Dynamic correlated Cu(2) magnetic moments in superconducting $YBa_2(Cu_{0.96}Co_{0.04})_3O_y$ ($y\sim7$)

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We have examined the magnetic properties of superconducting $YBa_2(Cu_{0.96}Co_{0.04})_3O_y(y\sim7, T_{sc}=65\ K)$ using elastic neutron scattering and muon spin relaxation (μSR) on single-crystal samples. The elastic-neutron-scattering measurements evidence magnetic reflections, which correspond to a commensurate antiferromagnetic Cu(2) magnetic structure with an associated Néel temperature $T_N\sim400$ K. This magnetically correlated state is not evidenced by the μSR measurements. We suggest that this apparent anomaly arises because the magnetically correlated state is dynamic in nature. It fluctuates with rates that are low enough for it to appear static on the time scale of the elastic-neutron-scattering measurements, whereas on the time scale of the μSR measurements, at least down to ~50 K, it fluctuates too fast to be detected. The different results confirm the conclusions reached from work on equivalent polycrystalline compounds: the evidenced fluctuating, correlated Cu(2) moments coexist at an atomic level with superconductivity.

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I. INTRODUCTION

For the cuprates showing high-temperature superconductivity, there is much interest in how the superconducting properties of the Cu(2)-O planes and the Cu(2)-based magnetic order evolve as a function of the doping level.[1] At the present time, the general consensus is that the ground state involves magnetically ordered Cu(2) for low dopings (typically for a doping level, $p<\sim0.05$), it involves *d*-wave superconductivity (for $p\sim0.05/0.10$ to ~0.25) and it involves a metallic state for higher dopings. The intermediate regions notably the underdoped pseudogap regime, where both superconductivity and Cu(2) magnetic order may be present, are not yet fully understood. In the underdoped region, it is problematic to experimentally determine whether or not superconductivity and Cu(2) magnetic order are mutually exclusive at an atomic level. However, since it is well established that at one end of the phase diagram there is a region with magnetically ordered Cu(2) and no superconductivity and that around optimum doping there is a region with superconductivity and no magnetically ordered Cu(2), the two phenomena are usually considered to be mutually exclusive at an atomic level. Previously, however,[2–5] we have shown that this view is not always valid.

In Ref. 4 (preceding paper), we reported that when Ni is substituted into fully oxidized $YBa_2Cu_3O_y(y\sim7)$, the Cu(2) in the neighborhood of each Ni carry magnetic moments, which are short range magnetically correlated. For the examined Ni concentration of 4%, essentially all the Cu(2) of the sample were found to carry correlated magnetic moments. These moments undergo temperature-dependent fluctuations with rates up to the GHz range.

We have also observed that when Co (rather than Ni) is substituted into fully oxidized $YBa_2Cu_3O_y(y\sim7)$, correlated magnetic Cu(2) moments are again clearly visible. These results were obtained from elastic-neutron-scattering measurements on a single crystal of $YBa_2Cu_3O_y(y\sim7)$ substituted with 1.3% of Co (Ref. 3) and ^{170}Yb Mössbauer (Ref 2) and μSR (Ref. 5) probe measurements on polycrystalline samples of $YBa_2Cu_3O_y(y\sim7)$ substituted with 1.0%–4.0% of Co. Here, we report additional results, obtained from elastic-neutron-scattering and μSR measurements, on superconducting single-crystal samples of $YBa_2Cu_3O_y(y\sim7)$ substituted with 4.0% Co.

II. SAMPLES, METHODOLOGY

The single-crystal samples of $YBa_2(Cu_{0.96}Co_{0.04})_3O_y(y\sim7)$ were prepared by the top seeding, melt texturing method and they were subsequently annealed in oxygen. Because of the growth technique used, the samples necessarily contain Y_2BaCuO_5 as a second phase ($\sim$40% by weight) as well as small amounts of some other impurities.

T_{sc}, the superconducting transition temperature, is 65 K, which is typical for a 4% Co substitution level. The elastic-neutron-scattering measurements were carried out at the Laboratoire Léon Brillouin, Saclay, France and the muon probe measurements were made at the ISIS facility of the Rutherford-Appleton Laboratory, Chilton, U.K.

