The interaction between the magnetic and superconducting order parameters in a La$_{1.94}$Sr$_{0.06}$CuO$_4$ wire

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We investigate the coupling between the magnetic and superconducting order parameters in an 8 m long meander line ("wire") made of a La$_{1.94}$Sr$_{0.06}$CuO$_4$ film with a cross section of 0.5 × 100 μm$^2$. The magnetic order parameter is determined using the Low-Energy muon spin relaxation technique. The superconducting order parameter is characterized by transport measurements and modified by high current density. We find that when the superconducting order parameter is suppressed by the current, the magnetic transition temperature, $T_m$, increases. The extracted sign and magnitude of the Ginzburg-Landau coupling constant indicate that the two orders are repulsive, and that our system is located close to the border between first and second order phase transition.

When cuprates are doped their low temperature ordered phase changes from an antiferromagnetic (AFM) to a superconducting (SC) one. The transition takes place over a range of doping levels where, at low enough temperatures, the samples are both superconducting and magnetic. It is natural to expect phase separation due to the inhomogenous doping. However local probe such as muon spin relaxation indicates that the magnetic volume fraction is 100%, namely, the magnetic field exists everywhere, even in the SC regions. Therefore, the nature of the presence of SC and magnetism is unclear. Are the two orders coupled, and if yes, what are the sign and strength of the coupling? What is the order of the transition between the AFM and SC phases as a function of doping? Is it first order with phase separation or second order with coexistence?

Here we answer this question by looking at the effect of current $I$ on the magnetic phase transition temperature, $T_m$. A current, on the scale of the second critical current $I_c$, diminishes the superconducting order parameter. If the two orders interact, the magnetic order parameter is expected to react to the current and either increase or decrease depending on the type of coupling between the two orders. This, in turn, will increase or decrease $T_m$, respectively. Therefore, we map the magnetic phase transition with and without current. We find that, with current, the magnetic phase transition temperature increases. This results implies that the orders are coupled, and that they are repulsive. Analysis based on the Ginzburg-Landau (GL) model shows that the phase transition is close to the border between first and second order.

The experiment is done with an 8 m long wire made of La$_{1.94}$Sr$_{0.06}$CuO$_4$ film. The film is prepared using laser ablation deposition on (100) LaAlO$_3$ substrate, standard photolithographic patterning and wet acid etching (0.05% HCl). The 6% Sr doping was chosen since the corresponding bulk material has a $T_c \approx 10$ K and $T_m \approx 6$ K, which makes both critical temperatures reachable in a standard cryostat. The cross section of the wire is 0.5 μm × 100 μm so that a typical applied current of a few mA is comparable to $I_c$. Probing the magnetic properties of such a thin wire is achieved by using the new low energy muon spin relaxation (LE-μSR) technique. In this technique, the muons are first slowed down in an Ar moderator where their kinetic energy drops from 4 MeV to 15 eV, while their initial full polarization is conserved. They are then electrostatically accelerated to 15 keV and transported in ultra high vacuum (UHV) to the sample. Four counters collect positrons from the asymmetric muon decay. One pair of counters is parallel to the initial muon spin direction and the other pair is perpendicular to it. The muon asymmetry in these directions is calculated by taking the difference over the sum of the count for each pair. This asymmetry is proportional to the component of the muon polarization in each direction. The field the muon experience is either internal, below $T_m$, or external (designated by $H$), or both. For more details on μSR in the presence of superconductivity and magnetism see Refs. The muons beam spot size has a 15 mm diameter (FWHM). In order to avoid muons missing the sample, the wire is folded in the form of a long meandering line covering a disc 3 cm in diameter. The inset of Fig. shows a magnified image of one corner of the sample.

