Magnetic-field-enhanced spin freezing on the verge of charge ordering in YBa$_2$Cu$_3$O$_{6.45}$

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Using ^{63}Cu NMR, we establish that the enhancement of spin order by a magnetic field H in YBa$_2$Cu$_3$O$_{6.45}$ arises from a competition with superconductivity because the effect occurs for H perpendicular, but not parallel, to the CuO$_2$ planes, and it persists up to field values comparable to H_{c2}. We also find that the spin freezing has a glassy nature and that the frozen state onsets at a temperature which is independent of the magnitude of H. These results, together with the presence of a competing charge-ordering instability at nearby doping levels, are strikingly parallel to those previously obtained in La-214. This suggests a universal interpretation of magnetic field effects in underdoped cuprates where the enhancement of spin order by the field may not be the primary phenomenon but rather a byproduct of the competition between superconductivity and charge order. We also observe that low-energy spin fluctuations are manifested up to relatively high temperatures where they partially mask the signature of the pseudogap in $1/T_1$ data of planar Cu sites.

I. INTRODUCTION

One of the most debated issue in high T_c cuprates is whether the pseudogap, and perhaps superconductivity itself, are related to the occurrence of electronic ordering competing with superconductivity.[1] Why has this issue not been settled yet? First, despite a plethora of ordering phenomena reported in these materials, there are actually very few instances where a competition with superconductivity is unambiguously demonstrated. The strongest evidence comes from those experiments which have shown either (1) that a magnetic field promotes spin and/or charge ordering if and only if this field weakens significantly superconductivity,[2–6] or (2) that the occurrence of one order reduces the amplitude of its competitor.[7–10] Second, it has been unclear whether there is a single, generic, competing order behind the different phenomena observed. In particular, which of spin order or charge order would be the leading phenomenon in that case has been a central question.[11–14] These issues obviously need to be settled before discussing any possible relation to the mechanism of superconductivity.

Recently an important result was obtained in YBa$_2$Cu$_3$O$_{6.45}$[15,16] where an enhancement of spin order by a magnetic field was observed in a cuprate other than La-214.[2,3,17–24] A significant step for establishing the ubiquity of competing orders in cuprates would be to demonstrate that the field-induced magnetism in YBa$_2$Cu$_3$O$_{6.45}$ arises from the weakening of superconductivity by the field, rather than from a direct effect of the field itself.[16,25] More generally, the degree of similarity between these two different families (Fig. 1) is related to the central, but controversial, issue of universality in cuprates.[16,25–28]

Here we report ^{63}Cu nuclear magnetic resonance (NMR) results in YBa$_2$Cu$_3$O$_{6.45}$ untwinned single crystals from the same batch as those studied in neutron scattering.[15,16] Owing to the unique capability of NMR to be operated in any field orientation and in the highest achievable field strengths, our results demonstrate that the field indeed promotes a competing

order at the expense of superconductivity, as it does in La-214. We further uncover a number of other similarities with La-214 and we make the, previously unnoticed, observation that *all* of the existing evidence of field-induced effects in underdoped cuprates has been obtained in a specific doping range over which superconductivity competes with a charge ordering instability (Fig. 1). This leads us to suggest a universal interpretation of magnetic field effects in underdoped cuprates where the enhancement of spin order by the field is not the central phenomenon, but rather a byproduct of the competition between superconductivity and charge order.

II. SUPERCONDUCTING PROPERTIES

A sharp superconducting transition is observed in zero field at $T_c = 35$ K. In a field the melting temperature of the vortex lattice T_{melt} was measured through the resonance frequency of the NMR tank circuit and the obtained values [Fig. 3(d)] were in agreement with data in YBa$_2$Cu$_3$O$_y$ at a similar doping level.[29] T_{melt} decreases much more with increasing fields $H \parallel c$ than with $H \parallel ab$, as expected for this anisotropic superconductor [Fig. 3(d)]. It is important to realize that the relaxation peak reported below cannot be related to the solid to liquid transition of the vortex lattice: The central line broadening starts far above T_c. At 28.5 T it has reached 90% of its low T value before T_{melt} and it corresponds to an amplitude of field distribution which is about three time larger than that possibly due to the vortex lattice. The peak in $1/T_1$ manifestly corresponds to the spin freezing seen by neutron scattering or muon spin rotation (μSR) and the peak temperature is unrelated to T_{melt} which shows an opposite field dependence.

III. Cu2 AND Cu1 NMR SPECTRA

The presence of two distinct sites in the NMR spectra of planar Cu2 [Fig. 2(a)] reveals an intrinsic degree of ortho-II oxygen order in the chains.[30] Nonetheless, disorder related to oxygen vacancies in the oxygen-filled chains is observed,
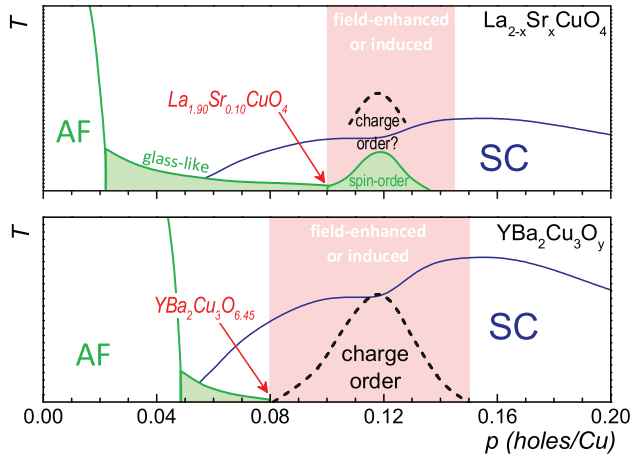
FIG. 1. (Color online) Phase diagrams of (a) $La_{2-x}Sr_xCuO_4$ and (b) $YBa_2Cu_3O_y$, based on data in Refs. 5, 6, 47, and 49–51. The shaded regions are those where field-tuned competing orders have been reported. The dotted line in (a) represents a putative charge-ordering transition which, although not directly observed, is suspected to be present by analogy with $La_{2-x}Ba_xCuO_4$ and $La_{2-y-x}Eu/Nd_ySr_xCuO_4$.[52] The boundary of the (field-induced) charge-ordered state in (b) is based on an interpretation of transport measurements.[5,47,50]

as expected from the actual doping level $y \simeq 6.45$ (hole content $p \simeq 0.07$–0.08) being lower than the ideal ortho-II composition $y = 6.50$: there are three different Cu1E (empty chains) sites and two Cu1F (oxygen-filled chains) sites instead of one Cu1E and one Cu1F for $y = 6.50$. These multiple sites correspond to the different positions with respect to the oxygen vacancies.[31]

