

Detection of the Unusual Magnetic Orders in the Pseudogap Region of a High-Temperature Superconducting $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$ Crystal by Muon-Spin Relaxation

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We present muon-spin relaxation (μSR) measurements on a large $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$ single crystal in which two kinds of unusual magnetic order have been detected in the pseudogap region by neutron scattering. A comparison is made to measurements on smaller, higher quality $\text{YBa}_2\text{Cu}_3\text{O}_y$ single crystals. One type of magnetic order is observed in all samples, but does not evolve significantly with hole doping. A second type of unusual magnetic order is observed only in the $\text{YBa}_2\text{Cu}_3\text{O}_{6.6}$ single crystal. This magnetism has an ordered magnetic moment that is quantitatively consistent with the neutron experiments, but is confined to just a small volume of the sample ($\sim 3\%$). Our findings do not support theories that ascribe the pseudogap to a state characterized by loop-current order, but instead indicate that dilute impurity phases are the source of the unusual magnetic orders in $\text{YBa}_2\text{Cu}_3\text{O}_y$.

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It is widely believed that the mysterious pseudogap region of high-transition temperature (high- T_c) copper oxide superconductors is caused by a “hidden order.” Varma [1,2] has proposed that the pseudogap is caused by a circulating-current (CC) state that breaks time-reversal symmetry and is characterized by a unique long-range pattern of loop currents in the CuO_2 planes that breaks rotational symmetry, but preserves the translational symmetry of the lattice (TSL). Alternatively, Chakravarty *et al.* [3] have attributed the pseudogap to a competing d -density wave (DDW) order. The DDW phase also breaks time-reversal and rotational symmetries, but has an orbital current pattern that breaks the TSL. In both models the loop-current order is predicted to weaken with increased doping, and to vanish at a quantum critical point somewhat above optimal doping.

Seemingly direct evidence for DDW order comes from neutron scattering experiments on underdoped $\text{YBa}_2\text{Cu}_3\text{O}_y$ (YBCO) with $y = 6.6$ [4,5] and $y = 6.45$ [6], which reveal a weak antiferromagnetic (AF) ordered magnetic moment predominantly directed perpendicular to the CuO_2 planes. However, no such static magnetic order was observed by Stock *et al.* [7] in a neutron study of $\text{YBa}_2\text{Cu}_3\text{O}_{6.5}$. A second unusual magnetic order recently detected in the pseudogap region of YBCO and $\text{HgBa}_2\text{CuO}_{4+\delta}$ (Hg1201) by polarized neutron scattering [8–10] does not break the TSL, and hence is instead qualitatively consistent with the CC phase. However, the ordered moment in this case is not perpendicular to the CuO_2 planes as expected for the CC phase, but rather has a large in-plane component. Spin-orbit coupling [11] or orbital currents involving the apical oxygens [12] have been offered as possible reasons for why the magnetic

moments are severely canted. The first scenario has also been invoked [13] to explain the onset of an accompanying weak ferromagnetism near the pseudogap onset temperature T^* , which has been detected in YBCO by high-resolution polar Kerr effect (PKE) experiments [14].

What is most surprising is the lack of evidence from previous zero-field (ZF) μSR experiments for the existence of loop-current magnetic order. While fields on the order of 100 G are expected in a μSR experiment, no field of this size has been detected in YBCO [15,16] or $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ [17]. To reconcile this discrepancy it has been suggested that charge screening of the positively charged muon (μ^+) severely underdopes its local environment, causing the loop current order to vanish over a distance of several lattice constants [18]. However, the assertion that the muon strongly perturbs its local magnetic environment runs contrary to the excellent agreement between μSR and nuclear magnetic resonance measurements of AF correlations in underdoped cuprates [19]. Furthermore, we demonstrate here that weak magnetic fields detected in YBCO are not caused by undisturbed loop current order located an appreciable distance from the muon.

