Inhomogeneous Magnetic-Field Response of YBa$_2$Cu$_3$O$_y$ and La$_{2-x}$Sr$_x$CuO$_4$ Persisting Above the Bulk Superconducting Transition

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We report that in YBa$_2$Cu$_3$O$_y$ and La$_{2-x}$Sr$_x$CuO$_4$ there is a spatially inhomogeneous response to magnetic field for temperatures $T$ extending well above the bulk superconducting transition temperature $T_c$. An inhomogeneous magnetic response is observed above $T_c$ even in ortho-II YBa$_2$Cu$_3$O$_6$ [5], which has highly ordered doping. The degree of the field inhomogeneity above $T_c$ tracks the hole doping dependences of both $T_c$ and the density of the superconducting carriers below $T_c$, and therefore is apparently coupled to superconductivity.

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The $T$-$p$-$H$ phase diagram of high-$T_c$ cuprates (i.e., temperature $T$, hole doping $p$, and magnetic field $H$) includes different magnetic phases discovered by the technique of muon spin rotation ($\mu$SR). As a sensitive local probe of static or quasi-static moments that are not necessarily ordered, $\mu$SR experiments have revealed that: (i) static Cu electronic moments remnant of the antiferromagnetic (AF) phase persist across the insulator-superconducting boundary [1, 2]; (ii) some form of weak static magnetism occurs in YBa$_2$Cu$_3$O$_y$ (YBCO) near the pseudogap transition temperature $T^*$ [3]; and (iii) an applied magnetic field induces static magnetism in and around the vortex cores of samples on the low-doping side of what has been loosely dubbed a “metal-to-insulator crossover” (MIC) [4]. Recently, (ii) has been independently verified by polar Kerr effect [5] and polarized neutron diffraction [6, 7] measurements that show the onset of some kind of magnetic order in YBCO near $T^*$. The neutron results indicate that the magnetic order is not AF Cu-spin order, but may be associated with either the circulating-current phase proposed by Varma [8], a ferromagnetic arrangement of Cu spins, or the existence of staggered spins on the oxygen sites of the CuO$_2$ layers. From magnetic-field training of the polar Kerr effect it is concluded in Ref. [5] that the magnetic order is associated with a time-reversal symmetry (TRS) breaking effect that persists above room temperature.

We have used transverse-field muon-spin rotation (TF-$\mu$SR) to measure the local response in YBCO and La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) single crystals to external magnetic fields up to $H = 7$ T. In this kind of experiment the initial muon spin polarization $P(t = 0)$ is oriented transverse to the field, which was applied perpendicular to the CuO$_2$ layers. The intrinsic spin of an implanted muon precesses in the plane perpendicular to the axis of the local magnetic field $B$ with a frequency $f_\mu = \gamma_\mu B$, where $\gamma_\mu = 851.6$ MHz/T is the muon’s gyromagnetic ratio. Spatial field inhomogeneity in the bulk of the sample causes the transverse polarization $P(t)$ to decay with time due to the dephasing of muon spins precessing in different local magnetic fields. Previously, we determined that none of the samples considered here contain Cu moments remnant of the AF phase that fluctuate slowly enough to be detected on the microsecond time scale of $\mu$SR [4]. Furthermore, we showed that an applied magnetic field induces quasi-static magnetism for $T < T_c$, but only in samples below $y \approx 6.55$ for YBCO and $x \approx 0.16$ for LSCO.

The TF-$\mu$SR spectra are the sum of a sample signal and a time-independent background signal, $aP(t) = a_sP_s(t) + a_{bg}$, where $a_{bg}$ is less than 4 % of the total signal amplitude $a$. The sample polarization function can be written as