The signal from Cu2 sites can be studied from room temperature down to ~50 K and we show in the Appendix that such measurements provide evidence for low-energy magnetic fluctuations masking the signature of the pseudogap in the spin-lattice relaxation rate ($1/T_1$) data of Cu2 so that no decrease of the magnitude of the pseudogap should be deduced from these measurements. Below 50 K, however, the only reliable NMR signal is that from ^{63}Cu1E sites which, unlike planar Cu2, is not wiped out by too fast relaxation times as the magnetic transition is approached (see Appendix).

The strong broadening [Fig. 2(b)] of the Cu1E line shows that a distribution of hyperfine fields $\langle h_z \rangle = gA\langle S_z \rangle$ develops on cooling in the CuO_2 planes, most likely due to oxygen disorder in the chains impacting onto the planes[32] ($A = 0.3$ T/μ_B[33] is the hyperfine coupling to the CuO_2 planes and g is Landé's factor). At 1.5 K and 28.5 T the value of $\delta\nu$ translates into a distribution of spin polarization of $\delta\langle S_z \rangle \simeq \pm 3 \times 10^{-2}\,\mu_B$.

IV. GLASSY SPIN FREEZING

The time dependence of the Cu1E magnetization after saturating the central line (where the three sites are unresolved) was fit to $m(t) = m_\infty\{1 - 0.1\exp[-(t/T_1^{\text{mean}})^\beta] - 0.9\exp[-(6t/T_1^{\text{mean}})^\beta]\}$ corresponding to a distribution of $1/T_1$ values whose width in a log scale is linear in β between $\beta = 1$ (i.e., a homogeneous system) and $\beta \simeq 0.5$.[34,35] $1/T_1^{\text{mean}}$ is the most probable value of the distribution, and is

approximately satisfying the standard definition of T_1, namely the time to reduce the perturbation by $1/e$, independently of the β value. This makes the fitted $1/T_1^{\text{mean}}$ values stable and mostly decoupled from β. The parametrization of the T or H dependence of $1/T_1^{\text{mean}}$ with models developed for homogeneous situations (like the BPP model below) thus provides a valuable insight into the intrinsic, *most probable*, behavior of the system, in particular as much as the corresponding β variations are not substantial over the relevant range (while those of $1/T_1^{\text{mean}}$ are).

The increase of $1/T_1^{\text{mean}}$ is evidence that the spin fluctuations slow down on cooling (i.e., their correlation time τ_c diverges), as expected from earlier evidence of spin ordering.[15,16,26] In agreement with the above-noted distribution of $\langle S_z \rangle$, the decrease of β [Figs. 3(a) and 3(b)] shows that heterogeneity develops on cooling along with the slowing down. The possible origin of this glassy dynamics has been addressed by many authors (Refs. 16, 27, 35, 38, and 39 and references therein). This discussion is beyond the scope of the present work but the important point here is that the very same glassy freezing is observed in La-214 cuprates.

The peak of $1/T_1^{\text{mean}}$, observed here at $T_{\text{spin}}^{\omega_{\text{NMR}}} \simeq 4$–$6$ K, is expected when the characteristic frequency of spin fluctuations becomes as small as the NMR frequency ω_{NMR}. As in previous studies of La-214,[36] the T dependence of $1/T_1^{\text{mean}}$ can be accounted for by the "BPP" formula[37] $\frac{1}{T_1} = \gamma_n^2 \langle h_\perp^2 \rangle \frac{2\tau_c}{1+\omega_{\text{NMR}}^2\tau_c^2}$, where $h_\perp$ is the component of the hyperfine field perpendicular to H and γ_n is the nuclear gyromagnetic ratio. Since we are analyzing $1/T_1^{\text{mean}}$ here, τ_c represents the most probable correlation time of the fluctuating spin system.

V. PROBE-FREQUENCY DEPENDENCE

The values of $T_{\text{spin}}^{\omega_{\text{NMR}}} = 4$–$6$ K are much lower than $T_{\text{spin}}^{\text{NS}} \simeq 30$ K, the temperature at which an elastic signal appears in neutron scattering. As shown by numerous studies of glasses, it is the broad temperature range of inhomogeneous slowing down which makes the apparent freezing temperature T_{spin} probe-frequency dependent. The difference of temperatures becomes significant here because the time scale of the neutron scattering experiment (100 μeV resolution here) is a hundred times faster than the typical NMR time scale of $10^{-2}\,\mu$s. This shows that the closeness of the values of $T_{\text{spin}}^{\text{NS}} \simeq 30$ K and $T_c = 35$ K is essentially fortuitous: Spin order does not occur near T_c but at much lower temperature. Moreover, μSR experiments with a time scale of $10\,\mu$s (thus slower than NMR) detect static fields below $T_{\text{spin}}^{\mu} \simeq 2$ K in this sample,[26] which is logically lower than $T_{\text{spin}}^{\omega_{\text{NMR}}} = 4$–$6$ K and is close to $T_{\text{spin}}^{\omega=0} = 2 \pm 2$ K, the freezing temperature extrapolated down to zero frequency with the BPP formula [Fig. 3(d)]. This probe-frequency dependence of T_{spin} is another striking similarity with the La-214 materials.