In contrast to neutron scattering, ZF- μSR is a local probe of magnetism that can distinguish between magnetic and nonmagnetic regions of the sample. The time evolution of the muon-spin polarization $P(t)$ for an ensemble of muons implanted one-by-one in the sample is dependent on the local magnetic field distribution, and is measured by detecting the muon-decay positrons. Magnetic order results in an oscillation of $P(t)$ with a frequency corresponding to the average field experienced by the muons. The ZF- μSR experiments reported here were performed on the M15 surface-muon channel at the Tri-University Meson

Facility (TRIUMF), with the initial spin polarization $P(0)$ parallel to the CuO_2 planes. One of the samples measured is the same large single crystal of YBCO with $y = 6.6$ ($T_c = 62.7$ K) in which neutron experiments [4,5,9] have detected both kinds of unusual magnetic orders in the pseudogap region. The $y = 6.6$ crystal has a cylindrical shape, with a diameter of 22.9 mm and a thickness of 10.8 mm. Measurements were also carried out on YBCO single crystals a thousandth of the size, grown by a self-flux method in fabricated BaZrO_3 crucibles [20]. Mosaics consisting of ~ 10 single crystals of thickness on the order of 0.1 mm, making up a total a - b surface area of 20 to 30 mm^2 , were measured for $y = 6.50$ ($T_c = 59$ K), $y = 6.57$ ($T_c = 62.5$ K), $y = 6.80$ ($T_c = 84.5$ K), and $y = 6.95$ ($T_c = 93.2$ K), and for Ca-doped YBCO with $y = 6.98$ ($T_c = 75$ K).

Figure 1(a) shows typical ZF- μ SR spectra for the large $y = 6.6$ crystal. The spectra for all samples are well described over a 9 μs time range by the following polarization function,

$$P(t) = P_{\text{KT}}(t) \cos(2\pi\nu_1 t) \\ = \left[\frac{1}{3} + \frac{2}{3}(1 - \Delta^2 t^2) e^{-\Delta^2 t^2/2} \right] \cos(2\pi\nu_1 t), \quad (1)$$

with a relaxation rate of $\Delta = 0.101264 \mu\text{s}^{-1}$. The static Gaussian Kubo-Toyabe function $P_{\text{KT}}(t)$ approximately describes the time evolution of the muon-spin polarization caused by the randomly oriented nuclear moments. The oscillatory component implies the presence of magnetic order that generates a field $B_1 = 2\pi\nu_1/\gamma_\mu$ at the muon site, where $\gamma_\mu = 0.0852 \mu\text{s}^{-1} \text{G}^{-1}$ is the muon gyromagnetic ratio. Hence, the temperature dependence of the muon-spin precession frequency ν_1 [Figs. 1(b)–1(g)] indicates changes in the average local field.

Figure 2 shows ν_1 for the $y = 6.6$ crystal superimposed on the temperature dependence of the square root of the neutron magnetic scattering [4]. Both quantities are proportional to the size of the ordered moment. The striking agreement between these two data sets suggests that ν_1 is caused by the same anomalous weak AF order detected by neutrons. In YBCO with $y = 6.6$, approximately 60% of the muons stop near a chain oxygen at (0.15, 0.44, 0.071), and 40% near an apical oxygen at (0.275, 0, 0.1333) [21,22]. Taking the ordered moment to be no larger than $0.02\mu_B$ (as established in Ref. [4]) and assuming DDW order [3], the calculated dipolar field at the chain and apical oxygen muon sites is less than 5.2 G and 12.1 G, respectively. This is consistent with $B_1 \sim 1.6$ G at the lowest temperature. However, since the temperature dependence of ν_1 is qualitatively similar in the other samples and evolves little with hole-doping concentration p [see Figs. 1(b)–1(g)], it is clear that the origin of the moments cannot be the orbital currents of the predicted DDW phase.

