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Glassiness and superconductivity in strongly Pr-doped $YBa_2Cu_3O_{7-\delta}$ single crystals

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We present magnetization data on superconducting strongly Pr-doped $YBa_2Cu_3O_{7-\delta}$ showing the existence of irreversibility at temperatures much higher than superconducting critical temperature and Neél temperatures for both planar Cu and Pr systems. A model in terms of cluster glass is proposed.

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Keywords: Superconductivity, Glassiness, Pr-doped $YBa_2Cu_3O_{7-\delta}$

1. Introduction

Doping Mott insulators with charge causes dramatic changes in their physics. Spectacular results of the doping are superconductivity in cuprates and colossal magnetoresistivity in manganites. In all cases, doping is accompanied by mesoscopic phase separation that signalises that the magnetic short-range forces start to be competed by the long-range Coulomb force. The long range order is, therefore, frustrated by the latter and, due to the spin-charge coupling, the system turn into a phase separated one [1] and displays a specific glassiness which is self generated [2, 3].

Glassy state is present in most of phase diagrams of cuprates, but its occurrence and the relation with the neighbouring phases is not universal [4]. In La-based cuprates, the glassy state emerges directly from the antiferromagnetic (AFM) state at a doping level lower than the critical one for the occurrence of superconductivity (SC) but survives at higher hole concentration [5-7]. The glassiness is still under debate in the case of the bilayered $YBa_2Cu_3O_{6+x}$ [8-13].

A singular case is $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ where the doping level is controlled by both oxygen and praseodymium. The increase of oxygen content increases the hole concentration, whereas Pr generate a depletion of the Zhang-Rice conduction band [14]. For fully oxygenated system, $\delta \cong 0$, the glassy state coexists with superconductivity for $x \geq 0.4$ but was not detected above $x_c \leq 0.55$, where SC state vanishes. Therefore there is no intermediate phase between AFM and SC states [15-17]. Actually, there are two subsystems susceptible to show magnetic interaction, specifically the Cu and Pr subsystems, and they equally display both AFM and glassy ordering at high Pr content. An attempt of phase diagram in a narrow concentration range around x_c with a remarkable richness in phases is shown in Figure 1 [17, 18]. This equilibrium picture goes along with strong phase fluctuations of low symmetry phases due to the decreased dimensionality and charge density. The fluctuations are expected to alter the magnetic response of the paramagnetic high temperature phase.

This communication present the results of the investigations of the magnetic response of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ in this rich-in-phase concentration range aiming to depict the role of the fluctuation in each phase. The main finding was that the paramagnetic phase is not completely reversible but is dressed with an important irreversibility extending up to 200 K.

2. Experimental

We used a $Y_{0.47}Pr_{0.53}Ba_2Cu_3O_{7-\delta}$ single crystal of size $0.77 \times 0.57 \times 0.067$ mm^3, for a set of magnetic investigations consisting in magnetization M vs magnetic field H, and *dc*- and *ac*-magnetization vs temperature T. The measurements were performed with a superconducting quantum interference device (SQUID) magnetometer. The single crystal was mounted on a Teflon support which was submitted to the same investigations as the sample in order to subtract the background. Each run was performed after the crystal was warmed up to temperatures higher than 200 K and cooled in zero-magnetic field (ZFC) down to the temperature of investigation. Field cooling (FC) protocol was used in certain cases. The critical temperature of the single crystal was T_c = 13 K and was obtained both from *ac*-susceptibility and transport measurements. Figure 2 shows the temperature dependence of the derivative of the real part of the *ac*-susceptibility.

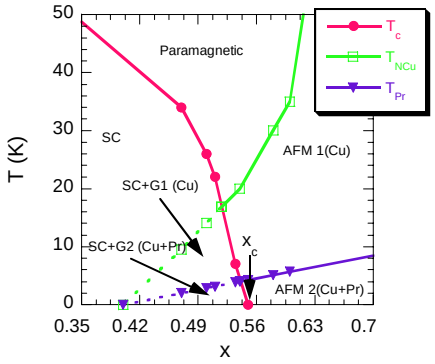

Fig. 1. Phase diagram of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ around the critical concentration xc. SC, G, and AFM stand for the superconducting state, glassy state, and antiferromagnetic state, respectively.

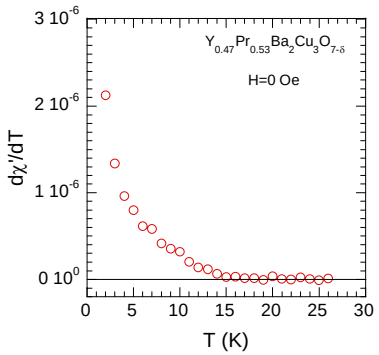

Fig. 2. *Temperature T dependence of the first derivative of the real part of the ac- susceptibility* $\frac{d\chi'}{dT}$.