VI. EVIDENCE OF COMPETITION WITH SUPERCONDUCTIVITY

As Fig. 3(a) shows, the amplitude of $1/T_1$ depends strongly on field up to (at least) 28.5 T for $H\|c$. Since $(1/T_1)^{\text{max}} = \gamma_n^2\langle h_\perp^2 \rangle/\omega_{\text{NMR}}$ at $T = T_{\text{spin}}^{\omega_{\text{NMR}}}$, the mean-squared fluctuating hyperfine field $\langle h_\perp^2 \rangle$ can be extracted at this temperature without supposing any T dependence for τ_c. It appears that
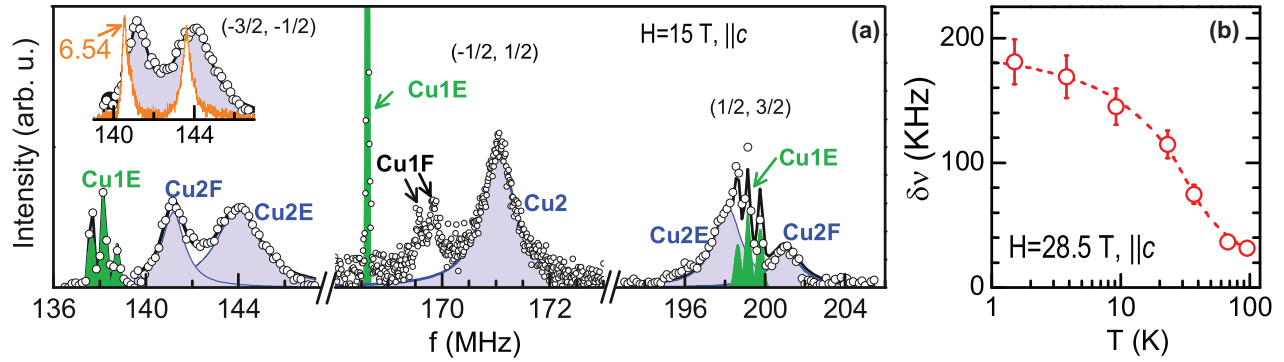
FIG. 2. (Color online) (a) ^{63}Cu NMR spectrum ($T = 60$ K) with quadrupole "satellites" ($\pm 3/2, \pm 1/2$ transitions) and central lines ($+1/2, -1/2$). The line assignment is made on the basis of previous NMR works in YBa$_2$Cu$_3$O$_y$. Note that "Cu2E" is an approximate naming here because, when the oxygen content is lower than $y = 6.5$, this line most likely also contains those Cu2F which lie below chain-oxygen vacancies.[31] This explains why the width and integrated area of this Cu2E line are larger than for Cu2F. Left inset: Comparison with Cu2 satellites of YBa$_2$Cu$_3$O$_{6.54}$.[5] (b) Full-width-at-half-maximum $\delta\nu$ (dashes are a guide to the eye) of the Cu1E central line. The shift of this line is negligible with respect to $\delta\nu$.

$\langle h_{\perp}^2 \rangle$ strongly varies with field [Fig. 3(b)]. Furthermore, whatever the supposed form of $\tau_c(T)$, fits in Fig. 3(a) show that the $1/T_1$ data are well described by a BPP model without any field dependence of τ_c. Of course the fits could be improved (especially at low fields) with a field-dependent τ_c but this is a minor correction which does not affect the results of this paper and the main message here is that field dependence of $1/T_1$ is, to a first approximation, entirely explained by the field dependence of $\langle h_{ab}^2 \rangle$. This field dependence should naturally be related to the observed enhancement of I_{NS} by the field.[15,16] Moreover, a similar field enhancement of T_1 is observed at low T in La$_{1.90}$Sr$_{0.10}$CuO$_4$[40] for which the quasielastic neutron scattering intensity I_{NS} (in general proportional to the square of the ordered moment) is also field dependent.[2]

The most direct evidence for competing orders, and therefore the most important result of this work, is the absence of a peak in $1/T_1$ when the field of 28.5 T is applied parallel to the CuO$_2$ planes. Since the peak must exist for any field orientation (frozen moments of $\sim 0.1 \mu_B$ are detected in μSR below 2 K, already for $H = 0$[26,51]), $\langle h_{\perp}^2 \rangle$ has to be smaller by more than an order of magnitude for $H \| ab$ than for $H \| c$ in order to make $1/T_1$ so small that no peak is seen above the relaxation background. The striking difference between $H \| c$ and $H \| ab$ cannot be explained by an extreme anisotropy of magnetic fluctuations $\langle h_c^2 \rangle \ll \langle h_{ab}^2 \rangle$ because $\langle h_{ab}^2 \rangle$ also contributes to $\langle h_{\perp}^2 \rangle = \frac{1}{2}(\langle h_c^2 \rangle + \langle h_{ab}^2 \rangle)$ when $H \| ab$. Instead, this result demonstrates that the enhancement of $1/T_1$ is not related to the field itself but it is due to the weakening of superconductivity which is indeed much more efficient for $H \| c$ than for $H \| ab$ [Fig. 3(d)]. This is precisely the phenomenology expected for competing orders. In this scenario, no field dependence should exist above the upper critical field $H_{c2}(\| c) \simeq 40 \pm 5$ T for this doping level.[29,41,42] Although this limit could not be reached here, our measurements probing the spin order for the first time in very high fields show that the enhancement of magnetism seems to occur on a field scale similar to that for the destruction of superconductivity, with $\langle h_{ab}^2 \rangle$ varying as $a_1 + a_2[H/(H_c - H)]$ and $H_c \simeq 39 \pm 1$ T (the use of this dependence has no other justification than introducing a field-scale H_c).

