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Anomaly in $YBa_2Cu_4O_8$ charge distribution below T_c: A zero-field muon spin relaxation study

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Zero-field muon spin-relaxation (μSR) measurements in ^{63}Cu isotope enriched and natural $YBa_2Cu_4O_8$ powders are presented. The μ^+ relaxation rate is characterized by a sizeable enhancement as the temperature is lowered below the superconducting transition temperature T_c. The comparison of the asymmetry decay in the two samples reveals that the μ^+ relaxation is driven by nuclear dipole interaction from 300 K down to 4.2 K. It is argued that the increase in the relaxation rate below T_c originates from a change either of the μ^+ site or of the orientation of the electric-field gradient at the Cu nuclei, due to a modification in the charge distribution within CuO chains.

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I. INTRODUCTION

In strongly correlated metals the electrons can be subject to interactions which are of the order of a fraction of the bandwidth and can cause an enhancement of instabilities, as superconductivity, and to crossovers among different regimes.[1] One of the most intriguing examples of such a scenario is the phase diagram of high-T_c superconductors, which are characterized by a relatively narrow bandwidth and by sizeable exchange and local lattice interactions. Although this phase diagram has been extensively explored and several crossover temperatures evidenced, the microscopic mechanisms leading to such a complex phase diagram are still subject of an intense debate. For instance, the origin of the pseudogap is not clear yet. The occurrence of a mesoscopic phase separation and/or of a charge order in the CuO_2 planes and their relevance to the mechanism of superconductivity is still controversial. Moreover, it is not established which is the role of CuO chains on the electronic properties of certain families of cuprates; namely, if CuO chains can be considered to a certain extent decoupled from the underlaying CuO_2 planes and be characterized by an independent phenomenology, typical of a Tomonaga-Luttinger liquid,[2] yielding some of the crossovers experimentally detected in these compounds.

A few years ago, Kramers and Mehring revealed a crossover below T_c in optimally doped $YBa_2Cu_3O_{6+x}$(Y123).[5] At the crossover temperature a peak in the Cu(2) nuclear transverse relaxation rate and a concomitant increase in the nuclear quadrupole resonance (NQR) linewidth were observed. It was argued that these anomalies could be associated with the onset of a charge order in the CuO_2 plane below $T \simeq 40$ K. Later on, Sonier *et al.*[3] observed in the same compound an anomalous increase in the zero-field (ZF) μ^+ relaxation rate below T_c which, at first, was associated with the pseudogap crossover temperature T^*. Afterwards,[4] it was realized that this anomalous increase occurred at the same temperature where the anomalies in NQR spectra and relaxation were detected, suggesting that both techniques were detecting the same type of crossover.

In order to clarify if the increase in the ZF muon spin relaxation (μSR) occurs at T^*, if it originates from a modification in the local-field distribution due to electron or nuclear spins and if it is related to a charge order, we have performed ZF μSR measurements in ^{63}Cu enriched and natural $YBa_2Cu_4O_8$(Y124). The peculiarity of Y124 is that, unlike Y123, it is characterized by a well-defined oxygen stoichiometry and by a precise value of $T^* \simeq 150$ K (Ref. 6) and hence it is the best system where one can check if there is any correlation between T^* and the anomalous increase in the muon relaxation rate. Moreover, the comparison of μ^+ relaxation rate in samples with different abundances of ^{63}Cu isotope allows one to clarify the origin of the local-field distribution at muon sites.

II. EXPERIMENTAL RESULTS

Y124 powder samples were grown following a standard procedure.[7] Appropriate amounts of Y_2O_3 (99.99%, Alfa Aesar), $BaCO_3$ (99.98%, Aldrich), enriched $Cu^{63}O$ (99.9%), and natural CuO (99.99%, Aldrich) were mixed, then pressed into pellets, and annealed in air at 850–910 °C for 150 h, with several intermediate regrindings. X-ray diffraction revealed that the resulting samples were a mixture of R-123 and CuO. These samples were placed into Al_2O_3 crucibles and subjected to the high oxygen treatment in a double-chamber high-pressure system. The temperature was first raised to 1000 °C at 10 °C/min and was held at this temperature for 60 h, followed by cooling to room temperature at 5 °C/min. The value of oxygen pressure was kept at 480 bar. X-ray powder diffraction characterization showed the presence of the Y124 phase with lattice parameters $a = 3.8369(6)$ Å, $b = 3.8666(6)$ Å, $c = 27.206(4)$ Å. A minor fraction of CuO was found in the spectra. However, this fraction must be below 3% as no change in the muon asymmetry was detected around 230 K, where CuO magnetically