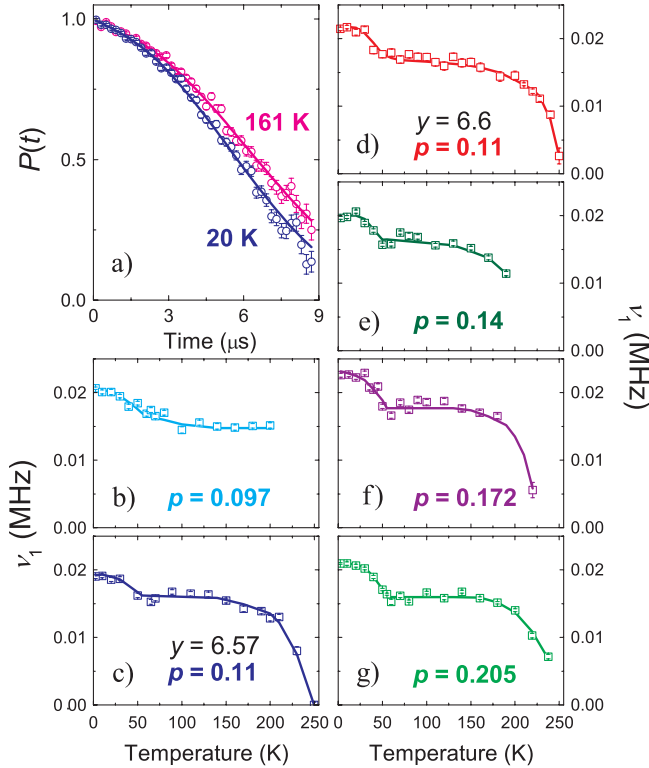


FIG. 1 (color online). (a) Representative ZF- μ SR signals for the large $y = 6.6$ single crystal. The solid curves are fits to Eq. (1). Also, temperature dependence of ν_1 for the smaller YBCO single crystals with oxygen content (b) $y = 6.50$, (c) $y = 6.57$, (e) $y = 6.80$, (f) $y = 6.95$, and for (g) the small Ca-doped YBCO single crystals with $y = 6.98$, and for (d) the large $y = 6.6$ single crystal. The solid curves in (b)–(g) are guides to the eye. The error bars come from the fits. The hole-doping concentration p for each sample is shown in the respective panel.

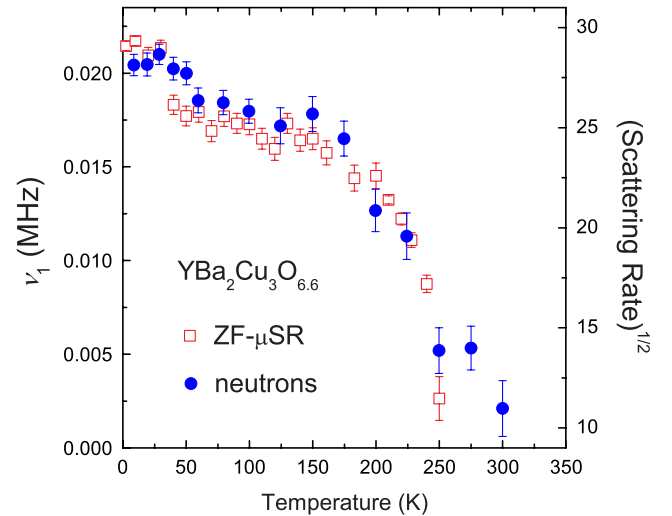


FIG. 2 (color online). A comparison of the temperature dependence of ν_1 for the large $y = 6.6$ single crystal with the square root of the neutron scattering rate at wave vector (0.5, 0.5, 2) from Ref. [4]. Both quantities are proportional to the ordered magnetic moment.

Here we note that the onset of the pseudogap [23] for YBCO with $y = 6.95$ ($p = 0.172$) is well within the temperature range of Fig. 1(f). Furthermore, loop-current magnetic order is not expected to occur in the Ca-doped sample with $p = 0.205$. The tiny size of the ordered moment detected in Refs. [4–6] may be an indication that the magnetic order resides in a small volume fraction of the sample. One cannot tell this from the neutron measurements. Unfortunately the ZF- μ SR measurements are equally unrevealing, because ν_1 is comparable in size to the dipolar fields generated by the nuclear moments.