Studies in La$_{2-x}$Sr$_x$CuO$_4$[22,23,43] and YBa$_2$Cu$_3$O$_y$[43] have suggested that the field tunes the relative volume fractions of the superconducting and magnetic phases, consistent with the idea that the order competing with superconductivity is enhanced in and about the vortex cores. This two-phase description might also rationalize the counterintuitive observation that the neutron scattering intensity in La$_{1.90}$Sr$_{0.10}$CuO$_4$ has a field-independent onset despite its field-dependent amplitude.[2] Our data in YBa$_2$Cu$_3$O$_{6.45}$ also support a field-independent T_{spin}. Indeed, in the BPP description, the peak temperature $T_{spin}^{\omega_{NMR}}$ is an apparent freezing temperature which depends on ω_{NMR}. In a standard NMR experiment, however, ω_{NMR} increases linearly with field. Therefore, $T_{spin}^{\omega_{NMR}}$ necessarily depends on field in NMR, although it would be field independent for any experimental probe operating at a fixed frequency ω_0. The excellent fit to the peak positions in Fig. 3(a) (with no field dependent parameter other than the amplitude $\langle h_{ab}^2 \rangle$) demonstrates that $T_{spin}^{\omega_{NMR}}$ values follow the H dependence expected from the trivial frequency dependence of the BPP model [Fig. 3(d)]. This implies that $T_{spin}^{\omega_{NMR}}$ would indeed be field independent if H could be varied without changing ω_{NMR}.

VII. DISCUSSION

In addition to demonstrating the existence of competing orders, our results have revealed striking analogies between YBa$_2$Cu$_3$O$_{6.45}$ and La-214 materials: similar glassy spin freezing, field-independent onset T_{spin}, enhancement of magnetism for $H \| c$.

In La$_{1.90}$Sr$_{0.10}$CuO$_4$ it is generally admitted[12,13,24] that the enhancement of spin order by the field actually occurs because incipient charge order[44] competes with superconductivity (and is thus field dependent). Namely, it is this charge order which, in turn, boosts spin order. This explanation does not suppose anything regarding the origin of spin order in zero field, which could well be triggered by quenched disorder rather than by charge order.[2,45,46]

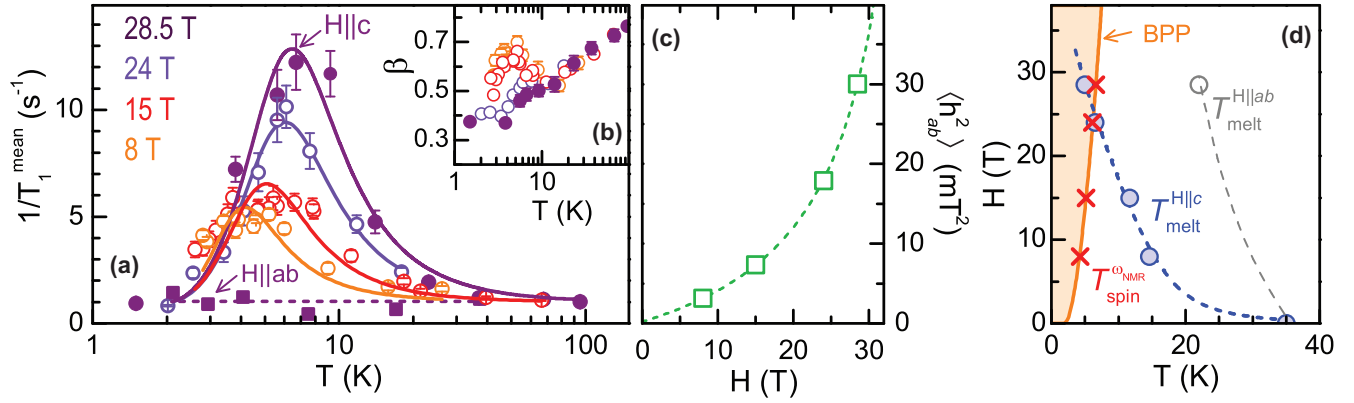As Fig. 1 shows, despite its different doping level, YBa$_2$Cu$_3$O$_{6.45}$ ($p \simeq 0.075$) lies in a similar position in the

FIG. 3. (Color online) (a) $1/T_1^{\mathrm{mean}}$ of Cu1E for $H\|c$ (except open circles for $H\|ab$). Lines are fits to the BPP formula assuming a field-independent $\tau_c = 0.042/(T-2)^{1.7}$,[36] where the numerical values were determined from fits to the data at 24 T. The only adjustable parameter between different fields is thus the amplitude factor $\langle h_\perp^2 \rangle = \langle h_{ab}^2 \rangle$. (b) Stretching exponent β with same color code as in (a). (c) Mean-squared hyperfine field $\langle h_\perp^2 \rangle = \langle h_{ab}^2 \rangle$ at $T = T_{\mathrm{spin}}^{\omega_{\mathrm{NMR}}}$. (d) Spin-freezing temperature $T_{\mathrm{spin}}^{\omega_{\mathrm{NMR}}}$ deduced from the peaks of the T_1 fits (following by construction the field dependence of the BBP model) and vortex-melting temperature T_{melt} deduced from the detuning of the NMR tank circuit.

phase diagram as $\mathrm{La_{1.90}Sr_{0.10}CuO_4}$, i.e., at the border between the glasslike phase and the region of field-dependent charge order. This leads us to propose the same description in both compounds: Regardless of the (debated) origin of the (incommensurate) spin order in zero field[28] for $\mathrm{YBa_2Cu_3O_{6.45}}$, its enhancement by a field does not necessarily imply a mere competition between superconductivity and spin order. Instead, the field-dependent spin order could be a more subtle byproduct of the recently discovered competition between superconductivity and a charge ordering instability at nearby doping levels.[5,6,8–10,47] Of course, a direct evidence of charge-density-wave correlations in $\mathrm{YBa_2Cu_3O_{6.45}}$ would be desirable but, as far as Cu NMR is concerned, disorder arising from the CuO chains and Cu(2) signal wipeout preclude the observation of a quadrupole splitting or broadening of the lines which would indicate a static charge density modulation in this sample. From this NMR point of view, the situation recalls more La-214 than $\mathrm{YBa_2Cu_3O_{6.6\pm0.1}}$. Still, there is a real possibility that a charge instability persists down to this doping level.[8,26,48,50] although it may be difficult to detect because higher oxygen disorder and stronger scattering from spin fluctuations could result in lower intensity and shorter correlation length.

