Superconducting fluctuations and anomalous diamagnetism in underdoped $YBa_2Cu_3O_{6+x}$ from magnetization and ^{63}Cu NMR-NQR relaxation measurements

P. Carretta, A. Lascialfari, A. Rigamonti, A. Rosso, and A. Varlamov*
Department of Physics ''A. Volta,'' Unitá INFM di Pavia-Via Bassi, 6 I-27100 Pavia, Italy
(Received 1 June 1999; revised manuscript received 11 November 1999)

Magnetization and ^{63}Cu nuclear magnetic resonance-nuclear quadrupole resonance relaxation measurements are used to study the superconducting fluctuations in $YBa_2Cu_3O_{6+x}$ (YBCO) oriented powders. In optimally doped YBCO the fluctuating negative magnetization $M_{fl}(H,T)$ is rather well described by an anisotropic Ginzburg-Landau (GL) functional and the curves $M_{fl}/\sqrt{H}$ cross at T_c. In underdoped YBCO, instead, over a wide temperature range an anomalous diamagnetism is observed, stronger than in the optimally doped compound by about an order of magnitude. The field and temperature dependence of M_{fl} cannot be described either by an anisotropic GL functional or on the basis of scaling arguments. The anomalous diamagnetism is more pronounced in samples with a defined order in the Cu(1)O chains. The $^{63}Cu(2)$ relaxation rate shows little, if any, field dependence in the vicinity of the transition temperature $T_c(H=0)$. It is argued how the results in the underdoped compounds can be accounted for by the presence of charge inhomogeneities, favored by chains ordering.

I. INTRODUCTION

The superconducting transition in conventional superconductors is rather well described by mean-field theories, essentially because in the coherence volume ξ^3 a large number of pairs is present. On the contrary, in cuprate superconductors the small coherence length ξ, the reduced carrier density, the marked anisotropy, and the high transition temperature T_c strongly enhance the superconducting fluctuations (SF). In a wide temperature range, which can extend up to 10 or 15 K above T_c, a variety of phenomena related to SF (Refs. 1–3) can be detected by several experiments such as specific heat,[4] thermal expansion,[5] penetration depth,[6] conductivity,[7] and magnetization[8] measurements. Also nuclear magnetic resonance-nuclear quadrupole resonance (NMR)–(NQR) relaxation has been used to detect SF.[9–13] Recently the field dependence of the NMR relaxation rate W and of the Knight shift have been studied, by varying the magnetic field from 2 up to 24 T.[11,12] Mitrovic *et al.*[11] attributed the field dependence of $W(H)$ to the corrections in the density of states (DOS) contribution to the spin-lattice relaxation. On the other hand, Gorny *et al.*[13] have observed $W(H)$ to be field independent, up to 14 T, from 150 K to 90 K, in a sample showing a decrease in $1/T_1T$ starting around 110 K. More recently, also Zheng *et al.*[14] found no effect of the magnetic field on ^{63}CuW in the underdoped $YBa_2Cu_4O_8$, above T_c. An analysis of the role of SF in NMR experiments and of the field dependence of W has been recently carried out by Eschrig *et al.*,[15] by extending the analitical approach of Randeria and Varlamov[16] to include short-wavelength and dynamical fluctuations.

Of particular interest is the role of SF in underdoped high-temperature superconductors. In fact, in these compounds one has the peculiar phenomena of the opening, at $T^* \gg T_c$, of a spin gap (as evidenced by NMR and neutron scattering) and of a pseudogap (as indicated by transport and ARPES experiments).[17–19] These gaps, which are possibly connected,[20] have been variously related to SF of a particular character.[21–23]

In this paper magnetization and $^{63}Cu(2)$ NMR-NQR relaxation measurements in underdoped $YBa_2Cu_3O_{6+x}$ (YBCO) are reported and compared with the ones in the optimally doped compound. The magnetization data yield information on the fluctuating diamagnetism (FD), while the nuclear relaxation rates W convey insights on the effects of SF on the **k**-integrated spin susceptibility in the zero-frequency limit.

The paper is organized as follows. In Sec. II some basic equations describing SF and FD are recalled. After some experimental details (Sec. III), in Sec. IV the experimental findings and their analysis are presented, with emphasis on the effect of the field on W and on the behavior of the diamagnetic magnetization in chain-ordered underdoped YBCO. The main results and conclusions are summarized in Sec. V.

II. THEORETICAL FRAMEWORK AND BASIC EQUATIONS

Because of SF the number of Cooper pairs per unit volume, which is given by the average value of the square of the modulus of the order parameter $\sqrt{\langle|\psi|^2\rangle}$, is different from zero above T_c. In the time-dependent Ginzburg-Landau (GL) description[2,3] the collective amplitudes and the correspondent decay times of SF are given by

$$\langle|\psi_{\mathbf{k}}|^2\rangle = \frac{\langle|\psi_0|^2\rangle}{1+\xi^2k^2} \quad \text{and} \quad \tau_{\mathbf{k}} = \frac{\tau_{GL}}{1+\xi^2k^2}, \tag{1}$$

where $\tau_{GL}=(\pi\hbar/8k_BT_c)\epsilon^{-1}$ and $\xi(T)=\xi(0)\epsilon^{-1/2}$, with $\epsilon=(T-T_c)/T_c$. $\langle|\psi_{\mathbf{k}}|^2\rangle$ plays the role of the Fourier components of the average number of pairs per unit volume, while $\tau_{\mathbf{k}}$ is the correspondent relaxation time.