The Co enters only the Cu(1) or chain sites and it pulls in additional oxygen atoms.[6–8] The Co tends to form dimers[6,8] or even short chains along the (110) direction,[7] which introduce microtwinning and trigger the change to the tetragonal phase that occurs for Co levels ~0.025. The introduction of Co reduces the carrier density[9,10] and the resulting underdoped samples evidence a pseudogap, which is seen, for example, in local spin susceptibility measurements.[11] Nuclear quadrupole resonance measurements have shown that the substitution of Co into $YBa_2Cu_3O_7$ introduces magnetic moments at both the chain Cu(1) and plane Cu(2) sites.[12]

III. EXPERIMENTAL RESULTS

Before presenting the results for the single-crystal samples, we briefly recall the results that have been obtained on the corresponding polycrystalline samples. Both ^{170}Yb Mössbauer Ref. (2) and μSR (Ref. 5) local probe measurements on superconducting samples of $YBa_2Cu_3O_y$(y ~ 7) substituted with Co have shown that the samples contain correlated magnetic Cu(2) moments. These moments are introduced over a distance of approximately 3 to 4 a/b lattice spacings around the Co. This length scale is similar to that of the staggered Cu(2) moments, which are introduced around Ni atoms substituted in $YBa_2Cu_3O_7$ as obtained from NMR measurements.[13,14] The Cu(2) magnetic correlations in superconducting $YBa_2Cu_3O_y$(y ~ 7) substituted with Co are short range since the magnetic reflections are too broad to be seen by neutron-diffraction measurements.[15] The correlated Cu(2) moments are dynamic with temperature-dependent rates. The rates increase as the temperature increases and they extend up to the GHz range. At each particular temperature, the local fluctuation rates show a distribution.

These characteristics are quite similar to those evidenced by polycrystalline superconducting $YBa_2Cu_3O_y$(y ~ 7) substituted with Ni,[4] with, however, one difference: at a given temperature, the average fluctuation rate of the correlated Cu(2) moments in samples containing Co (Refs. 2 and 5) is considerably lower than that in samples containing an equivalent amount of Ni (Ref. 4).

A. Elastic-neutron-scattering measurements

The details concerning the measurement procedure are given in Ref. 3, which reported well-defined magnetic reflections in a superconducting, single-crystal of $YBa_2Cu_3O_y$(y ~ 7) substituted with 1.3% Co. The reflections corresponded to commensurate antiferromagnetically (AF) correlated Cu(2) magnetic moments with correlation lengths typically > ~ 20 nm. For the superconducting single crystal of $YBa_2Cu_3O_y$(y ~ 7) substituted with 4% Co considered here, similar well-defined magnetic reflections are observed and they again evidence commensurate antiferromagnetically correlated Cu(2) magnetic moments. As is discussed below, this magnetically correlated state possesses some, but not all, of the characteristics generally associated with conventional (static) long-range magnetic order. To distinguish the presently reported state from that of conventional long-range magnetic order, we refer to it as an extended range, magnetically correlated state.

The temperature dependence of the neutron-scattering intensities at **Q**=(0.5,0.5,−1.5) and (0.5,0.5,−2) is shown in Fig. 1. As the temperature is reduced below ~300 K, the intensity at **Q**=(0.5,0.5,−2) initially increases progressively and then below ~90 K it decreases. Near 90 K, an intensity appears at (0.5,0.5,−1.5), which progressively increases as the temperature is lowered. This behavior indicates that near 90 K, the system undergoes a AF1-AFII transition characterized by a doubling of the AF unit cell along the *c* axis. A similar doubling of the AF unit cell also occurred in the superconducting sample containing 1.3% Co but at a lower temperature (~12 K).[3] It also occurs in nonsuperconducting $YBa_2Cu_3O_{6+\delta}$ when substituted at the Cu(1) site.[16,17] In the nonsuperconducting samples, the temperature of the AF1-AFII transition also increases as the concentration of the substituted cation increases.