In a broader perspective, the following, so far unnoticed, observation can be made: To the best of our knowledge, *all* of the field-induced effects (shaded areas in Fig. 1) were actually observed at doping levels around $p \simeq 0.12$, where a charge-ordering instability manifestly competes with superconductivity.[5–10] Likewise, no effect of the field has ever been reported in the "glasslike" region of the phase diagram in Fig. 1 (see particularly Ref. 16 for $\mathrm{YBa_2Cu_3O_{6.35}}$). It is therefore possible that all of the observations of field-enhanced spin order in cuprates are actually explained by a competition between superconductivity and charge order.

Note added: Recently, several related papers appeared. Spin freezing has been observed with NMR in $\mathrm{YBa_2Cu_3O_{6.45}}$ but its dependence on the magnitude and on the orientation of the field was not studied.[65] Two x-ray scattering studies in $\mathrm{YBa_2Cu_3O_{6.54}}$ (ortho-II)[66,67] report a charge ordering

wave vector unrelated to that of the spin modulation in $\mathrm{YBa_2Cu_3O_{6.45}}$. These results prompt for further investigation of charge correlations in $\mathrm{YBa_2Cu_3O_{6.45}}$ and of field effects on spin order. With these results and ours, the question of universality and competing orders is more than ever center stage in cuprates.

APPENDIX

1. Samples and methods

The typical size of the untwinned, high quality, single crystals was $1.7 \times 1.6 \times 0.4$ mm^3 with a sharp superconducting transition (Fig. 4). NMR measurements at 24 and 28.5 T were performed in the M10 resistive magnet of the LNCMI Grenoble. All measurements for both Cu1 and Cu2 sites were performed on the ^{63}Cu isotope. Spin-lattice relaxation times (T_1) were measured by saturating the central transitions.

2. Electronic transition at $p \simeq 0.08$

$1/(T_1T)$ data for ^{63}Cu2 [a measure of the dynamic spin susceptibility $\chi''(q,\omega_n)$ at the NMR frequency ω_n] show a significant difference with data obtained at higher doping level in underdoped $\mathrm{YBa_2Cu_3O_y}$: The values are much larger over the whole temperature range and a broad maximum occurs around $T^\dagger \simeq 70$ K [Fig. 5(a)] instead of the usual $T^\dagger \simeq 150$ K at higher doping.[53] We cannot exclude that no $T^\dagger$ would even be observed in this sample if no wipeout occurred. Anyhow,
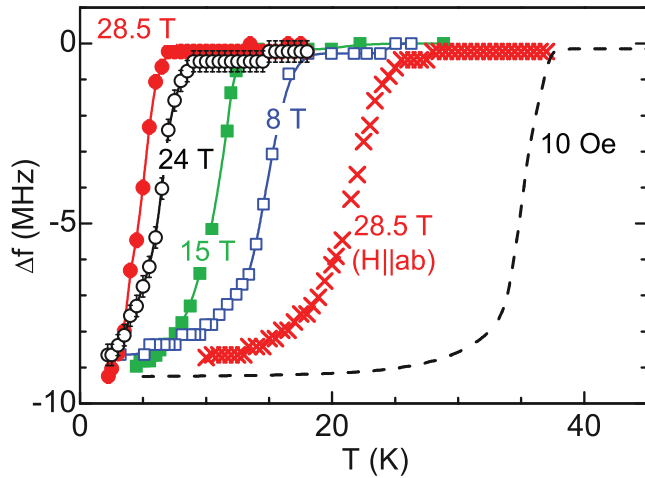
FIG. 4. (Color online) Frequency shift of the resonance of the NMR tank circuit (symbols) and bulk magnetization measurements in a field of 10 Oe (dashed line, scaled to the frequency shift data). When not specified, the field is applied parallel to the c axis.

FIG. 5. (Color online) (a) $1/(T_1T)$ in $H\|c = 15$ T, measured on the central line of $^{63}Cu2$ nuclei. The data are in agreement with an earlier study at similar doping level.[53] (b) Central-line intensity integrated over frequency and corrected for the T_2 decay. Unlike Cu2, the Cu1E signal does not suffer any wipeout, thus allowing reliable measurements at low T. Open (closed) symbols in (b) correspond to $H = 15$ (28.5) T.

a decrease of $T^\dagger$ for $p \leqslant 0.08$ has already been reported in YBa$_2$Cu$_3$O$_y$[53] as well as in three- and five-layer cuprates near $p \simeq 0.09$.[54] Since the maximum of $1/(T_1T)$ at $T^\dagger > T_c$ is typical of the pseudogap phase, it would be tempting to associate the lower $T^\dagger$ to a decrease in the magnitude of the pseudogap for $p \leqslant 0.08$. However, $T^\dagger$ is significantly smaller than T^* defining the onset of the pseudogap in the uniform spin susceptibility[55] or in the resistivity[56] and T^* does not decrease below $y = 6.5$.[55,56] Also, nonmagnetic impurities do not affect T^*[57] while they lead to a decrease of $T^\dagger$ in some T_1 measurements.[58] Actually, $k_B T^\dagger$ may not represent any characteristic energy scale, especially given the complex nature of the processes involved in nuclear relaxation. Thus,
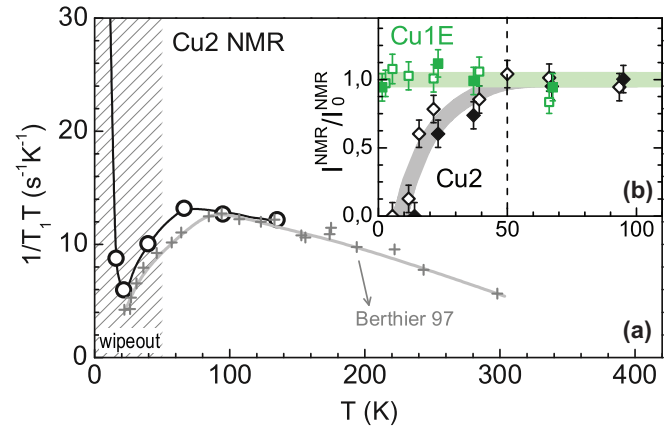
the data cannot be taken as evidence of a decrease in the pseudogap energy scale below $p \simeq 0.08$. Instead, the decrease of $T^\dagger$ likely arises from the spin fluctuations which must be present at energy much lower than $k_B T^*$, and thus fill the pseudogap, on approaching T_{spin}. Note that this statement does not conflict with the well-documented evidence for a crossover (or transition) from a magnetic, disorder-sensitive, 2D anisotropic, bad metal to a spin-gapped 3D metallic ground state near $p \simeq 0.08$ in YBa$_2$Cu$_3$O$_x$.[48,50,51,59–64] It simply means that this phenomenon is not directly related to the pseudogap.