First, we discuss the sample characterization. In order to verify that the wire is indeed a bulk superconductor and that the current flows in the bulk of the wire we performed transverse field LE-μSR measurements in a field of $H = 1$ kG. Figure (a) depicts the results from the magnetic phase ($T = 2.9$ K) in a rotating reference frame, using zero field cooling (ZFC). The muons depolarize very quickly and after 3 μs the remaining decay asymmetry is due to muons that have stopped in the substrate. For comparison, data from a blank substrate, normalized by its effective area, are also shown. We also
present the decay asymmetry in the pure superconducting phase \((T = 6\ \text{K})\) using field cooling (FC) conditions. In this case, the muon polarization is lost exponentially versus time at a rate \(r_{sc}\) due to the magnetic field distribution of the vortices in the superconducting phase. After 6 \(\mu\)s the polarization reaches the level of the substrate and the ZFC run, and thus most of the muons are affected by vortices.

We fit the function:

\[
	ext{Asym}(t) = A_{sc}e^{-(r_{sc}t)^2/2} \cos(\omega_{sc}t) + A_{sb}e^{-(r_{sb}t)^2/2} \cos(\omega_{sb}t) + A_n e^{-(r_nt)^2/2} \cos(\omega_{nt}t)
\]

to the muon decay asymmetry at all temperatures. Here \(A_{sc}, A_{sb},\) and \(A_n\) represent the respective contributions from the part of the meander that turns superconducting upon cooling, the substrate, and the part of the meander that remains normal upon cooling. \(r_{sc}, r_{sb},\) and \(r_n\) are the relaxation rates of muons that land in a superconducting, substrate, and normal material, respectively. \(\omega_{nt} = \omega_{nt}(t)\) is the rotation frequencies in the normal material and the substrate (taken to be equal). \(\omega_{sc}\) is the rotation frequency in the superconducting part. The only parameters that are allowed to vary with \(T\) are \(r_{sc}\) and \(\omega_{sc}\).

The superconducting volume fraction is estimated from \(A_{sc}/(A_{sc} + A_n)\) and was found to be \(90 \pm 5\%\).

Figure (a) shows \(r_{sc}\) and the resistivity versus temperature. The midpoint of the resistivity transition to the superconducting state, and the onset of superconductivity are fitted to the function \(\Theta(T - T_c)\) with \(T_c = 16\ \text{K}\). The London penetration depth \(\lambda_{ab}\) at \(T = 7\ \text{K}\) is 500 nm as estimated from the relation \(r_{sc} = 0.04 \gamma \mu_\phi / \lambda_{ab}^2\) where \(\gamma = 13.5\ \text{MHz/kg}\) is the muon gyromagnetic ratio, and \(\phi_0\) is the magnetic flux quantum. This penetration depth value is similar to the meander thickness and therefore the current will flow uniformly in the bulk of the meandering wire.

It is challenging to flow a current in the meander line during a LE-\(\mu\)SR experiment while keeping its temperature well determined. This results from the fact that the sample is cooled by a cold finger in a UHV ambient. Above the first critical current, \(I_1\), the superconducting wire acts as a heater and is not in thermal equilibrium with either the cold finger or any attached thermometer. Therefore, the wire’s temperature can be measured only by an \textit{a priori} calibration procedure. For this, we chose to take the V-I curve of the wire at each temperature in a flow cryostat. In such a cryostat the thermal contact between the wire and a thermometer, even at high currents, is good. Using this calibration, the wire acts as its own thermometer. To account for possible drifts in the calibration we repeated the calibration in the flow cryostat also after the LE-\(\mu\)SR experiment. This proved the temperature uncertainty to be smaller than 0.01 K, namely, when we say that we are comparing two runs with equal temperatures we mean that we managed to keep the two runs 0.01K away from each other.

Figure (a) shows several V-I curves recorded at different temperatures on a short segment (1 cm long) of the wire. These V-I curves are used for the determination of \(I_{c1}\) and \(I_{c2}\) which are needed for the analysis. The curves are fitted to the function \(\Theta(T - T_c)\ e^{k(I - I_c)}\), where \(\Theta\) is the Heaviside step function. It is seen in Fig. (b) that, at \(T = 12\ \text{K}\), \(I_{c1}\) drops to zero and the 1 cm segment of the wire shows Ohmic behavior with a normal resistance of \(R_n = 60\Omega\). We estimate \(I_{c2}\) using a variation of the offset criterion. The exponential dependence of \(V\) on \(I\) is extrapolated to the value of \(I\) that gives a differential resistance equal to \(R_n\). The obtained values of both critical currents as a function of temperature are plotted in Fig. (b).