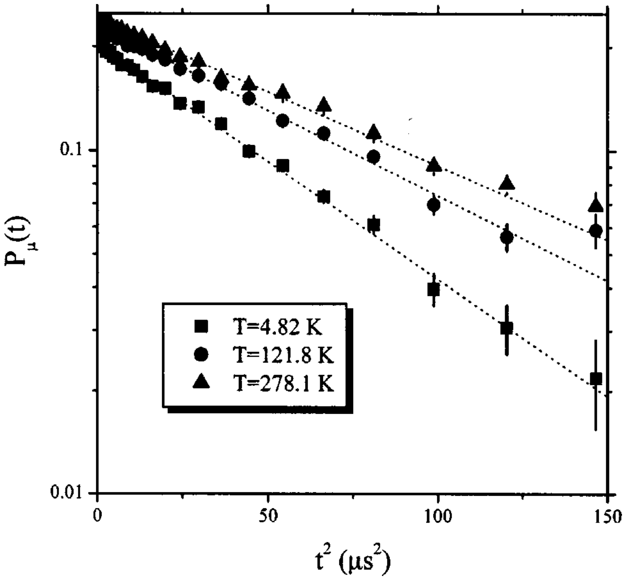

FIG. 1. Decay of the ZF μ^+ polarization in Y124 powder samples with natural abundant ^{63}Cu at a few selected temperatures. The decay is reported in semilog scale vs t^2 in order to show the accuracy of the fit (dotted lines) according to Eq. (1).

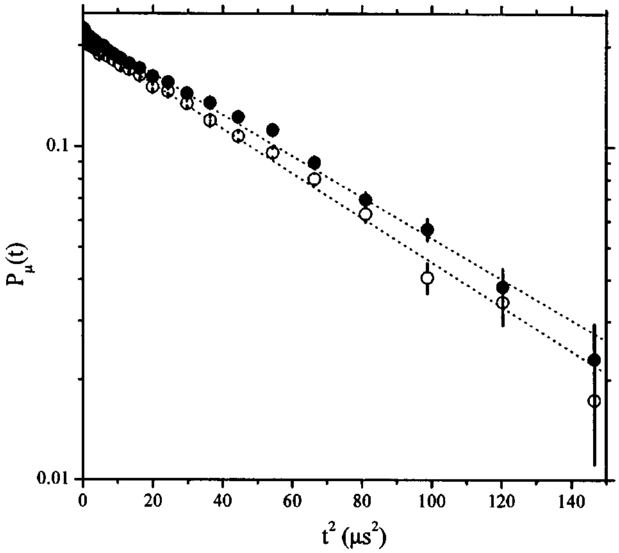

FIG. 2. Decay of the ZF μ^+ polarization at $T \simeq 20$ K for the ^{63}Cu-enriched (closed circles) and nonenriched (open circles) Y124 powder samples. The decay is reported in semilog scale vs t^2 in order to evidence the accuracy of the fit with Eq. (1).

orders.[8] The superconducting transition temperature T_c was estimated from the magnetization curves in a magnetic field of 10 G. It was found that $T_c = 80.2 \pm 0.2$ K for the isotope-enriched sample and $T_c = 82 \pm 0.2$ K for the natural one. Such a small difference in T_c will be irrelevant in the analysis of the μSR results (see Fig. 3).

ZF μSR measurements were performed on the MUSR instrument at the ISIS Pulsed Muon Facility, using 29 MeV/c spin-polarized muons. The use of an intense pulsed muon source at ISIS has the major advantage that allows to measure slow relaxation rates with the highest accuracy. The background signal due to the cryostat and sample holder was estimated from the slowly decaying part of the polarization in low-temperature transverse field measurements. During the ZF measurements an automatic compensation of the magnetic field was active in order to grant a magnetic field on the sample below a few tenths of mGauss. This is important to assure that below T_c there is no extra contribution to the relaxation associated with the trapping of the magnetic flux.