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[1] S. Sachdev and S.-C. Zhang, Science **295**, 452 (2002); M. R. Norman, D. Pines, and C. Kallin, Adv. Phys. **54**, 715 (2005).

[2] B. Lake, H. M. Rønnow, N. B. Christensen, G. Aeppli, K. Lefmann, D. F. McMorrow, P. Vorderwisch, P. Smeibidl, N. Mangkorntong, T. Sasagawa, M. Nohara, H. Takagi, and T. E. Mason, Nature (London) **415**, 299 (2002).

[3] B. Lake, K. Lefmann, N. B. Christensen, G. Aeppli, D. F. McMorrow, H. M. Rønnow, P. Vorderwisch, P. Smeibidl, N. Mangkorntong, T. Sasagawa, M. Nohara, and H. Takagi, Nat. Mater. **4**, 658 (2005).

[4] J. E. Hoffman, E. W. Hudson, K. M. Lang, V. Madhavan, H. Eisaki, S. Uchida, and J. C. Davis, Science **295**, 466 (2002).

[5] T. Wu, H. Mayaffre, S. Krämer, M. Horvatić, C. Berthier, W. N. Hardy, R. Liang, D. A. Bonn, and M.-H. Julien, Nature (London) **477**, 191 (2011).

[6] T. Wu, H. Mayaffre, S. Krämer, M. Horvatić, C. Berthier, P. L. Kuhns, A. P. Reyes, R. Liang, W. N. Hardy, D. A. Bonn, and M.-H. Julien, Nat. Commun. **4**, 2113 (2013).

[7] J. M. Tranquada, J. M. Tranquada, B. J. Sternlieb, J. D. Axe, Y. Nakamura, and S. Uchida, Nature (London) **375**, 561 (1995).

[8] G. Ghiringhelli, M. Le Tacon, M. Minola, S. Blanco-Canosa, C. Mazzoli, N. B. Brookes, G. M. De Luca, A. Frano, D. G. Hawthorn, F. He, T. Loew, M. M. Sala, D. C. Peets, M. Salluzzo, E. Schierle, R. Sutarto, G. A. Sawatzky, E. Weschke, B. Keimer, and L. Braicovich, Science **337**, 821 (2012).

[9] A. J. Achkar, R. Sutarto, X. Mao, F. He, A. Frano, S. Blanco-Canosa, M. Le Tacon, G. Ghiringhelli, L. Braicovich, M. Minola, M. Moretti Sala, C. Mazzoli, R. Liang, D. A. Bonn, W. N. Hardy, B. Keimer, G. A. Sawatzky, and D. G. Hawthorn, Phys. Rev. Lett. **109**, 167001 (2012).

[10] J. Chang, E. Blackburn, A. T. Holmes, N. B. Christensen, J. Larsen, J. Mesot, R. Liang, D. A. Bonn, W. N. Hardy, A. Watenphul, M. v. Zimmermann, E. M. Forgan, and S. M. Hayden, Nat. Phys. **8**, 871 (2012).

[11] D. P. Arovas, A. J. Berlinsky, C. Kallin, and S.-C. Zhang, Phys. Rev. Lett. **79**, 2871 (1997).

[12] Y. Zhang, E. Demler, and S. Sachdev, Phys. Rev. B **66**, 094501 (2002); S. Sachdev and E. Demler, ibid. **69**, 144504 (2004).

[13] S. A. Kivelson, D.-H. Lee, E. Fradkin, and V. Oganesyan, Phys. Rev. B **66**, 144516 (2002).

[14] E. G. Moon and S. Sachdev, Phys. Rev. B **80**, 035117 (2009).

[15] D. Haug, V. Hinkov, A. Suchaneck, D. S. Inosov, N. B. Christensen, C. Niedermayer, P. Bourges, Y. Sidis, J. T. Park, A. Ivanov, C. T. Lin, J. Mesot, and B. Keimer, Phys. Rev. Lett. **103**, 017001 (2009).

[16] D. Haug, V. Hinkov, Y. Sidis, P. Bourges, N. B. Christensen, A. Ivanov, T. Keller, C. T. Lin, and B. Keimer, New J. Phys. **12**, 105006 (2010).

[17] S. Katano, M. Sato, K. Yamada, T. Suzuki, and T. Fukase, Phys. Rev. B **62**, R14677 (2000).

[18] B. Khaykovich, S. Wakimoto, R. J. Birgeneau, M. A. Kastner, Y. S. Lee, P. Smeibidl, P. Vorderwisch, and K. Yamada, Phys. Rev. B **71**, 220508(R) (2005).

[19] J. Chang, Ch. Niedermayer, R. Gilardi, N. B. Christensen, H. M. Rønnow, D. F. McMorrow, M. Ay, J. Stahn, O. Sobolev, A. Hiess, S. Pailhes, C. Baines, N. Momono, M. Oda, M. Ido, and J. Mesot, Phys. Rev. B **78**, 104525 (2008).

[20] J. Chang, N. B. Christensen, C. Niedermayer, K. Lefmann, H. M. Ronnow, D. F. McMorrow, A. Schneidewind, P. Link, A. Hiess, M. Boehm, R. Mottl, S. Pailhes, N. Momono, M. Oda, M. Ido, and J. Mesot, Phys. Rev. Lett. **102**, 177006 (2009).

[21] M. Fujita, M. Matsuda, H. Goka, T. Adachi, Y. Koike, and K. Yamada, J. Phys: Conf. Series **51**, 510 (2006).

