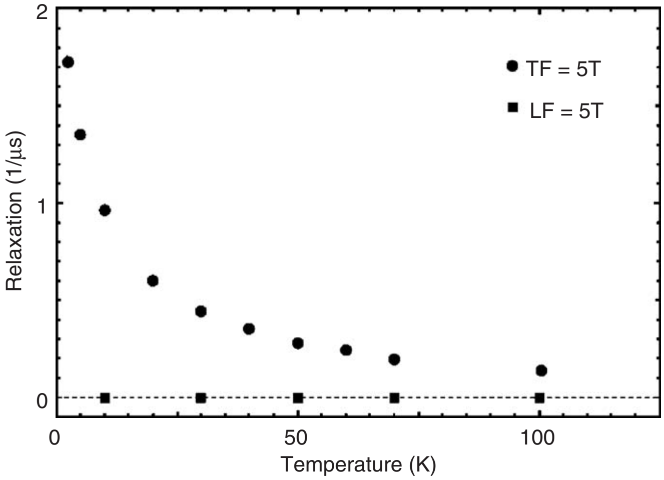

Fig. 2. Comparison of relaxations for the TF and LF geometries.

Singer et al. have put forth an electronic inhomogeneity model to explain many of their results in NMR experiments on LSCO [11]. In this model, charge carriers are distributed randomly through the sample when doped, but are still inhomogeneous on a microscopic level. Properties at a given spin site are then determined by the local charge density, creating a natural distribution. The ability of such a model to explain line broadening depends critically on the large variation of Knight shift with doping. The Knight shifts seen in μSR, however, are orders of magnitude smaller than the observed broadening, making such a picture unlikely.

Examination of zero-field spectra shows that the overdoped materials have no tendency to order in the absence of applied field. There is also no sign of line broadening as temperature is lowered, consistent with our previous observation that the anomalous relaxation is a field-induced effect. Lack of order in zero field, however, does not rule out magnetism as a relaxation mechanism. It is quite possible that the presence of an applied magnetic field lowers the free energy of a magnetic ground state, making it more favourable as field is increased. As such, we have made several attempts to model the observed lineshapes by superimposing internal order on the externally applied field. Conventional anti-ferromagnetism and sinusoidal spin density waves (SDW) were ruled out as possibilities because they tend to create bimodal field distributions, inconsistent with what is observed. Ferromagnetism was similarly eliminated due to the lack of observed Knight shift.

It is important to note, however, that this does not eliminate less conventional forms of magnetism with different field distributions. We have had some success modelling our data by assuming a Gaussian distribution of sinusoidal SDW amplitudes. Also possible are 2D SDWs or forms of SDW which are not sinusoidal in nature but have a more sharply peaked profile. This work is ongoing.

In conclusion, we have performed the first known μSR measurements on heavily overdoped LSCO single crystals. We have observed an anomalously large relaxation with a C–W temperature dependence and a linear field dependence. Such line broadening has been confirmed in several samples across the phase diagram. Different relaxation mechanisms have been discussed. A possible explanation lies in a field-induced magnetism with an unconventional distribution of magnetic fields.

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