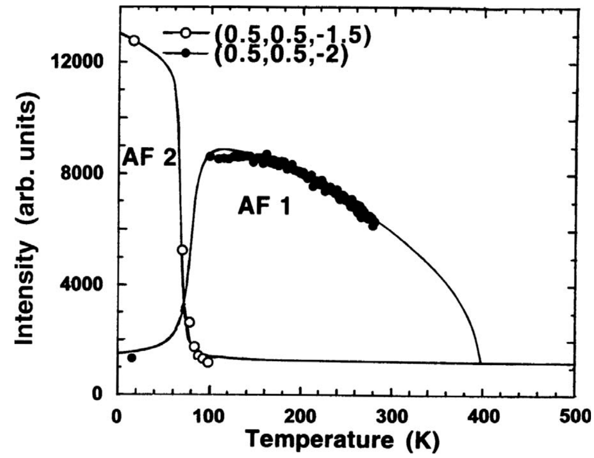

FIG. 1. From elastic-neutron-scattering measurements on single-crystal superconducting $YBa_2(Cu_{0.96}Co_{0.04})_3O_7$ (y ~ 7) mixed with Y_2BaCuO_5: temperature dependence of the scattering intensity at **Q**=(0.5,0.5,−2), which displays a re-entrant behavior below ~90 K and at **Q**=(0.5,0.5,−1.5), where the intensity grows below ~90 K. The Néel temperature, T_N ~ 400 K, is obtained by extrapolation as described in the text.

Although the measurements were made at room temperature and below, this range is sufficiently wide to provide a reasonable estimate of the Néel temperature (T_N), which lies above room temperature. The extrapolation was made assuming that the thermal dependence of the scattering intensity, normalized to T_N, is the same as in the sample with 1.3% Co.[3] The obtained value, T_N ~ 400 K, is higher than that for the sample with 1.3% Co (T_N ~ 330 K).[3] The maximum size of the correlated Cu(2) magnetic moment (~$0.3\mu_B$), is approximately twice that (~$0.14\mu_B$) observed in the part of the sample containing 1.3% Co, where the moments are extended range correlated.[3]

Using elastic neutron scattering, a total of six separately prepared superconducting samples of $YBa_2(Cu_{1-x}Co_x)_3O_y$(y ~ 7) have been examined. Each of them evidences well-defined magnetic reflections corresponding to antiferromagnetically correlated Cu(2) magnetic moments. This characteristic is thus very robust. However, in samples having the same concentration of Co, the size of the Cu(2) magnetic moment obtained assuming all the Cu(2) in the sample contribute to the magnetic reflections, is found to vary from sample to sample. It was generally lower (by up to 40%) than the value ~$0.3\mu_B$ reported for the sample above.

The origin of this sample-dependent behavior may be considered in terms of two limiting cases. It could be due to a variation in the size of the extended-range-correlated moments [assuming all the Cu(2) contribute to the magnetic reflections in each sample] or it could be due to a variation in the volume fraction over which the Cu(2) are extended range correlated. In this context, we recall that our local probe measurements on a number of separately prepared single phase, polycrystalline samples of $YBa_2Cu_3O_y$($y\sim7$) substituted with 4% Co show that in each sample essentially all the Cu(2) carry (short range) correlated magnetic moments[2] and that the average size of the moment remains essentially the same. We thus anticipate that in the single-crystal samples, all the Cu(2) also carry correlated magnetic moments having a common average size. We then attribute the variation in the neutron-measured Cu(2) magnetic moment from sample to sample to variations in the relative volumes where respectively the correlations are extended range (which contribute to the narrow magnetic reflections) and short range (which do not contribute to the narrow magnetic reflections).

With this description in terms of different relative volumes of the extended-range-correlated and short-range-correlated fractions, it also follows that it is possible that not all the Cu(2) are extended-range-correlated in any of the single-crystal samples including that reported above (Fig. 1). In this case, the average size of the Cu(2) moment for this sample would be higher than the value of $\sim0.3\mu_B$ obtained assuming that all the Cu(2) in the sample are extended-range-correlated.