[22] A. T. Savici, A. Fukaya, I. M. Gat-Malureanu, T. Ito, P. L. Russo, Y. J. Uemura, C. R. Wiebe, P. P. Kyriakou, G. J. MacDougall, M. T. Rovers, G. M. Luke, K. M. Kojima, M. Goto, S. Uchida, R. Kadono, K. Yamada, S. Tajima, T. Masui, H. Eisaki, N. Kaneko, M. Greven, and G. D. Gu, Phys. Rev. Lett. **95**, 157001 (2005).

[23] L. H. Machtoub, B. Keimer, and K. Yamada, Phys. Rev. Lett. **94**, 107009 (2005).

[24] J. Wen Q. Jie, Q. Li, M. Hücker, M. v. Zimmermann, S. J. Han, Z. Xu, D. K. Singh, L. Zhang, G. Gu, and J. M. Tranquada, Phys. Rev. B **85**, 134513 (2012).

[25] C. Stock, W. J. L. Buyers, K. C. Rule, J.-H. Chung, R. Liang, D. Bonn, and W. N. Hardy, Phys. Rev. B **79**, 184514 (2009).

[26] V. Hinkov, D. Haug, B. Fauqué, P. Bourges, Y. Sidis, A. Ivanov, C. Bernhard, C. T. Lin, and B. Keimer, Science **319**, 597 (2008).

[27] C. Stock, W. J. L. Buyers, Z. Yamani, C. L. Broholm, J.-H. Chung, Z. Tun, R. Liang, D. Bonn, W. N. Hardy, and R. J. Birgeneau, Phys. Rev. B **73**, 100504(R) (2006); C. Stock, W. J. L. Buyers, Z. Yamani, Z. Tun, R. J. Birgeneau, R. Liang, D. Bonn, and W. N. Hardy, *ibid.* **77**, 104513 (2008).

[28] A. I. Milstein and O. P. Sushkov, Phys. Rev. B **78**, 014501 (2008); C.-P. Chou, N. Fukushima, and T. K. Lee, *ibid.* **78**, 134530 (2008); H. Yamase, *ibid.* **79**, 052501 (2009); K. Sun, M. J. Lawler, and E.-A. Kim, Phys. Rev. Lett. **104**, 106405 (2010); M. Vojta, Eur. Phys. J. Special Topics **188**, 49 (2010); M. A. Metlitski and S. Sachdev, New J. Phys. **12**, 105007 (2010); G. Seibold, M. Capati, C. Di Castro, M. Grilli, and J. Lorenzana, Phys. Rev. B **87**, 035138 (2013).

[29] B. J. Ramshaw, J. Day, B. Vignolle, D. LeBoeuf, P. Dosanjh, C. Proust, L. Taillefer, R. Liang, W. N. Hardy, and D. A. Bonn, Phys. Rev. B **86**, 174501 (2012).

[30] N. H. Andersen, M. von Zimmermann, T. Frello, M. Käll, D. Mønster, P.-A. Lindgård, J. Madsen, T. Niemöller, H. F. Poulsen, O. Schmidt, J. R. Schneider, Th. Wolf, P. Dosanjh, R. Liang, and W. N. Hardy, Physica (Amsterdam) **317C**, 259 (1999).

[31] T. Wu *et al.* (unpublished).

[32] W. Chen and P. J. Hirschfeld, Phys. Rev. B **79**, 064522 (2009).

[33] A. V. Dooglav, H. Alloul, M. V. Eremin, and A. G. Volodin, Physica (Amsterdam) **272C**, 242 (1996).

[34] D. C. Johnston, Phys. Rev. B **74**, 184430 (2006).

[35] V. F. Mitrović, M.-H. Julien, C. de Vaulx, M. Horvatić, C. Berthier, T. Suzuki, and K. Yamada, Phys. Rev. B **78**, 014504 (2008).

[36] B. J. Suh, P. C. Hammel, M. Hücker, B. Büchner, U. Ammerahl, and A. Revcolevschi, Phys. Rev. B **61**, R9265 (2000); N. J. Curro, P. C. Hammel, B. J. Suh, M. Hücker, B. Büchner, U. Ammerahl, and A. Revcolevschi, Phys. Rev. Lett. **85**, 642 (2000); M.-H. Julien, A. Campana, A. Rigamonti, P. Carretta, F. Borsa, P. Kuhns, A. P. Reyes, W. G. Moulton, M. Horvatić, C. Berthier, A. Vietkin, and A. Revcolevschi, Phys. Rev. B **63**, 144508 (2001); A. W. Hunt, P. M. Singer, A. F. Cederström, and T. Imai, *ibid.* **64**, 134525 (2001); G. B. Teitel'baum, I. M. Abu-Shiekah, O. Bakharev, H. B. Brom, and J. Zaanen, *ibid.* **63**, 020507(R) (2000); B. Simovič, P. C. Hammel, M. Hücker, B. Büchner, and A. Revcolevschi, *ibid.* **68**, 012415 (2003); H.-J. Grafe, N. J. Curro, M. Hücker, and B. Büchner, Phys. Rev. Lett **96**, 017002 (2006).

[37] C. P. Slichter, *Principles of Magnetic Resonance* (Springer, New York, 1990).

[38] I. Sega and P. Prelovšek, Phys. Rev. B **79**, 140504(R) (2009).

[39] S. A. Kivelson and V. J. Emery, Selected Topics Superconduct. **8**, 91 (2002).

[40] M.-H. Julien *et al.* (unpublished).

[41] Y. Ando and K. Segawa, Phys. Rev. Lett. **88**, 167005 (2002).

[42] J. Chang, N. Doiron-Leyraud, O. Cyr-Choinière, G. Grissonnanche, F. Laliberté, E. Hassinger, J-Ph. Reid, R. Daou, S. Pyon, T. Takayama, H. Takagi, and Louis Taillefer, Nat. Phys. **8**, 751 (2012).