In Fig. 1 the ZF decay of the muon polarization for the ^{63}Cu-enriched sample is reported versus t^2 for a few selected temperatures. As one can notice, the form of the decay law is Gaussian below 12 μs and does not change upon cooling from about 200 K down to 4.2 K. In fact, the data can be nicely fit with a static Gaussian Kubo-Toyabe function,[9,10] the one theoretically expected when the relaxation is driven by nuclear moments,

$$P_\mu(t) = A(0)\left[\frac{1}{3} + \frac{2}{3}(1 - \sigma^2 t^2)\exp\left(-\frac{1}{2}\sigma^2 t^2\right)\right], \qquad (1)$$

where $A(0) \simeq 0.23$ is the initial asymmetry, $\sigma = \gamma\sqrt{\langle \Delta h^2 \rangle}$, with $\gamma = 2\pi \times 13.55$ kHz/Gauss the muon gyromagnetic ratio, and $\sqrt{\langle \Delta h^2 \rangle}$ the amplitude of the local-field distribution experienced by the muons. To second order in time t this function is identical to a Gaussian and the asymmetry decay plotted as $\log P_\mu(t)$ versus t^2 (Fig. 1) is given by a straight line. The decay of the muon polarization was observed to be faster in the sample with natural isotope abundance with respect to the ^{63}Cu-enriched samples (Fig. 2). The values derived for σ from Eq. (1) for both samples in the 300–4.2 K temperature range are finally reported in Fig. 3. A small decrease in the relaxation with increasing temperature is observed around 200 K and possibly associated with μ^+

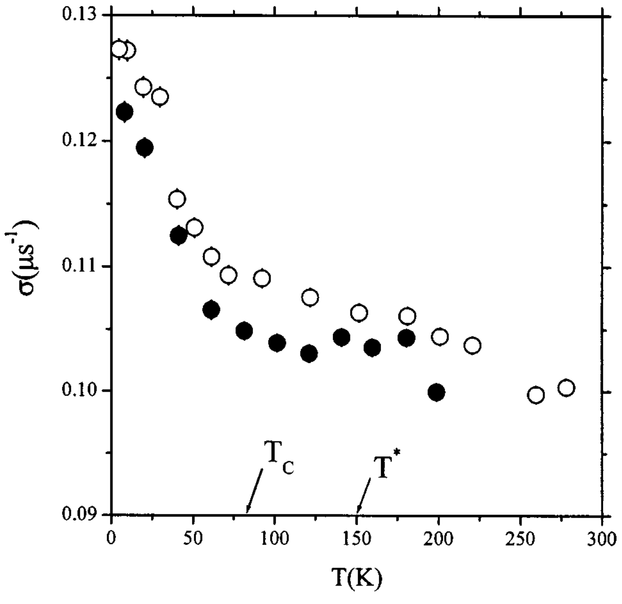

FIG. 3. Temperature dependence of the ZF μSR relaxation rate σ [see Eq. (1)] in isotope-enriched (closed circles) and nonenriched (open circles) Y124 powder samples.

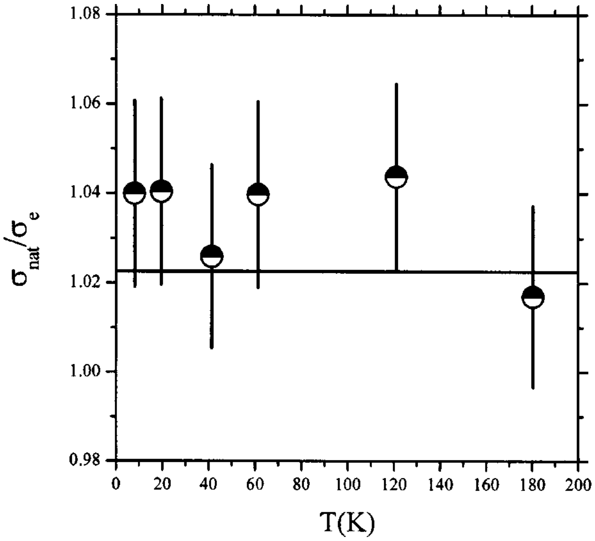

FIG. 4. Ratio of the ZF μSR relaxation rate σ in isotope-enriched and nonenriched Y124 powder samples. The solid line shows the theoretical value for this ratio calculated according to Eq. (2).

diffusion.[11] One notices that no anomaly is observed at $T^* \simeq 150$ K, definitely ruling out the occurrence of any increase in σ at T^*. However, a pronounced increase is clearly visible below T_c and is quantitatively similar to the one observed in optimally doped Y123 by Sonier *et al.*[3] for $T \simeq 40$ K.