Fluctuating pairs can give rise to a diamagnetic magnetization above T_c. The diamagnetic magnetization M_{fl} in gen-

eral is not linear in the field H. The curves M_{fl} vs H and M_{fl} vs T can be tentatively analyzed by generalizing the Lawrence-Doniach model, using the free-energy functional[24,25]

$$F[\psi]=\sum_n \int d\mathbf{r}\left[a|\psi_n|^2+\frac{b}{2}|\psi_n|^4 + \frac{\hbar^2}{4m_\parallel}\left|\left[\nabla_\parallel-\frac{2ie}{\hbar c}\mathbf{A}_\parallel\right]\psi_n\right|^2+t|\psi_{n+1}-\psi_n|^2\right], \quad (2)$$

where the last term takes into account the tunneling coupling between adjacent layers. From Eq. (2), deriving the free energy in the presence of the field and by means of a numerical derivation ($M_{fl}=-\partial F/\partial H$), one can obtain the fluctuating magnetization.

According to scaling arguments[26–28] for moderate anisotropy [quasi-three-dimensional (3D) case] one expects that the magnetization curves, at constant field, cross at $T_c(H=0)$ when the magnetization is scaled by $\sqrt{H}$. The amplitude M_{fl} at $T=T_c$ departs from Prange's result[29] by a factor ranging from 3 to 7, corresponding to the anisotropy ratio $(\xi_\parallel/\xi_\perp)$. Accordingly,[30] the data at different fields collapse onto an universal curve when M_{fl}/T is plotted as a function of $\epsilon/H^{1/2\nu}$, with a critical exponent for the coherence length $\nu\simeq 0.66$.

For strong anisotropies, namely, quasi-2D systems, the M_{fl} curves at constant field cross each other at $T_c(0)$. $M_{fl}(T_c)$ is larger than the value obtained from the Gaussian approximation by a factor[28] of approximately 2. Collapse of the data onto a universal curve occurs for ν well above 0.66, usually in the range 1.2–1.4.

The contributions to the relaxation rates W due to SF can be derived within a Fermi liquid scenario, without specifying the nature of the interactions.[2] The direct and most singular contribution, equivalent to the Aslamazov-Larkin paraconductivity, is not effective as a nuclear relaxation mechanism. The positive Maki-Thompson (MT) contribution W_{MT} results from a purely quantum process, involving pairing of the electron with itself at a previous stage of motion, along intersecting trajectories. A negative SF contribution W_{DOS} comes from the density of states reduction when electrons are subtracted to create pairs. Approximate expressions for W_{DOS} and W_{MT} can be derived by resorting to simple physical arguments as follows. The relaxation rates can be approximated by

$$2W\simeq\frac{\gamma^2}{2}\langle A_{\mathbf{k}}\rangle k_B T\sum_{\vec{k}}\left(\frac{\chi''_{spin}(\mathbf{k},\omega)_\perp}{\omega}\right)_{\omega\to 0} \simeq\frac{\gamma^2}{2N}\langle A_{\mathbf{k}}\rangle k_B T\chi_{spin}(0,0)\sum_{\mathbf{k}}\tau_{\mathbf{k}}, \quad (3)$$

where $\langle A_{\mathbf{k}}\rangle$ is an average form factor for Cu nuclei and $\tau_{\mathbf{k}}=J_{\mathbf{k}}(0)$ an effective spectral density.[17,31] SF modify the static spin susceptibility $\chi_{spin}(0,0)$ and the effective correlation time $\Sigma_{\mathbf{k}}\tau_{\mathbf{k}}$ in Eq. (3). From Eq. (1) the in-plane density of pairs is $n_c=\Sigma_{\mathbf{k}}\langle|\psi_{\mathbf{k}}|^2\rangle=2n_e(k_BT_c/E_F)\ln(1/\epsilon)$ and therefore the SF imply

$$\chi_{spin}(0,0)=\frac{n_e\mu_B^2}{E_F}\left[1-\frac{2k_BT_c}{E_F}\ln\frac{1}{\epsilon}\right]. \quad (4)$$

For the dynamical part in Eq. (3), the DOS contribution can be obtained by averaging over the Brillouin zone (BZ) the collective correlation time in Eq. (1):

$$\langle\tau_{DOS}\rangle=\sum_{\mathbf{k}}\tau_{\mathbf{k}}^{DOS}=\hbar\rho(E_F)\frac{\hbar}{4E_F\tau}\ln\frac{1}{\epsilon}, \quad (5)$$

in the dirty limit, differing from the exact calculation[16] only by a small numerical factor. τ is the electron collision time. The MT contribution has to be evaluated in the framework of diagrammatic theories.[2] The final result corresponds to the 2D average over the BZ of a decay rate of diffusive character $\Gamma_{\mathbf{k}}=Dk^2$ (with $D=v_F^2\tau/2$ being the carrier diffusion constant), phenomenologically accounting for phase-breaking processes by adding in the decay rate a frequency τ_ϕ^{-1}:

$$\langle\tau_{MT}\rangle=\sum_{\mathbf{k}}\{(Dk^2+\tau_\phi^{-1})\epsilon(1+\xi^2k^2)\}^{-1} =\hbar\rho(E_F)\frac{\hbar}{4E_F\tau}\frac{1}{(\epsilon-\gamma_\phi)}\ln\frac{\epsilon}{\gamma_\phi}, \quad (6)$$

where $\gamma_\phi=\xi^2(0)/\tau_\phi D=\pi\hbar/8K_BT_c\tau_\phi$ is a dimensionless pair-breaking parameter.

By indicating with W^0 the relaxation rate in the absence of SF and by neglecting the correction to $\chi_{spin}(0,0)$ [Eq. (4)], from Eqs. (3), (5), and (6) the relaxation rate in 2D systems becomes

$$W^{SF}=W^0\left[\frac{\pi\hbar}{8F_F\tau}\frac{1}{(\epsilon-\gamma_\phi)}\ln\frac{\epsilon}{\gamma_\phi}-\frac{0.8\hbar}{E_F\tau}\ln\frac{1}{\epsilon}\right]. \quad (7)$$

To extend this equation to a layered system, when dimensionality crossover (2D→3D) occurs, one has to substitute[2] $\ln(1/\epsilon)$ by $2\ln[2/(\sqrt{\epsilon}+\sqrt{\epsilon+r})]$ and $\ln(\epsilon/\gamma_\phi)$ by $2\ln[(\sqrt{\epsilon}+\sqrt{\epsilon+r})/(\sqrt{\gamma_\phi}+\sqrt{\gamma_\phi+r})]$, with $r=2\xi_c^2(0)/d^2$ anisotropy parameter (d is the interlayer distance). It is noted that the MT contribution [first term in Eq. (7)] is present only for s-wave orbital pairing, while it is averaged to almost zero for d symmetry.