Next, we study the effect of the current on the magnetic order. Figure (c) shows raw muon decay asymmetry data from the meander wire at several temperatures with no external field and in the laboratory frame. The open symbols represent measurements at low currents (used only for temperature determination) and the solid symbols are measurements at high currents. At \(T > T_m\), the asymmetry resembles a Gaussian with relatively slow
relaxation, typical of magnetic fields generated by copper nuclear magnetic moments. As the temperature decreases, there is a clear increase in the muon spin depolarization rate indicating that the magnetic order has set in. For comparison, we show in the inset of Fig. 3 standard $\mu$SR measurements taken with a He flow cryostat on the bulk powder used for making the film. In this case the measurements could be extended to $T = 1.65$ K. We find that the magnetic transition in the wire is very similar to that of ours and others bulk samples, having similar $T_m$. In addition, the data in the bulk at low enough temperatures is typical of the case where muons in the full sample volume experience frozen magnetism, with spontaneous precession below about 2 K with a frequency $f \simeq 3$ MHz, again in agreement with others.

The effect of the current is demonstrated by the $T = 5$ K measurement (red symbols in Fig. 3). The depolarization of the muons spin is faster when a higher current is applied. The difference between the two measurements is emphasized by the shaded area. The change in the asymmetry line shape caused by the application of current is equivalent to cooling by about 0.3 K, although, as mentioned before, the sample temperature is stable to within 0.01 K. This effect was observed at several temperatures along the magnetic transition.

Above $T_m$ and below 4 K the application of current has no effect on the asymmetry. This finding is particularly important since, $a$ priori, the current might affect the muon asymmetry directly by means of the magnetic field it produces, or by colliding with the muons. However, we found that once the electronic spins are fully frozen the current does not change the muon asymmetry indicating that there is no direct current muon coupling. This is in agreement with calculations showing that the magnetic field the current produces is very small compared to the internal field. Similarly, the lack of current effect above $T_m$ rules out collisions between muon and electron charge.

In order to determine the magnetic phase transition temperature, without assuming a specific spatial field distribution or temporal fluctuation model, we define the order parameter in a model-free way. At each temper-
The asymmetry as a function of time is averaged to produce \( \langle Asy \rangle = \frac{1}{t_m} \int_0^{t_m} Asy(t)dt \) where the measurement time \( t_m = 8 \ \mu\text{sec} \). We expect \( \langle Asy \rangle \) to decrease with increasing magnetic moment size \( M(T) \), and therefore defined
\[
\frac{M(T)}{M(0)} = \frac{\langle Asy \rangle^{-1}(T) - \langle Asy \rangle^{-1}(\infty)}{\langle Asy \rangle^{-1}(0) - \langle Asy \rangle^{-1}(\infty)} \quad (2)
\]
For \( \langle Asy \rangle(\infty) \) we take the averaged \( Asy \) at \( T = 7.35 \ K \), which is above the transition. The magnetic phase transition temperature \( T_m \) is taken as the onset of the sudden change in \( M(T) \). The magnetic transition is sharp enough that other, model-based, analysis methods gave indistinguishable \( M(T) \). The temperature dependence of \( M \) with and without current is presented in Fig. 4. We find that the application of a current of about \( 0.2-I_{c2}(T) \) increases the magnetic phase transition temperature by \( 0.4 \pm 0.1 \ K \). This effect means that the two orders interact repulsively. It is complementary to the effect of a strong magnetic field on doped samples, where the magnetic order is enhanced while the superconducting order is suppressed\(^{11,12}\). However, since current, in contrast to magnetic field, does not couple directly to spins, the effect presented here is more simply analyzed. For example, it shows that the enhanced magnetism in the applied field could be a result of supercurrent in the bulk\(^{11,12}\), and not necessarily due to magnetism in the vortex core\(^{11,12}\).