III. DISCUSSION AND CONCLUSION

The dependence of σ on the ^{63}Cu isotope abundance is straightforward evidence that the ZF muon relaxation in Y124 is driven by the interaction with nuclear magnetic moments, over all the explored temperature range. In Y124 the dominant nuclear dipole interaction is the one with ^{63,65}Cu nuclei. Taking into account that the natural abundance is given by 69% of ^{63}Cu $(\gamma_{63}/2\pi = 11.285\ \mathrm{MHz/Tesla})$ and 31% of ^{65}Cu $(\gamma_{65}/2\pi = 12.089\ \mathrm{MHz/Tesla})$, the ratio of the second moment of the field distributions due to nuclear dipolar interaction in the enriched and natural samples should scale as[12]

$$\frac{\sigma_{nat}^2}{\sigma_e^2} = \left(0.31 \times \frac{\gamma_{65}^2}{\gamma_{63}^2} + 0.69\right) = 1.046, \qquad (2)$$

with σ_{nat} and σ_e the relaxation rates for the natural and enriched samples, respectively. One observes in Fig. 3 that the relaxation rate of the natural sample is slightly larger than the one of the isotope-enriched sample, as expected from Eq. (2). In Fig. 4 the ratio σ_{nat}/σ_e is reported for a few selected temperatures at which measurements in both samples were performed and a reasonable agreement with the ratio expected on the basis of nuclear dipole interaction is found.

In principle one could associate the increase in σ below 40 K with a crossover from a high-temperature regime, where μ^+ diffusion occurs, to a low-temperature one where the muon is localized, as observed in several metals.[12] However, this hypothesis is in conflict with the observation that the decay of the muon polarization is nicely fit with a static Kubo-Toyabe function over all the explored temperature range. Hence, the increase in the muon relaxation rate is not associated with a slowing down of the muon dynamics but rather to a modification in the field distribution probed by the muons.

A modification in the nuclear dipole interaction can occur only if the relative position between the muon and the nuclei changes or if the zero-field quantization axes of nuclear spins, corresponding to the electric-field gradient (EFG) principal axes, varies. A change in the μ^+ site or in the EFG principal axes can result from a variation of the crystal field, induced by a modification in the surrounding charge distribution. Also, Sonier *et al.*[4] have suggested a similar scenario for optimally doped Y123, after reconsidering the interpretation of their data.

In principle, one could associate the increase in σ with a lattice distortion. However, in the cuprates, although clear signs of a modification in the phonon spectra have been detected,[13] suggesting a strong electron-phonon coupling, no structural distortions have been observed at T_c. Moreover, it has to be remarked that in optimally doped Y123 the analogous increase in σ is detected well below T_c, pointing out that it cannot be directly related to the superconducting transition. On the other hand, a change in the μ^+ site or of the EFG orientation could arise from the onset of a charge order in the CuO chains. In $PrBa_2Cu_3O_7$ and Y123 Grevin *et al.*[14,15] have suggested on the basis of NQR measurements that a charge-density wave (CDW) order might set in. In Y124 the NQR results do not seem to support unambiguously such a scenario, although a clear anomaly in the NQR frequency of the chain copper was revealed at T_c,[16] suggesting a modification in the charge distribution within the chains. It is worth mentioning that the microscopic mechanism yielding the increase of σ does involve the chains since it is observed only in compounds with completely filled CuO chains, as optimally doped Y123 and Y124, while it is absent in compounds without chains as $La_{2-x}Sr_xCuO_4$.[17] Therefore, the increase in the μSR relaxation rate at low temperature in Y124 and optimally doped Y123 seems to signal a crossover from a high-temperature disordered arrangement of the charge distribution to a regime where at least short-range correlations in the charge density set in within the CuO chains.[18] Hence, it appears that the CuO chains in Y123 and Y124 could be characterized by an independent phenomenology, as if they were almost decoupled from the superconducting CuO_2 layers. In fact, a commonly accepted scenario for high-T_c superconductors is that CuO_2 planes are Josephson coupled across the CuO chains, which would play the role of a nonsuperconductive layer. This hypothesis is supported also by the observation of coexisting magnetic order and superconductivity in the adjacent CuO_2 and RuO layers of ruthenocuprate superconductors.[19]

In conclusion, from a careful analysis of the ZF μSR relaxation in isotope-enriched and natural Y124 powders we have observed an anomalous increase in the relaxation rate below T_c which has to be unambiguously associated with a change in the dipolar coupling with the nuclear spins, either due to a modification in the μ^+ site or in the EFG at the Cu nuclei. This modification suggests a common scenario for optimally doped Y123 and Y124, with a crossover to a low-temperature regime where the charge distribution within the CuO chains varies, possibly due to growing charge-density correlations.

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