III. EXPERIMENTAL DETAILS

The measurements have been carried out on oriented powders of optimally doped YBCO, on one chain-disordered underdoped, and on two chain-ordered underdoped YBCO samples.[32] By thermogravimetric analysis the oxygen content in the underdoped compounds was found close to 6.66, with slight differences in T_c. In the chain-ordered compounds electron diffraction microscopy evidenced the expected tripling of the a axis, while resistivity measurements show a sharp transition with zero resistivity at $T=62$ K and occurrence of paraconductivity below about 75 K.[32] Table I shows the main properties of the samples, as obtained from a combination of measurements.

The ^{63}Cu relaxation rates $2W\equiv T_1^{-1}$ have been measured by standard pulse techniques. In NQR the recovery towards the equilibrium after the saturation of the $(\pm\frac{1}{2}\to\pm\frac{3}{2})$ line is well described by a single exponential, directly yielding $6W$.

TABLE I. Main properties of the samples, as obtained from a combination of measurements.

	YBCO optimally doped	YBCO underdoped		
		Disordered	Chain-ordered	
$T_c(0)$	91.1 ± 0.4 K	63.3 ± 0.4 K	64.5 ± 0.4 K	64.2 ± 0.4 K
$T_c(H=6\text{ T})$	87 ± 0.5 K	60 ± 1 K		
$T_c(H=9.4\text{ T})$		55 ± 1	54 ± 1 K	
$\left(\frac{\partial H_{c2}}{\partial T}\right)_{T_{c0}}$ T/K	-1.2 ± 0.2	-1.05	-1.1 ± 0.2	-1.0 ± 0.1
$H_{c2}(T\to0)$ $\mathbf{H}\parallel\mathbf{c}$	100 ± 20 T	~70 T	~65 T	
$H_{c2}(T\to0)$ $\mathbf{H}\perp\mathbf{c}$	~67 T		~44 T	

For the NMR relaxation a good alignment of the $\vec{c}$ axis of the grains along the magnetic field is crucial to extract reliable values of $W(H)$. The NMR satellite line, corresponding to the $(\frac{1}{2}\to\frac{3}{2})$ transition, can be used to adjust the alignment and to monitor the spread in the orientation of the c axis, the resonance frequency being shifted at the first order by the term $eQV_{zz}(3\cos^2\theta-1)/h$, due to the quadrupole interaction (θ angle of the $\vec{c}$ axis with the field). From the width of the spectrum [Fig. 1(a)] the spread in the orientation of the c axis appears within about two degrees.

The recovery law for the NMR satellite transition is

$$y(t)=0.5\exp(-12Wt)+0.4\exp(-6Wt)+0.1\exp(-2Wt). \quad (8)$$

This formula fits the experimental data [Fig. 1(b)] and indicates that the estimate of $2W$ is not affected by other resonance lines.

The magnetization has been measured by means of a Metronique Ingegnerie superconducting quantum interface device (SQUID) magnetometer, on decreasing the temperature at constant field and, at selected temperatures, by varying H. The paramagnetic contribution to M was obtained from M vs H at $T\geqslant110$ K, where practically no fluctuating magnetization is present. Then M_{fl} was derived, accounting for an intrinsic Pauli-like paramagnetic term as well as for a small contribution from paramagnetic impurities. The paramagnetic susceptibility turns out to be a little temperature dependent around T_c and this dependence will be neglected in discussing the much stronger diamagnetic term.

In optimally doped YBCO the measurements have been carried out to confirm the results recently obtained in single crystals by other authors.[24,28,30,33] The isothermal magnetization curves are satisfactorily described on the basis of the anisotropic GL functional [Eq. (2)], for $\epsilon\geqslant4\times10^{-2}$. Only close to T_c and for $H\leqslant1$ T an observable departure is detected, indicating crossover to a region of non-Gaussian SF, in agreement with recent thermal expansion measurements.[5] The M_{fl} vs T curves cross at $T_c(0)$ when M_{fl} is scaled by $\sqrt{H}$, as expected for moderate anisotropy.[28]

IV. RESULTS AND ANALYSIS

Let us first comment on the data in optimally doped YBCO [Fig. 2(a)]. The comparison between the NMR and NQR W's has been already discussed in previous papers.[9,10] Here we only add a few comments motivated by more recents works[11,13,34] involving the remarkable aspect of the field dependence.

The NQR W is compared to Eq. (7), using the values[2]