We do not know why the correlations are sufficiently extended range in the single crystals to give rise to magnetic reflections whereas in the equivalent polycrystalline samples they remain short range. It is possible that the $\sim40\%$ of Y_2BaCuO_5, which is distributed throughout the single-crystal sample volume could play a role. To test this, it would be of interest to examine if the extended range correlations are present in equivalent single-crystal samples, which do not contain any secondary phases.

The AFI and AFII-type magnetic structures we observe in the superconducting samples are also present in nonsuperconducting $YBa_2Cu_3O_6$ (AFI)[18] and in nonsuperconducting $YBa_2Cu_3O_6$ substituted at the Cu(1) sites (AFI and AFII).[16,17] Comparing the results obtained here with those in nonsuperconducting $YBa_2Cu_3O_6$, we note that the Néel temperature in the superconducting sample (~400 K) is only marginally lower than that of the nonsuperconducting sample (~420 K), whereas the sample-averaged size of the Cu(2) magnetic moment in the superconducting sample ($\sim0.3\mu_B$) amounts to a considerable fraction of that in nonsuperconducting sample ($0.61\mu_B$).

There are thus a number of similarities between the extended-range magnetically correlated state observed in superconducting $YBa_2(Cu_{0.96}Co_{0.04})_3O_y$($y\sim7$) and the conventional long-range magnetically ordered state observed in nonsuperconducting $YBa_2Cu_3O_6$. There is, however, one important difference. Whereas for $YBa_2Cu_3O_6$, the conventional (static) magnetic order observed by neutron measurements[18] is also strongly evidenced by μSR measurements[19] (in fact, the μSR measurements preceded the neutron measurements in identifying the existence of magnetic order), for single crystal $YBa_2(Cu_{0.96}Co_{0.04})_3O_y$($y\sim7$), as is described in the following subsection, the extended-range-correlated state (both AFI and AFII structures) evidenced by the neutron-scattering measurements is not evidenced by the μSR measurements.

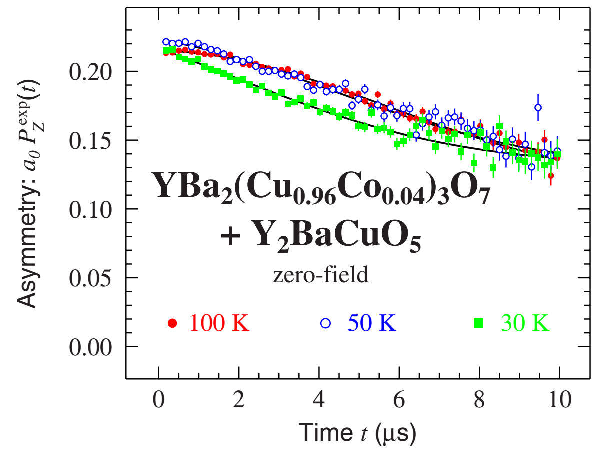

FIG. 2. (Color online) μSR spectra for single crystal $YBa_2(Cu_{0.96}Co_{0.04})_3O_y$ ($y\sim7$) mixed with of Y_2BaCuO_5 at temperatures above the magnetic ordering temperature of Y_2BaCuO_5. Despite the fact that neutron-scattering measurements on this sample show the Cu(2) in $YBa_2(Cu_{0.96}Co_{0.04})_3O_7$ give rise to magnetic reflections, which correspond to a commensurate antiferromagnetic structure with an associated Néel temperature above room temperature, the presence of this correlated state does not markedly influence the depolarization (there is no reduction in the initial asymmetry and there are no oscillations).

B. μSR measurements

The μSR measurements were made on a sample where the value of the extended-range-correlated Cu(2) magnetic moment measured by neutron scattering was $\sim0.2\mu_B$. Since the sample contains $\sim40\%$ of Y_2BaCuO_5, which orders magnetically near 15 K and since the muons are implanted over the whole sample volume, the μSR response will involve contributions from the two main constituents as well as from the muons which contribute to the background. It turns out that it is not possible to unambiguously define the contributions coming from each of the two main phases in the sample or to precisely define the contribution coming from the background. Because of this, it is not feasible to make a detailed quantitative analysis and this is especially the case at temperatures near and below the magnetic ordering temperature of Y_2BaCuO_5. Nevertheless, even limiting the analysis to the temperatures of 30 K and above, it is possible to obtain pertinent information.