Next we turn our attention to the early times of the ZF- μ SR spectra. Figures 3(a)–3(d) demonstrate a small-amplitude oscillatory signal below $T \sim 250$ K in the large $y = 6.6$ crystal. The ZF- μ SR spectra over the first 1.5 μ s are best described by the addition of a second oscillatory term to Eq. (1):

$$P(t) = (1 - f)P_{\text{KT}}(t) \cos(2\pi\nu_1 t) + f e^{-\Lambda t} \cos(2\pi\nu_2 t), \quad (2)$$

where f is the volume fraction of the oscillating signal in Fig. 3. The temperature dependence of f and ν_2 are shown

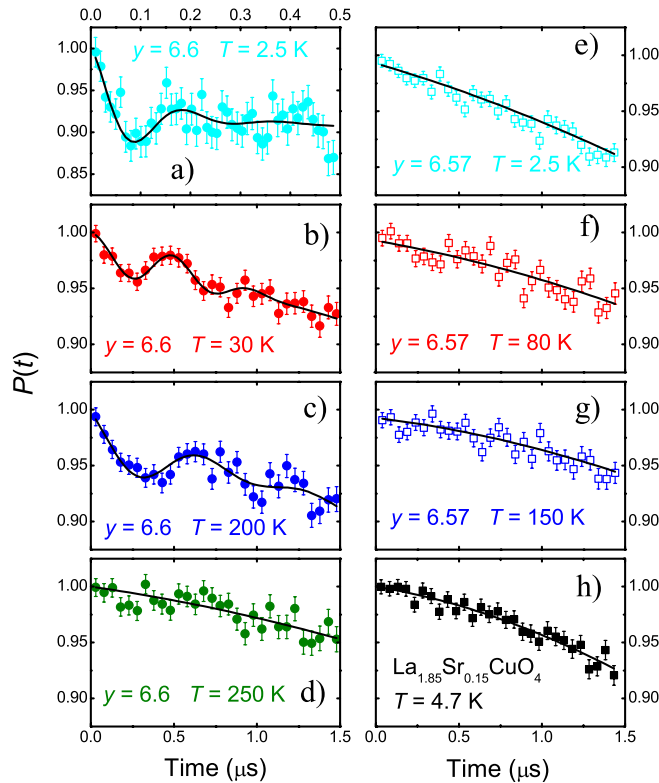


FIG. 3 (color online). Time evolution of the ZF- μ SR signal at early times for (a)–(d) the large $y = 6.6$ single crystal, (e)–(g) the mosaic of $y = 6.57$ single crystals, and (h) a single crystal of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$. The spectra are made up of ~ 11 million muon-decay events, with the exception of (e) and (h), which have 3 and 5 times this number of statistics, respectively. The solid curve in each panel is a fit to Eq. (2). Note the difference in the time scale of (a) relative to (b)–(h).

in Fig. 4. Below $T \sim 15$ K we observe a rapidly damped oscillatory signal coming from approximately 8% of the sample [see Fig. 3(a)]. At $T = 2.5$ K the oscillatory frequency is $\nu_2 = 5.1 \pm 0.4$ MHz, which corresponds to an average local field of $B_2 = 376 \pm 30$ G. We attribute this 8% component to the presence of the “green phase” Y_2BaCuO_5 that is known to occur in this sample [5] and undergo an AF transition at $T \sim 15$ K [24]. Between $T = 30$ K and $T = 250$ K a smaller, slower decaying oscillatory signal is observed [see Figs. 3(b)–3(d)] indicating the occurrence of an additional kind of magnetic order in approximately 3% of the sample. The fact that we cannot follow this below $T = 30$ K is simply a consequence of the 8% impurity phase dominating the smaller signal. A substantially higher number of counts would be needed to simultaneously resolve both of these oscillatory components at low T . At $T = 30$ K the frequency of the 3% oscillatory signal is $\nu_2 = 1.92 \pm 0.15$ MHz, corresponding to a local field of $B_2 = 142 \pm 11$ G. Figures 3(e)–3(g) show the absence of these oscillatory signals in the $y = 6.57$ sample, which has the same hole-doping concentration as the large $y = 6.6$ single crystal. We have searched hard with higher counting statistics, and find no evidence for either oscillatory signal in any of the other YBCO samples or in a single crystal of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ [see Fig. 3(h)].