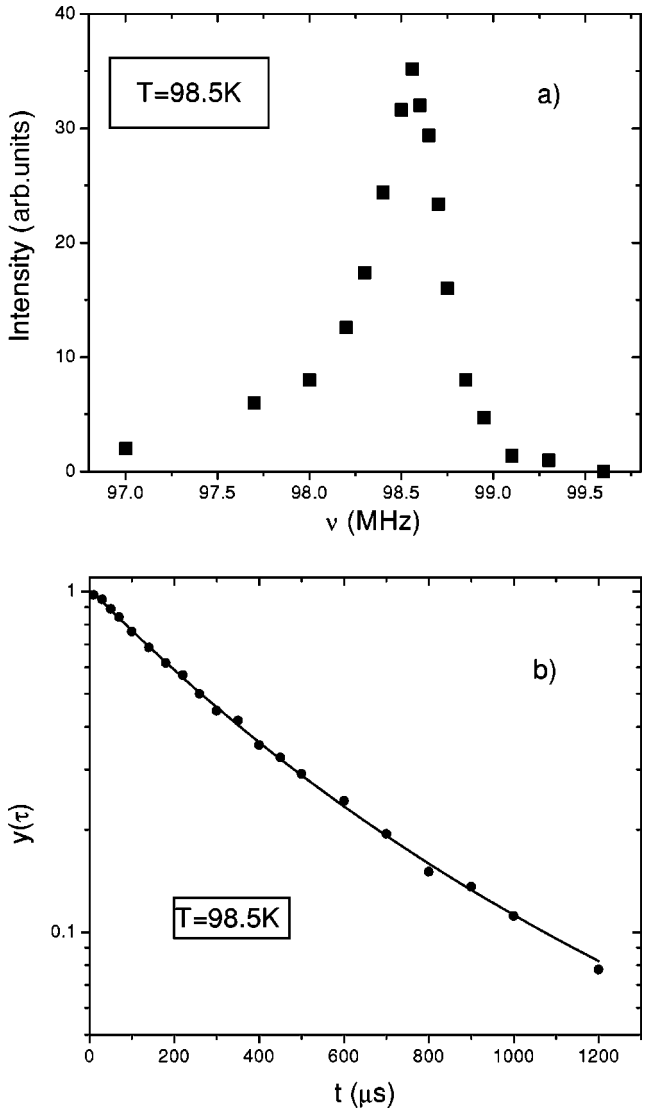

FIG. 1. (a) NMR spectrum of the ^{63}Cu (2) satellite line and (b) the correspondent relaxation law in optimally doped YBCO [the solid line is the best fit according to Eq. (8) in the text].

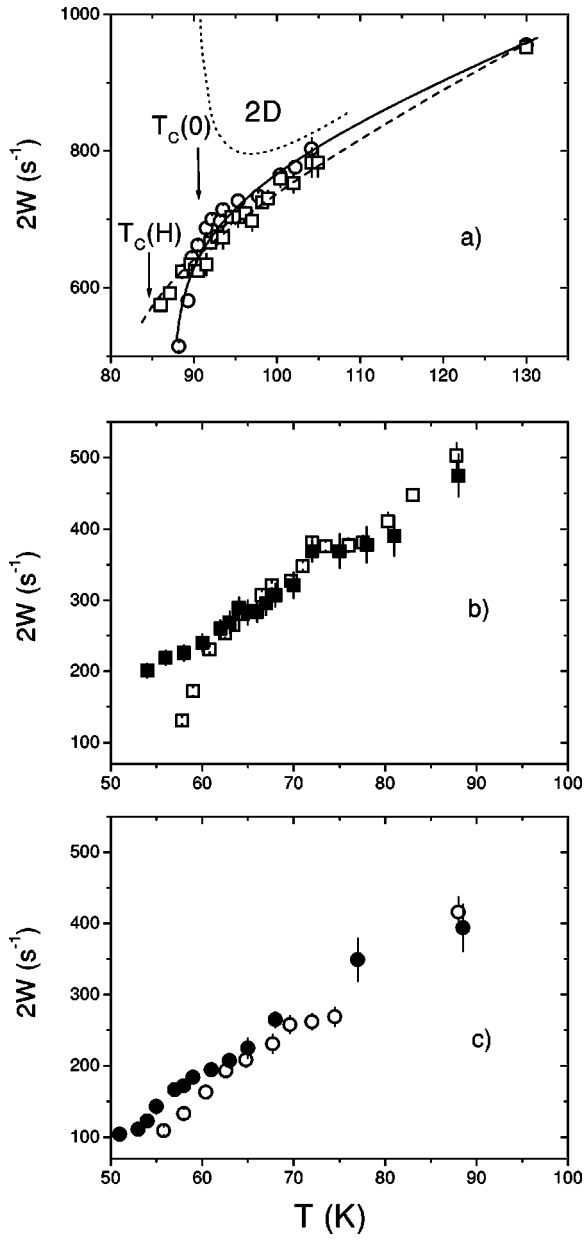

FIG. 2. (a) NQR (○) and NMR (□) ($H_0=6\,\mathrm{T}\|\vec{c}$) ^{63}Cu relaxation rates $2W$ in optimally doped YBCO. The solid and dashed lines are guides for the eye. The dotted line is the temperature behavior according to Eq. (7) in the text for $r=0$ (namely, a 2D spectrum of fluctuations) and $\gamma_\phi=0.2$. NQR (empty symbols) and NMR (full symbols) relaxation rates $2W$ (in a field of 9.4 T along the c axis) (b) in chain-disordered and (c) in chain-ordered underdoped YBCO around T_c.

$\tau=10^{-14}$ s, $\tau_\phi=2.10^{-13}$ s, and a dephasing time parameter $\gamma_\phi=0.2$. A quantitative fitting is inhibited by the fact that the background contribution to W does not follow the Korringa law, in view of correlation effects among carriers.[17] A firm deduction, however, is that an anisotropy parameter $r\neq 0$ (i.e., a crossover to a 3D regime) is required to avoid unrealistic values [2D line in Fig. 2(a)] as indicated also by magnetization measurements (see later on).

Mitrovic *et al.*[11] have discussed the field dependence of T_1 in terms of quenching of the DOS term only, by resorting to the theory of Eschrig *et al.*[15] The field dependence of the SF contribution to the relaxation rate is a delicate issue, because of the nontrivial interplay of many parameters that include reduced temperature ϵ, reduced field $\beta=2H/H_{c2}$, anisotropy parameter r, and elastic and phase-breaking times τ and τ_ϕ. Eschrig *et al.*[15] have extended the analytical approach[16] by taking into account arbitrary values of $(k_BT\tau/\hbar)$, short-wave fluctuations, and dynamical fluctuations. The price of this generalization is the restriction to a 2D spectrum of SF fluctuations. In view of our experimental findings the analysis of the field effect in a purely 2D framework is questionable. On the other hand, a reexamination of the field effect for a layered system is now possible by applying the method of the DOS term regularization devised by Buzdin and Varlamov,[34] which has indicated how the divergences can be treated. Since the dynamical fluctuations could be relevant only for fields of the order of H_{c2}, up to 20–30 T the fluctuations can be safely treated in the nearly static limit. The field dependence for weak fields ($\beta\ll\epsilon$) turns out to be[35]