[43] J. E. Sonier, F. D. Callaghan, Y. Ando, R. F. Kiefl, J. H. Brewer, C. V. Kaiser, V. Pacradouni, S. A. Sabok-Sayr, X. F. Sun, S. Komiya, W. N. Hardy, D. A. Bonn, and R. Liang, Phys. Rev. B **76**, 064522 (2007).

[44] H.-H. Wu, M. Buchholz, C. Trabant, C. F. Chang, A. C. Komarek, F. Heigl, M.v. Zimmermann, M. Cwik, F. Nakamura, M. Braden, and C. Schüßler-Langeheine, Nat. Commun. **3**, 1023 (2012).

[45] T. Sasagawa, P. K. Mang, O. P. Vajk, A. Kapitulnik, and M. Greven, Phys. Rev. B **66**, 184512 (2002).

[46] B. M. Andersen, M. Schmid, S. Graser, P. J. Hirschfeld, and A. P. Kampf, J. Phys. Chem. Solids **72**, 358 (2011).

[47] F. Laliberté, J. Chang, N. Doiron-Leyraud, E. Hassinger, R. Daou, M. Rondeau, B. J. Ramshaw, R. Liang, D. A. Bonn, W. N. Hardy, S. Pyon, T. Takayama, H. Takagi, I. Sheikin, L. Malone, C. Proust, K. Behnia, and L. Taillefer, Nat. Commun. **2**, 432 (2011).

[48] Y. Ando, K. Segawa, S. Komiya, and A. N. Lavrov, Phys. Rev. Lett. **88**, 137005 (2002).

[49] M.-H. Julien, Physica B **329–333**, 693 (2003).

[50] D. LeBoeuf, N. Doiron-Leyraud, B. Vignolle, M. Sutherland, B. J. Ramshaw, J. Levallois, R. Daou, F. Laliberte, O. Cyr-Choinière, J. Chang, Y. J. Jo, L. Balicas, R. Liang, D. A. Bonn, W. N. Hardy, C. Proust, and L. Taillefer, Phys. Rev. B **83**, 054506 (2011).

[51] R. F. Kiefl, J. H. Brewer, J. Carolan, P. Dosanjh, W. N. Hardy, R. Kadono, J. R. Kempton, R. Krahn, P. Schleger, B. X. Yang, Hu Zhou, G. M. Luke, B. Sternlieb, Y. J. Uemura, W. J. Kossler, X. H. Yu, E. J. Ansaldo, H. Takagi, S. Uchida, and C. L. Seaman, Phys. Rev. Lett. **63**, 2136 (1989); S. Sanna, G. Allodi, G. Concas, A. D. Hillier, and R. De Renzi, *ibid.* **93**, 207001 (2004); R. I. Miller, R. F. Kiefl, J. H. Brewer, F. D. Callaghan, J. E. Sonier, R. Liang, D. A. Bonn, and W. Hardy, Phys. Rev. B **73**, 144509 (2006); F. Coneri, S. Sanna, K. Zheng, J. Lord, and R. De Renzi, *ibid.* **81**, 104507 (2010).

[52] M. Vojta, Adv. Phys. **58**, 699 (2009).

[53] C. Berthier, M.-H. Julien, O. Bakharev, M. Horvatić, and P. Ségransan, Physica (Amsterdam) C **282–287**, 227 (1997).

[54] H. Mukuda, S. Shimizu, A. Iyo, and Y. Kitaoka, J. Phy. Soc. Jpn. **81**, 011008 (2012).

[55] H. Alloul, T. Ohno, and P. Mendels, Phys. Rev. Lett. **63**, 1700 (1989).

[56] T. Ito, K. Takenaka, and S. Uchida, Phys. Rev. Lett. **70**, 3995 (1993).

[57] H. Alloul, J. Bobroff, M. Gabay, and P. J. Hirschfeld, Rev. Mod. Phys. **81**, 45 (2009).

[58] G.-q. Zheng, T. Odaguchi, Y. Kitaoka, K. Asayama, Y. Kodama, K. Mizuhashi, and S. Uchida, Physica (Amsterdam) C **263**, 367 (1996).

[59] X. F. Sun, K. Segawa, and Y. Ando, Phys. Rev. Lett. **93**, 107001 (2004).

[60] S. E. Sebastian, N. Harrison, M. M. Altarawneh, C. H. Mielke, R. Liang, D. A. Bonn, W. N. Hardy, and G. G. Lonzarich, Proc. Natl. Acad. Sci. USA **107**, 6175 (2010).

[61] F. Rullier-Albenque, H. Alloul, F. Balakirev, and C. Proust, Europhys. Lett. **81**, 37008 (2008).

[62] S. Li, Z. Yamani, H. J. Kang, K. Segawa, Y. Ando, X. Yao, H. A. Mook, and P. Dai, Phys. Rev. B **77**, 014523 (2008).

[63] V. Balédent, D. Haug, Y. Sidis, V. Hinkov, C. T. Lin, and P. Bourges, Phys. Rev. B **83**, 104504 (2011).

[64] B. Vignolle, B. J. Ramshaw, J. Day, D. LeBoeuf, S. Lepault, R. Liang, W. N. Hardy, D. A. Bonn, L. Taillefer, and C. Proust, Phys. Rev. B **85**, 224524 (2012).

[65] S.-H. Baek, T. Loew, V. Hinkov, C. T. Lin, B. Keimer, B. Büchner, and H.-J. Grafe, Phys. Rev. B **86**, 220504(R) (2012).

[66] E. Blackburn, J. Chang, M. Hücker, A. T. Holmes, N. B. Christensen, R. Liang, D. A. Bonn, W. N. Hardy, U. Rütt, O. Gutowski, M. v. Zimmermann, E. M. Forgan, and S. M. Hayden, Phys. Rev. Lett. **110**, 137004 (2013).

[67] S. Blanco-Canosa, A. Frano, T. Loew, Y. Lu, J. Porras, G. Ghiringhelli, M. Minola, C. Mazzoli, L. Braicovich, E. Schierle, E. Weschke, M. Le Tacon, and B. Keimer, Phys. Rev. Lett. **110**, 187001 (2013).