$$P_s(t) = G(t) \exp(-\Delta^2 t^2) \cos(f_\mu t + \phi), \quad (1)$$

where the Gaussian function $\exp(-\Delta^2 t^2)$ accurately accounts for the random nuclear dipole fields and is temperature independent, and $G(t)$ is a phenomenological function that accounts for additional relaxation of $P_s(t)$ by other sources. The relaxation function $G(t) \exp(-\Delta^2 t^2)$ is the “envelope” of the TF-$\mu$SR signal. Figure 1(a) shows TF-$\mu$SR envelopes for pure Ag, which does not exhibit superconductivity and does not contain electronic magnetic moments. The TF-$\mu$SR signals for Ag are well described by Eq. (1) with a temperature-independent exponential relaxation function $G(t) = \exp(-\Lambda_A g t)$, where $\Lambda_A$ is a measure of the field inhomogeneity of the superconducting magnet used to generate the applied field. Alternatively, in YBCO and LSCO, the formation of a vortex lattice below $T_c$ creates a broad temperature-dependent internal magnetic field distribution $n(B)$. Below $T \approx 0.5 T_c$, the TF-$\mu$SR signals in YBCO and LSCO are well described by Eq. (1) with a stretched-exponential relaxation function $G(t) = \exp[-(\Delta t)^{\beta}]$, where $1.19 \leq \beta \leq$
correspond to data taken for
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for all samples.

Since Λ is a measure of the width of n(B) originating from sources other than nuclear dipoles, the difference Λ−Λ_{Ag} should vanish at T_{c}. Instead we find that a nonzero value of Λ−Λ_{Ag} persists to temperatures well above T_{c} (see Fig. 2). Note that above T≈200 K, the observed Λ in YBCO is reduced by thermally activated hopping of the muon [9]. Hence the temperature at which Λ−Λ_{Ag} vanishes can only be determined by extrapolation. We now discuss possible origins of the anomalous spatial field inhomogeneity above T_{c}.

Remnant Copper Spins.—Previously Savici et al. detected field-induced inhomogeneity above T_{c} in underdoped cuprates, including LSCO x=0.12. In contrast to the current study, quasi-static Cu spins remnant of the AF phase were detected for H=0. Field-induced ordering of these spins in a significant volume of the sample adds a second faster-relaxing exponential component to G(t). This was observed in LSCO x=0.12, but also here in LSCO x=0.145—i.e. on the low-doping side of the MIC near x=0.16. However, in both cases, this second component vanishes well below T_{c}, as does disordered static magnetism induced at lower fields [4].

We note that neutron scattering measurements on LSCO x=0.10 also show field-induced magnetic order only below T_{c} [11], and measurements on LSCO x=0.163 show no static magnetism up to H=14.5 T and dynamic AF correlations only below T_{c} [12].

Figure 3 shows the hole-doping dependence of Λ−Λ_{Ag}. Despite YBCO y=6.67 being close to 1/8 hole doping where spin/charge stripe correlations [13] and a suppression of superconductivity [14] occur, above T_{c} the value of Λ−Λ_{Ag} is smaller than that for y=6.50, 6.57 and 6.80 samples. Furthermore, the largest field inhomogeneity detected in YBCO above T_{c} is in the y=6.80 sample, which is furthest away from the AF phase. In other words, the hole doping dependence of Λ−Λ_{Ag} presented in Fig. 3(c) is inverted from the usual tendency for the...
field inhomogeneity due to remnant static or fluctuating Cu spins.

For \( T < T_c \), \( \Lambda - \Lambda_{Ag} \) tracks the inhomogeneous field of the vortex lattice and hence tracks the superfluid density \( n_s \propto 1/\lambda_{ab}^2 \), where \( \lambda_{ab} \) is the in-plane magnetic penetration depth [see inset of Fig. 3(c)]. Since \( T_c \) varies as a function of \( \rho_s \) [16], we see in Figs. 3(c) and 3(d) that \( \Lambda - \Lambda_{Ag} \) also tracks \( T_c \). Remarkably this continues to be the case well above \( T_c \) [Figs. 3(e) and 3(f)]. It is thus evident that nonzero \( \Lambda - \Lambda_{Ag} \) above \( T_c \) is in some way coupled to superconductivity.