$$W(\beta\ll\epsilon)-W(0,\epsilon)=W^{(0)}\frac{\hbar}{24E_F\tau}\left(\kappa(T\tau)-\frac{\pi}{8\gamma_\phi}\right)\frac{\epsilon+r/2}{[\epsilon(\epsilon+r)]^{3/2}}\beta^2, \qquad (9)$$

where

$$\kappa(T\tau)=\frac{7\zeta(3)}{\pi}\frac{1}{4\pi T\tau[\psi(1/2)-\psi(1/2+1/4\pi T\tau)]+\psi'(1/2)}=\begin{cases}14\zeta(3)/\pi^3, & T\tau\ll 1\\ 4T\tau, & 1\ll T\tau\ll 1/\sqrt{\epsilon}\end{cases}$$

(here $\hbar=k_B=1$).

Therefore, W can increase or decrease with increasing magnetic field, depending on the mean-free path in the specific sample. If $T\tau\lesssim 0.1$ the main correction to W is due to the MT term and one should observe W decreasing with increasing field, while if $T\tau\gtrsim 0.1$ the DOS correction becomes dominant and W is expected to increase.

As it appears from Fig. 2 the effect of the field for $T\geqslant T_c(H=0)$ is small, if any. It is noted that if Eq. (7) is applied to the underdoped compounds the reduction in the Fermi energy E_F and the increase in the anisotropy (i.e., decrease of r) would imply a sizable increase in W^{SF}. This effect is absent in the underdoped compounds [see Figs. 2(b) and 2(c) for the chain-disordered and chain-ordered YBCO's, respectively]. The decrease of W over a wide temperature range ($T\lesssim 160$ K), usually related to the spin-gap opening,[17] in principle could be attributed to a field-independent DOS term. On the other hand, the conventional SF of GL character should occur only close to T_c, where, on the contrary, $\chi_{spin}(0,0)$ is weakly temperature dependent. Equation (9), in principle, does predict a field dependence. However, if typical values $E_F\simeq 3000$ K, $\tau\simeq 10^{-14}$, $\kappa(T\tau)-(\pi/8\gamma_\phi)\simeq 1$, and $r\simeq 0.1$ are used, for $\epsilon\simeq 3\times 10^{-2}$ one has $W(\beta)-W(0)\simeq 0.15W^{(0)}\beta^2$, which is difficult to observe for $\beta\ll 1$. A comprehensive discussion of the field dependence of the SF contribution to the relaxation rate is given

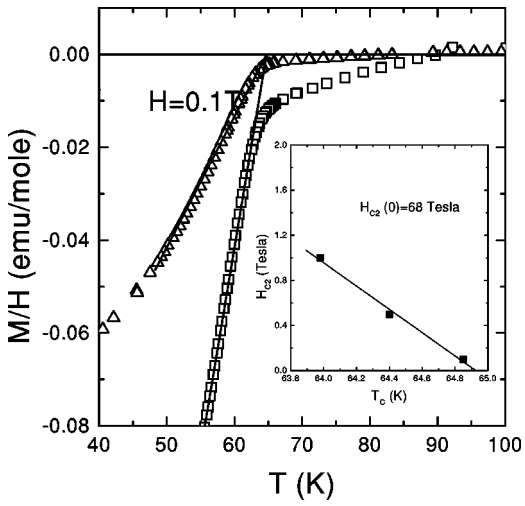

FIG. 3. Raw data for M/H ($H=0.1$ T) in chain-ordered YBCO, for field along the c axis (□) and in the a-b plane (△). In the inset the results for H_{c2} around T_c are shown (estimated experimental error $\leqslant 2\%$). The solid lines indicate the procedure to evaluate $T_c(H)$ and $T_c(0)$.

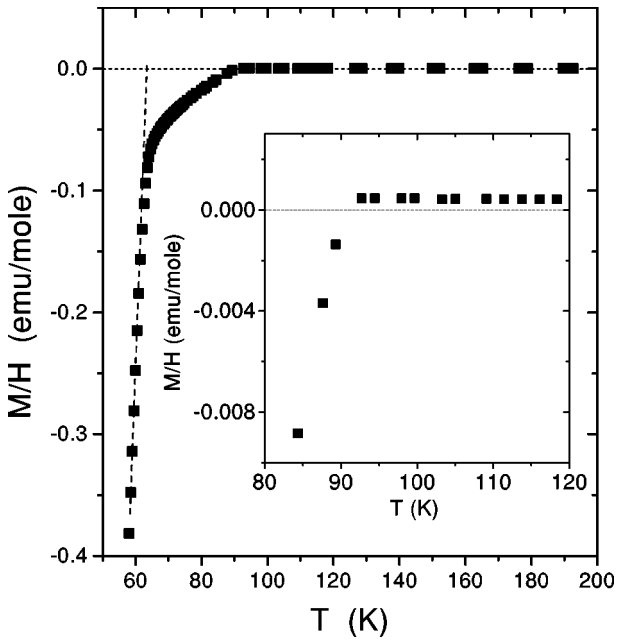

FIG. 4. Ratio M/H, in a field of 0.02 T along the c axis, in chain-ordered YBCO. The inset is the blowup of the data for $T \geqslant 85$ K. Similar results have been obtained in another sample prepared with the same procedure.

elsewhere.[35] Here we only remark that the results in strong fields ($\beta \gtrsim 0.2$) from Mitrovic *et al.*[11] cannot be justified on the basis of Eq. (9).