In a deliberate search for the CC phase in the $y = 6.6$ crystal with polarized neutron scattering [9], a transition to magnetic order that preserves the TSL was observed at $T = 235 \pm 15$ K. The results are similar to that reported earlier by Fauqué *et al.* [8]. The ordered moment is about $0.1 \mu_B$ and is directed $55 \pm 7^\circ$ from the c axis. To determine whether the 142 G field detected here by ZF- μ SR is

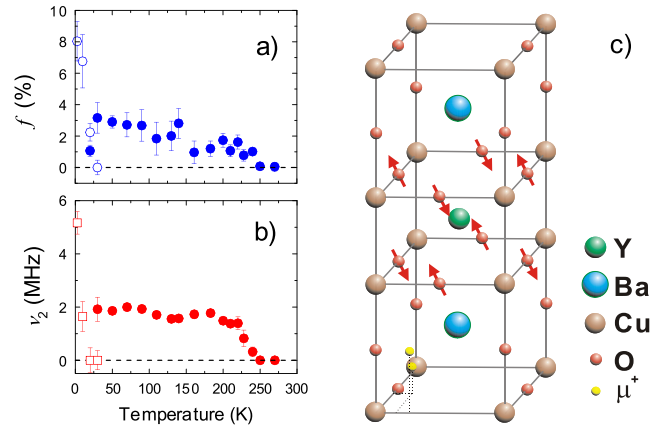


FIG. 4 (color online). Temperature dependence of (a) f and (b) ν_2 from fits to the ZF- μ SR signals for the $y = 6.6$ single crystal to Eq. (2). (c) A staggered arrangement of spins on the oxygen sites in the CuO_2 planes that accounts for the average field of the 3% oscillatory component. The spins are rotated in the CuO_2 planes $\pm 45^\circ$ from the a axis, and are directed 55° (or 235°) from the c axis. Also shown are the two muon-stopping sites in YBCO with $y = 6.6$.

associated with the same magnetic order, we consider a model of staggered spins on the oxygen sites—which was introduced in Ref. [8] as one possible interpretation of the neutron measurements. Assuming the neutron scattering intensity associated with the magnetic order comes from only 3% of the sample, the true ordered moment is $(1/0.03)^{1/2} \times 0.1\mu_B = 0.58\mu_B$. A staggered moment of this size on the oxygen sites canted at $55 \pm 7^\circ$ [see Fig. 4(c)] gives a resultant field of 107 ± 5 G and 192 ± 14 G at the chain and apical muon sites, respectively. The corresponding muon-site population average is 141 ± 6 G, which is in remarkable agreement with the average field of the 3% signal. Because the muon sites are outside the CuO_2 bilayers, reversing the direction of the spins in one of the CuO_2 planes of Fig. 4(c) increases the field only slightly to 144 ± 7 G. Alternatively, circulating currents involving the apical oxygens [12] give a muon-site-averaged field of about 127 G, which is also compatible with the observed precession frequency. However, the exceedingly small 3% magnetic volume fraction and the absence of a similar oscillating component in the other samples do not support the existence of a CC phase.

While our experiments indicate that the unusual magnetic orders are associated with impurity phases, we can only speculate on what these are. The first kind of magnetic order may be remnant traces of the starting material CuO. Bulk CuO exhibits AF phase transitions at $T_{N1} = 230$ K and $T_{N2} = 213$ K, which are strongly coupled to the crystal lattice [25]. A rise in magnetization also occurs in CuO near $T = 50$ K [26], where lattice structure changes are observed in YBCO [27,28]. These different behaviors are noticeable in Fig. 1, and as expected do not change appreciably with hole doping. We note that the onset of the ferromagnetic-like PKE signal in YBCO with $y = 6.92$ ($T_c = 92$ K) occurs at $T \sim 50$ K, and is disproportionately weak compared to the PKE signal observed at higher temperatures in underdoped samples [14]. As mentioned in Ref. [14], impurities can induce a ferromagnetic component in an otherwise AF environment. At the very least our findings create ambiguity about the interpretation of the PKE signal near optimal doping, and hence whether there is a quantum critical point under the superconducting “dome.”

Since variations in oxygen content are the doping mechanism for both YBCO and Hg1201, oxygen defects may play some role in the second kind of unusual magnetic order. In YBCO, short-range oxygen-vacancy ordering occurs in small dilute regions [28–30]. However, something else more exotic, such as a small concentration of Cu vacancies in the CuO_2 planes [31], may be required to induce the observed magnetic order.

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