Figure 2 shows the μSR depolarization at 100, 50, and 30 K with the incident muons propagating parallel to the c axis of the single crystal. We first describe the simplified approach, made in terms of sample-averaged parameters, which was used to obtain the data fits shown. At 100 K, the time-dependent part of total depolarization has the standard Kubo-Toyabe form ($a_sP_Z^{KT}$), which is characteristic of an interaction between the muon spins and adjacent nuclear magnetic moments. The fitted value for the (sample averaged) field width at the muon site is $\Delta^{KT}=0.147(3)$ mT. On reducing the temperature to 50 and 30 K, the depolarization shows modest changes. At 50 and 30 K, the time-dependent part of the depolarization is well-described with a relaxation function $a_s P_Z^{KT} \exp(-\lambda_Z t)$, where λ_Z is the (sample averaged) muon relaxation rate. Assuming Δ^{KT} is independent of temperature, we obtain (sample averaged) values for λ_Z of $0.051(7)\times 10^6$ (50 K) and $0.124(10)\times 10^6 (30\ \mathrm{K})\ \mathrm{s}^{-1}$.

We return to these values for λ_z below. Here, we consider the qualitative aspects of the μSR depolarization. Down to $\sim$100 K, the depolarization only evidences interactions between the muon spin and adjacent nuclear moments and there is no evidence of any interaction between the muon spin and electron-based magnetic moments. An interaction with electron-based moments is detected near 50 K and below through the appearance of a measurable muon spin-relaxation rate, whose value increases as the temperature is further reduced. These results pertain to a sample, where the major part $(YBa_2(Cu_{0.96}Co_{0.04})_3O_y, y\sim 7)$ evidences extended-range Cu(2) magnetic correlations with a T_N above room temperature. The magnetically correlated Cu(2) thus have no (at 100 K and above) or little (at 50 K and below) influence on the muon depolarization. We also note that whereas conventional (static) long-range magnetic order systematically has a very marked influence on the μSR depolarization (reduction in the initial asymmetry or/and oscillations in the time dependence), Fig. 2 shows that at each of the three temperatures, the initial asymmetry remains at its full value and there are no oscillations.

We suggest that the neutron scattering evidenced extended-range magnetically correlated state has little or no influence on the μSR measurements because the correlated state is dynamic: it fluctuates with rates that are fast enough to "motionally narrow" its influence on the μSR measurements.

Combined elastic-neutron-scattering and μSR measurements on single crystal $YBa_2Cu_3O_{6.5}(T_{sc}=55\ \mathrm{K})$, prepared by the top seeding method have previously identified similar behaviors, where a commensurate Cu(2) antiferromagnetic order with $T_N=310$ K, and magnetic moments of $\sim 0.05\mu_B$ [assuming a homogeneous distribution of the moments at all the Cu(2) sites] involved dynamic features with a time scale, which is longer than that associated with the neutron-scattering measurements but shorter than that associated with the μSR measurements.[20] The arguments presented there to outline why the moments of $\sim 0.05\mu_B$ would have been readily detected by μSR if they had been "static" are also relevant to the present case, where the moments are approximately four times bigger.

We now return to the (sample averaged) muon spin-relaxation rates given above since their thermal dependences provide some information concerning the dynamically correlated state. As Y_2BaCuO_5 magnetically orders near 15 K, we anticipate that in the range of 30 to 50 K, it will have a paramagnetic behavior and the local moments will undergo spin-spin-driven fluctuations with rates that are essentially independent of temperature. We thus attribute the changes observed in the (sample averaged) muon relaxation rate in this temperature range to changes that occur in the dynamics of the extended-range-correlated moments in the $YBa_2(Cu_{0.96}Co_{0.04})_3O_y(y\sim 7)$ fraction. Thus, it is the slowing down of the fluctuation rate of the correlated Cu(2) moments in the superconducting fraction, which gives rise to the appearance (near 50 K) and to the increase (below 50 K) in the muon spin-relaxation rate. A quite similar appearance (near 50 K) and a quite similar increase (below 50 K) in the muon spin-relaxation rate are also observed in the equivalent single-phase superconducting polycrystalline samples,[5] where they are clearly related to the properties of the (short range) magnetically correlated Cu(2).[2,5]