As regards the magnetization measurements in optimally doped YBCO, as already mentioned, our data indicate a 3D regime crossing from Gaussian to critical fluctuations close to T_c. The value of $\gamma_{an} m_3(\infty)$ (Ref. 28) at the crossing point of the curves $M_{fl}/\sqrt{H}$ vs T turns out around -1.5, corresponding to an anisotropy factor $\gamma_{an}=4.5$. A collapse onto a common function is obtained when $M_{fl}/\sqrt{HT}$ is plotted as a function of $\epsilon/H^{0.747}$. The collapse fails in magnetic fields less than about 0.3 T. A small-field departure from the universal function has been already observed in underdoped compounds,[30] but no mention about it is found in the literature for optimally doped YBCO.

In the underdoped compounds a marked enhancement of the fluctuating magnetization is observed (Figs. 3 and 4). In Fig. 4 the susceptibility, defined as M/H for $H=0.02$ T, is plotted. The inset is a blowup of the results around the temperature where reversing of the sign of the magnetization occurs. The susceptibilities for chain-ordered and chain-disordered underdoped samples are compared with the one in optimally doped YBCO in Fig. 5.

In underdoped YBCO the magnetization curves dramatically depart from the ones expected on the basis of Eq. (2) and numerical derivative. The curves M_{fl} vs T do not cross at any temperature. No collapse on a common function is found for $M_{fl}/\sqrt{H}$ or for M_{fl} as a function of $\epsilon/H^{1/2\nu}$ (Fig. 6). These anomalies are more manifested in the chain-ordered compound, as it can be realized from the peculiar shape of the isothermal magnetization vs H (Fig. 7). One could remark that a fraction of a few percent of nonpercolating local superconducting regions can account for the absolute value of the diamagnetic susceptibility and for the shape of the magnetization curves. A trivial chemical inhomogeneity could be suspected. The fact that the anomalous diamagnetism has been observed in three samples differently prepared and since it is strongly enhanced in the chain-ordered ones, supports the intrinsic origin of the phenomenon. Recent theories[36,37] have discussed how a distribution of local superconducting temperatures, related to charge inhomogeneities, can cause anomalous diamagnetic effects.

Mesoscopic charge inhomogeneities have been predicted on the basis of various theoretical approaches.[22,39] In underdoped compounds one could expect the occurrence of preformed Cooper pairs causing ''local'' superconductivity without long-range phase coherence. An inhomogeneous distribution of carriers, on a mesoscopic scale, is supported by *ab* plane optical conductivity[38] and it is the basic ingredient of the stripes model.[40] Since the anomalous diamagnetism is

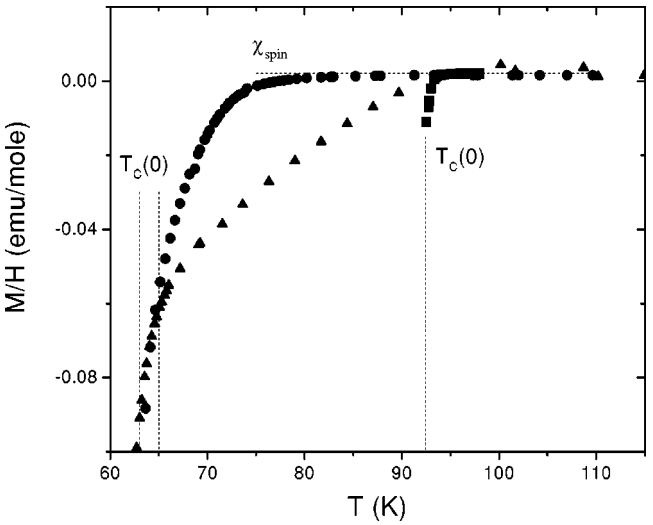

FIG. 5. Comparison of the susceptibility (defined as M/H, for $H=0.02$ T along the c axis) in optimally doped YBCO (squares) and in chain-disordered (circles) and chain-ordered (triangles) underdoped YBCO.