A simple interpretation of the result can be given in the framework of the GL model. In this model the free energy density near the critical temperature \( T_m \) can be written as
\[
F = -a(T) \left( 1 - T^2/I^2_{c2} \right) |\psi|^2 + U_{s0}|\psi|^4 - b \left( T_m^2 - T \right) |\phi|^2 + U_m|\phi|^4 + 2U_{sm}|\phi|^2|\psi|^2 \quad (\text{plus gradient terms})
\]
where \( \psi \) and \( \phi = M/\sqrt{\tau_4} \) are the superconducting and magnetic order parameters respectively, \( U_{sm} \) is their coupling constant, \( v \) is the unit cell volume, \( b \) is a dimensionless parameter, \( T_m^2 \) is the magnetic phase transition temperature for \( |\psi|^2 = 0, a(T), U_s \) and \( U_m \) are the standard GL parameters. All the parameters can be experimentally determined\(^{13-16}\): \( a(T) = h^2/2m^* \xi^2 \) where \( \xi = 2 \ \text{nm} \) is the superconducting coherence length\(^ {17,18}\); \( \psi_0 = m^*/4\mu_0e^2\lambda^2 \) where \( \lambda = 500 \ \text{nm} \) is the London penetration depth; \( U_s = a/2\psi_0^2 \) according to the minimum condition; \( bT_m = h^2/2m^* \kappa \) where \( \kappa = 4 \ \text{nm} \) is the magnetic coherence length\(^ {18,19}\); the electron mass can be approximated by the stiffness of the xy model where \( h^2/2mA = J \), \( A \) is the cell area and \( J \approx 10^3 \) K is the superexchange; from the ratio of muon oscillation frequency between our sample and pure \( \text{La}_2\text{CuO}_4 \) we find a local magnetic moment \( M = 0.33\mu_B \) giving \( \phi^2 = 0.33^2/v \); \( U_{sm} = bT_m/2\phi_0^2 \) again by the minimum condition.

\( U_{sm} \) is obtained from our current dependent measurement (neglecting gradient terms at this stage). Since \( T_c \) is higher than \( T_m \) we do not expect \( |\phi|^2 \) to affect \( |\psi|^2 \). Therefore \( |\psi(I, T)|^2 = |\psi(0, T)|^2(1 - I^2/I_{c2}^2) \). The minimization of \( F \) with respect to \( |\phi|^2 \) yields, \( |\phi|^2 = b(T_m^2 - 2U_{sm}|\psi(I, T)|^2/b - T)/2U_m \). The measured magnetic transition temperature is given by \( T_m = T_m^0 - 2U_{sm}|\psi(0, T)|^2/b \). We assume that near \( T_m \), \( |\psi|^2(0, T) = \psi_0^2 \) where \( \psi_0 \) is the ground state value of \( \psi \). Therefore, the change in the transition temperature, \( \delta T_m = T_m - T_m(0) \), caused by the current is \( \delta T_m(I) = 2U_{sm}\psi_0^2 I^2/bI_{c2}^2 \). The interesting parameter is
\[
R \equiv \frac{U_{sm}}{\sqrt{\psi_0 I_{c2}}} = \frac{e\xi^2 M I_{c2}^2 \delta T_m}{\mu_\gamma h\kappa I^2 T_m} \sqrt{\frac{J_{t0}}{h}}
\]
where \( h \) is the unit cell height. For \( R > 1 \) the GL model predicts phase separation and first order phase transition. For \( R < 1 \) the model predicts coexistence and a second order phase transition. The \( R = 1 \) condition is essential for SO(5) symmetry\(^ {20}\). At \( T = 5 \ K \) we found that \( I_{c2} = 17 \ \text{mA} \) (see Fig. 2b) and used \( I = 4 \ \text{mA} \) in the LE-\( \mu \)SR. This yields a positive \( R = 1.4 \). Although numerical factors can change \( R \), they cannot change its proximity to unity.

In summary, we demonstrated the presence of interaction between the magnetic and superconducting order parameters and measured its sign and strength. We find that phase transition at zero temperature from magnetic to superconducting orders, as a consequence of doping, must be very close to the boarder between first and second order.

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