IV. COMMENTS

Although it is not commonplace to encounter a state involving fluctuating extended-range-correlated magnetic moments, in addition to the case of $YBa_2Cu_3O_{6.5}$ mentioned above,[20] this state has been observed recently in the rare-earth pyrochlores, where frustration plays a role.[21–27] In a manner analogous to that reported here, Ref. 26 reported both the observation of neutron-scattering peaks and of "motional narrowing" of the μSR response in a particular pyrochlore. The frequency scale of the fluctuations was on the order of $10^{10}\ \mathrm{s}^{-1}$ and the maximum magnetic correlation length was $\sim$20 nm.[23] For single crystal $YBa_2(Cu_{0.96}Co_{0.04})_3O_y(y\sim 7)$, it is difficult to obtain good estimates of the fluctuation rates, which are probably both strongly temperature dependent and distributed in size. We anticipate that depending on the temperature, they could lie in the range from $\sim 10^7\ \mathrm{s}^{-1}$ [the lowest value measured in polycrystalline $YBa_2(Cu_{0.96}Ni_{0.04})_3O_y, y\sim 7$ (Ref. 4)] to $\sim 10^{11}\ \mathrm{s}^{-1}$ (observed in polycrystalline $YBa_2(Cu_{0.96}Co_{0.04})_3O_y, y\sim 7$).[2]

Clearly more experimental work on the single crystals is required (including samples not containing impurity phases) so as to better define the dynamic properties of the correlated Cu(2) moments. The present study simply suggests that the extended range, commensurate, antiferromagnetic Cu(2) state evidenced by the elastic-neutron-scattering measurements on superconducting $YBa_2(Cu_{0.96}Co_{0.04})_3O_y, y\sim 7$ is not conventional in that it possesses dynamic characteristics.

Finally, we note that although the details of the properties of the correlated Cu(2) magnetic moments are of interest, in the context of the present work, a more important aspect is simply that these fluctuating moments exist.

V. SUMMARY AND CONCLUSIONS

When Co is substituted [at the Cu(1) sites] into fully oxidized $YBa_2Cu_3O_y(y\sim 7)$, the Cu(2) in its neighborhood carry antiferromagnetically correlated moments. From measurements on polycrystalline samples, we previously found that a Co substitution level of $\sim$2% was sufficient for essentially all the Cu(2) to carry magnetic moments (in such samples $T_{sc}=84$ K). This indicates that the range around the Co with correlated Cu(2) moments is approximately three to four a or b lattice spacings.

A mechanism has been proposed to explain the magnetically correlated Cu(2) observed in *underdoped* cuprates,[28] so that this mechanism could play a role here. It is possible, however, that a different and so far undefined mechanism is responsible for the correlated Cu(2) moments observed both in underdoped ($YBa_2Cu_3O_7$+Co) cuprates and in optimally doped ($YBa_2Cu_3O_7$+Ni) cuprates.[4]

For reasons that remain to be established, the omnipresent fluctuating, short-range magnetically correlated Cu(2) state observed in polycrystalline $YBa_2(Cu_{0.96}Co_{0.04})_3O_y (y \sim 7)$ becomes extended range in single-crystal samples. There are few existing examples of a state involving fluctuating, extended-range-correlated moments but this has been evidenced recently both in $YBa_2Cu_3O_{6.5}$ (Ref. 20) and in some of the rare-earth pyrochlores, where magnetic frustration plays a key role.[21–27] Whether frustration plays a role in fashioning some of the exotic properties of the superconducting cuprates remains to be seen.

The results presented here provide further evidence that the Cu(2)-O network of the planes is capable of supporting superconductivity when all the Cu(2) carry fluctuating correlated magnetic moments.

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