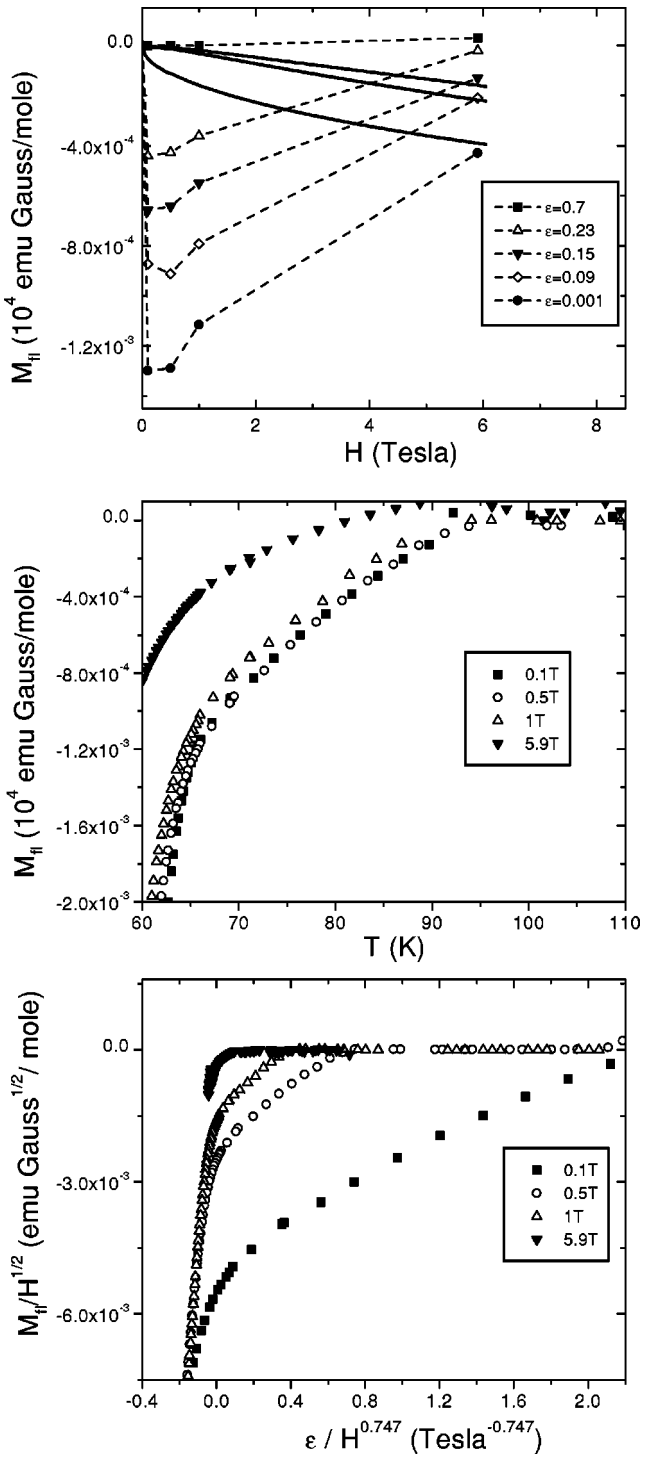

FIG. 6. Evidence of the failure of anisotropic GL functional and of scaling arguments to describe the magnetization curves in chain-ordered underdoped YBCO. The solid lines in the upper part of the figure are the behaviors obtained from the numerical derivative of Eq. (2). The dashed curves are guides for the eye.

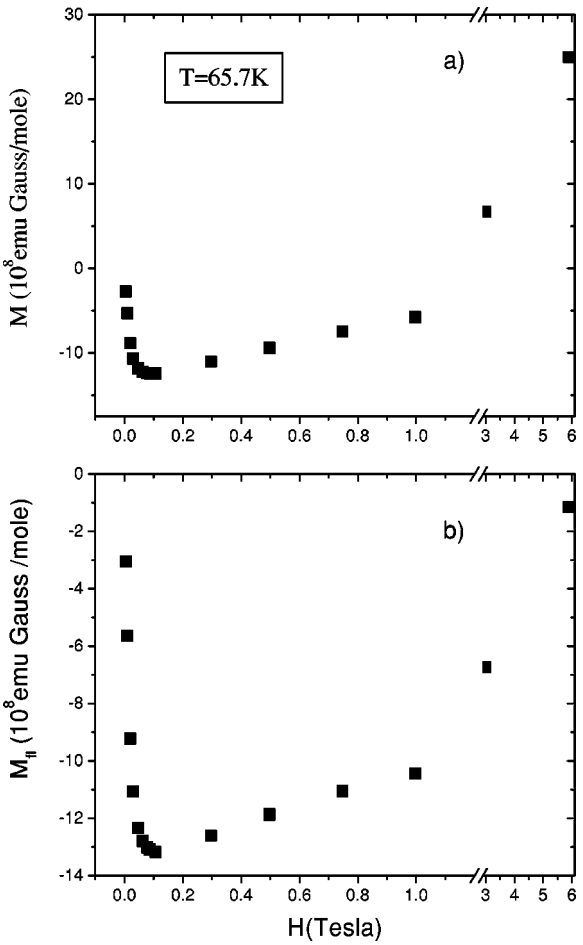

FIG. 7. Magnetization curve in chain-ordered underdoped YBCO under high stabilization temperature ($T=65.67$ K). (a) Raw data for the total magnetization, (b) fluctuating magnetization M_{fl} as a function of the field.

enhanced in chain-ordered underdoped YBCO one is tempted to relate it to the presence of stripes. The insurgence of phase coherence among adjacent charge-rich regions can qualitatively be expected to yield strong screening above the bulk superconducting temperature.[36,37,41] Therefore, one could phenomenologically describe the role of SF in underdoped compounds, as follows. At $T \gg T_c$, just below T^*, fluctuating pairs are formed, without long-range phase coherence. Below T_{SF}, about 20–30 K above T_c, phase coherence among adjacent charge-rich drops[37] start to develop, yielding the formation of superconducting loops with strong screening and causing the onset of the anomalous diamagnetism. Close to T_c, SF of GL character occurs and long-range phase coherence sets in.

V. SUMMARIZING REMARKS AND CONCLUSIONS

By combining magnetization and NMR-NQR relaxation measurements, an attempt has been made to clarify the role of superconducting fluctuations and of fluctuating diamagnetism in underdoped YBCO, *vis-à-vis* the optimally doped compound. In the latter case the fluctuations above T_c are rather well described by an anisotropic Ginzburg-Landau (GL) functional and by scaling arguments for slightly anisotropic systems. A breakdown of the Gaussian approximation for small magnetic fields has been observed close to T_c. The ^{63}Cu relaxation rates W around T_c do not show a noticeable field dependence. One cannot rule out the presence of a MT contribution, and then of a small *s*-wave component in the spectrum of the fluctuations, which should be sample dependent in view of the role of impurities in the pair-breaking mechanisms. In underdoped YBCO an anomalous diamagne-

tism is observed, on a large temperature range. The diamagnetic susceptibility at T_c is about an order of magnitude larger than the one in the optimally doped sample and the isothermal magnetization curves cannot be described by the anisotropic GL functional. The data cannot be fit by a scaling function of the form $\epsilon/H^{1/2\nu}$. The anomalies are more marked in the chain-ordered samples. ^{63}Cu NQR W in the underdoped YBCO almost coincide with the NMR ones at $H=9.4\,\mathrm{T}$, in agreement with a theoretical estimate of the effect of the magnetic field. Another conclusion drawn from the field and temperature dependences of the ^{63}Cu relaxation rate is that the spin gap does not depend on the magnetic field and that the behavior of the spin susceptibility for $T \gg T_c$ cannot be ascribed to SF of GL character.

The fluctuating diamagnetism observed in underdoped YBCO can be phenomenologically accounted for by the presence of charge inhomogeneities at mesoscopic level. The anomalies in the magnetization curves can be attributed to ''locally superconducting'' nonpercolating drops, present above the bulk T_c and favored by chain ordering.