\textbf{Vortex Liquid.—} Ong et al. [17] have established that the application of a sizeable field creates a Nernst effect and a corresponding field-enhanced diamagnetic signal indicative of a two-dimensional (2D) vortex liquid, which persists above \( T_c \), but is contained within the more extensive pseudogap region. In cuprates, vortices fluctuate about their equilibrium positions with a characteristic fluctuation time (\( \sim 10^{-11} \) s) [18] that is much smaller than the timescale (\( 10^{-6} \) s) of \( \mu \)SR, with a fluctuation-amplitude in the liquid phase on the order of the inter-vortex spacing. These factors conspire to produce severe motional narrowing of the field distribution detected by \( \mu \)SR [19], as observed in Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_{8+\delta}\) (BSCCO) at \( T < T_c \) and \( H < 0.1 \) T [20]. While YBCO and LSCO are less anisotropic than BSCCO, vortex-lattice melting and a loss of vortex line tension also occur before \( T_c \) is reached [21]. As the external magnetic field is increased, a signature of a 2D vortex liquid is a reduction of the \( \mu \)SR line width [19, 21]. However, here we observe the exact opposite. As shown in Figs. 2(b) and 4 \( \Lambda \) increases with increasing field for \( T > T_c \) and remains nonzero even beyond \( T = 2T_c \). Thus the field inhomogeneity detected above \( T_c \) cannot be caused by vortices, which is not to say a vortex liquid does not exist.

\textit{Inhomogeneous Superconductivity.—} It has been proposed that superconductivity first develops in small domes at \( T^* \) [22, 23, 24] that proliferate with decreasing temperature, eventually resulting in the formation of the bulk-superconducting phase at \( T_c \) via percolation or Josephson coupling. This picture is supported by the recent detection of spatially inhomogeneous pairing gaps in BSCCO above \( T_c \) [25], and is compatible with the hysteresis observed in low-field magnetization measurements on LSCO above \( T_c \) by Panagopoulos et al. [26].

Geshkenbein et al. [27] have shown that the presence of small superconducting regions with \( T_c \) greatly exceeding the bulk \( T_c \) explains an unusual linear diamagnetic response (\( M \sim H \)) observed above the resistive transition of highly-overdoped Tl\(_2\)Ba\(_2\)CuO\(_6+\delta\) [28]. In their model, the magnetization of a superconducting grain is proportional to \( H \). Because \( \Lambda / H \) is proportional to the spread
in local magnetic susceptibility at the muon site, then a signature of inhomogeneous superconductivity is a linear dependence of $\Lambda$ on $H$. As shown in Fig. 4 this is observed for $T > 0.8 T_c$, and is distinct from the behavior at lower $T$ where the width of $n(B)$ is dominated by the field inhomogeneity of the vortex lattice. This was also observed in Ref. [10]. The onset temperature for $\Delta \propto H$ behavior is compatible with a thermally induced break-up of the bulk superconducting state into small superconducting domains, beginning at a temperature slightly below $T_c$. Nevertheless, this interpretation must be regarded as speculative, since we cannot say whether there is a diamagnetic response. Also, while electronic phase separation in the form of domains is understandable in cuprates doped by cation substitution, such as BSCCO and LSCO, it is not clear why this should be equally prevalent in YBCO where doping occurs via a change in the oxygen concentration of the CuO-chain layers. Of particular note is ortho-II YBCO $y=6.50$, which has alternating full and empty CuO chains, and therefore has highly ordered doping. Even so, the degree of field inhomogeneity in YBCO is comparable to LSCO, as indicated by the similar values of $\Lambda - \Lambda_{Ag}$ at $p \approx 0.14$ and $T \geq 90 \, \textrm{K}$ [Figs. 3(e) and 3(f)].

We conclude by showing that the spatially inhomogeneous response of YBCO to a $7 \, \textrm{T}$ field persists above the anomalous magnetic order that occurs in zero field near $T^*$ (Fig. 5). If the response is due to electronic moments, what is novel here is that it tracks superconductivity. A compatible picture is one in which magnetic normal-state carriers form pairs and condense at $T_c$. Alternatively, this behavior could arise from spin magnetism being confined to isolated regions by inhomogeneous superconductivity.

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