ACKNOWLEDGMENTS

A. Bianconi, F. Borsa, J. R. Cooper, and M. H. Julien are thanked for useful discussions. The collaboration of Paola Mosconi in processing the SQUID magnetization data is gratefully acknowledged. Thanks are due to B. J. Suh and P. Manca, for having provided well-characterized underdoped YBCO compounds. The availability of the Metronique Ingegnerie SQUID magnetometer from the Department of Chemistry, University of Florence (Professor D. Gatteschi) is gratefully acknowledged. The research has been carried out in the framework of a ''Progetto di ricerca avanzata (PRA),'' sponsored by Istituto Nazionale di Fisica della Materia (PRA-SPIS 1998-2000).

*On leave of absence from Institute of Steel and Alloys, University of Moscow, Russia. Present address: Unitá INFM-Tor Vergata, 00133 Roma, Italy.

[1] For a recent collection of papers, see *Fluctuations Phenomena in High Temperature Superconductors*, edited by M. Ausloos and A. A. Varlamov (Kluwer Academic, Netherlands, 1997).

[2] A. A. Varlamov, G. Balestrino, E. Milani, and D. V. Livanov, Adv. Phys. **48**, 655 (1999).

[3] M. Thinkham, *Introduction to Superconductivity* (Mc Graw-Hill, New York, 1996), Chaps. 8 and 9.

[4] See A. Junod, in *Studies of High Temperature Superconductors*, edited by A. V. Narlikar (Nova Science, New York, 1996), Vol. 18.

[5] V. Pasler *et al.*, Phys. Rev. Lett. **81**, 1094 (1998).

[6] Z. H. Lin *et al.*, Europhys. Lett. **32**, 573 (1995), and references therein.

[7] M. A. Howson *et al.*, Phys. Rev. B **51**, 11 984 (1995), and references therein.

[8] See the review paper by L. N. Bulaevskii, Int. J. Mod. Phys. B **4**, 1849 (1990), and several papers in *Fluctuations Phenomena in High Temperature Superconductors* (Ref. 1).

[9] P. Carretta, D. Livanov, A. Rigamonti, and A. A. Varlamov, Phys. Rev. B **54**, R9682 (1996).

[10] P. Carretta, A. Rigamonti, A. A. Varlamov, and D. Livanov, Nuovo Cimento D **19**, 1131 (1997).

[11] V. Mitrovic *et al.*, Phys. Rev. Lett. **82**, 2784 (1999).

[12] H. N. Bachman *et al.*, Phys. Rev. B **60**, 7591 (1999).

[13] K. Gorny *et al.*, Phys. Rev. Lett. **82**, 177 (1999).

[14] G. Zheng *et al.*, Phys. Rev. B **60**, R9947 (1999).

[15] M. Eschrig *et al.*, Phys. Rev. B **59**, 12 095 (1999).

[16] M. Randeira and A. A. Varlamov, Phys. Rev. B **50**, 10 401 (1994).

[17] A. Rigamonti, F. Borsa, and P. Carretta, Rep. Prog. Phys. **61**, 1367 (1998).

[18] A. G. Loeser *et al.*, Science **273**, 325 (1996).

[19] H. Ding *et al.*, Nature (London) **382**, 51 (1996).

[20] G. V. M. Williams, J. L. Tallon, R. Michalak, and R. Dupree, Phys. Rev. B **54**, R6909 (1996).

[21] T. Dahm, D. Manske, and L. Tewordt, Phys. Rev. B **55**, 15 274 (1997).

[22] V. J. Emery, S. A. Kivelson, and O. Zachar, Phys. Rev. B **56**, 6120 (1997); see also V. J. Emery and S. A. Kivelson, J. Phys. Chem. Solids **59**, 1705 (1998).

[23] C. Castellani, C. Di Castro, and M. Grilli, Z. Phys. B: Condens. Matter **103**, 137 (1997).

[24] C. Baraduc, A. Budzin, J.-Y. Henry, J. P. Brison, and L. Puech, Physica C **248**, 138 (1995).

[25] A. Budzin and V. Dorin, in *Fluctuations phenomena in High Temperature Superconductors* (Ref. 1), p. 335.

[26] M. B. Salamon *et al.*, Phys. Rev. B **47**, 5520 (1993).

[27] T. Schneider and H. Keller, Int. J. Mod. Phys. B **8**, 487 (1993).

[28] A. Junod, J. Y. Genoud, G. Triscone, and T. Schneider, Physica C **294**, 115 (1998).

[29] R. E. Prange, Phys. Rev. B **1**, 2349 (1970).

[30] M. A. Hubbard, M. B. Salamon, and B. W. Veal, Phys. Rev. B **259**, 309 (1996).

[31] C. P. Slichter, *Principles of Magnetic Resonance* (Springer-Verlag, Berlin, 1990).

[32] P. Manca, P. Sirigu, G. Castellani, and A. Migliori, Nuovo Cimento D **19**, 1009 (1997); G. Castellani, A. Migliori, P. Manca, and P. Sirigu, *ibid.* **19**, 1075 (1997).

[33] U. Welp *et al.*, Phys. Rev. Lett. **67**, 3180 (1991).

[34] A. I. Buzdin and A. A. Varlamov, Phys. Rev. B **58**, 14 195 (1998).

[35] P. Mosconi, A. Rigamonti, and A. A. Varlamov, Appl. Magn. Reson. (to be published).

[36] Yu. N. Ovchinikov, S. A. Wolf, and V. Z. Kresin, Phys. Rev. B **60**, 4329 (1999).

[37] E. Z. Kuchinskii and M. V. Sadovskii, cond-mat/9910261 (unpublished).

[38] D. Mihailovic, T. Mertelj, and K. A. Muller, Phys. Rev. B **57**, 6116 (1998); see also V. V. Kabanov *et al.*, *ibid.* **59**, 1497 (1999).

[39] M. R. Norman, cond-mat/9904048 (unpublished).

[40] A. Bianconi *et al.*, Phys. Rev. Lett. **76**, 3412 (1996).

[41] C. Bergemann *et al.*, Phys. Rev. B **57**, 14 387 (1998).