3. Results and discussion

The magnetization M vs applied magnetic field H displays irreversibility up to 200 K as can be seen in Fig. 3. At low temperature, below T_c, it consists of a superconducting contribution superposed on a paramagnetic term (see the Inset to Fig. 3 for T = 5 K). The paramagnetic term is dominant over the diamagnetic screening at relatively low fields even at the lowest temperature. The presence of irreversibility is not expected at temperatures higher than 18 K where the equilibrium phase diagram predicts a paramagnetic state for both Cu and Pr moments.

The temperature dependence of the *dc*-susceptibility χ is shown in Fig. 4 for an applied field of 100 Oe. A general feature of χ is the strong field dependence which is not noticed in low x $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$. Irreversibility is present only for $H \leq 400$ Oe, hence, there is always a zero-field-cooled susceptibility χ_{ZFC} which is smaller than the field-cooled one χ_{FC} in this field range.

The zero-field-cooled susceptibility χ_{ZFC} is typical superconducting,hence, negative, at $T < T_c(H)$. At higher temperatures, χ_{ZFC} becomes positive, reaches a wide maximum at T_g, and at even higher temperatures decreases following a Curie-Weiss (CW) law with antiferromagnetic character, i.e., has negative Curie-Weiss temperature θ_{CW}. In the following we will use the absolute value of θ_{CW}. The field-cooled susceptibility χ_{FC} obeys also an AFM CW dependence at high temperatures but with parameters slightly different from χ_{ZFC}. Below T_g, $\chi_{FC}(T)$ roughly follows a $T^{-\alpha}$ dependence. The exponent α is a monotonous function of field reaching a maximum of $\alpha \approx 0.5$ at 400 Oe. Above 300 Oe, the transition between the two dependencies smeared out but is still visible in the derivative.

As we have mentioned, both high temperature susceptibilities $\chi_{ZFC}(T)$ and $\chi_{FC}(T)$ obey a CW dependence, $\chi = \chi_0 + C_{CW}/(T+\theta_{CW})$, with slightly different parameters. Here, χ_0 includes the Pauli and Van Vleck paramagnetism, and the core diamagnetism; $C_{CW} = p_{eff}\mu_B^2 n/3k_B$ is the Curie-Weiss constant with p_{eff} the effective number of Bohr magnetons μ_B, n the concentration of paramagnetic ions, and k_B thsolute value of θ_{CW} e Boltzmann constant.

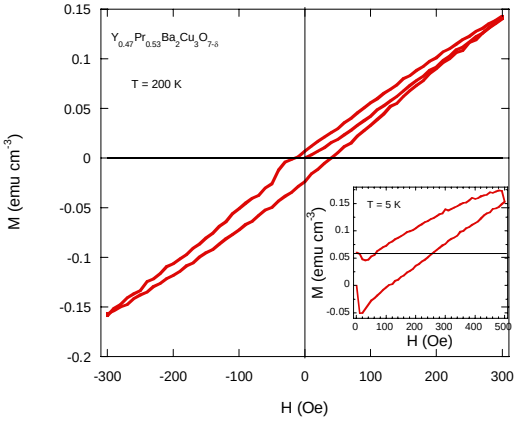

Fig. 3 Field H dependence of the volume magnetization M of a $Y_{0.47}Pr_{0.53}Ba_2Cu_3O_{7-\delta}$ single crystal at 200 K. Inset: Magnetization loop at 5 K.

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Normally, a CW dependence is the result of the free motion of the magnetic ions. Here, we have an unusual field and history dependent paramagnetism which suggests, however, the existence of a certain internal structure of these moments. A fit of both susceptibilities with a CW law provides field dependent parameters χ_0, θ, and C. All these parameters decrease with increasing H, another behavior which rules out the ordinary paramagnetism (Fig. 5). The weak irreversibility have suggested the separation of the total susceptibility into a

reversible and irreversible contribution $\chi = \chi_{rev} + \chi_{irr}$, where $\chi_{rev} = (\chi_{ZFC} + \chi_{FC})/2$ and $\chi_{irr} = (\chi_{ZFC} - \chi_{FC})/2$. The CW dependence is also valid in the high field region as is conspicuous in the $\chi(T)$ vs $(T + \theta_{CW})^{-1}$ plot (see Inset to Fig. 4 for H = 800 Oe). The CW parameters C_{CW} (Fig. 5) and θ_{CW} (Fig. 6) extracted from the reversible curve depend on field as a power law at low fields, $H < 500$ Oe, $C \sim H^{\gamma}$ and $\theta \sim H^{\beta}$ with $\gamma \approx 0.58$ and $\beta \approx 2/3$, respectively, whereas for the parameter extracted from ZFC and FC susceptibilities is hardly to assign an analytical dependence (Fig. 5 and 6). It is noteworthy that the ZFC parameters are almost always larger than FC parameters.

The real part of the *ac*-susceptibility data show the typical diamagnetic screening at low temperature (Fig. 1) and a residual diamagnetic response above T_c whereas the imaginary part χ'' is non zero in the whole investigated temperature range but changes the slope at T_c (Fig. 7). Diamagnetic-like response of *ac*-susceptibility was reported also in almost optimally doped $La_{2-x}Sr_xCuO_4$ above T_c [19] and was attributed to the superconducting phase fluctuations of the order parameter.

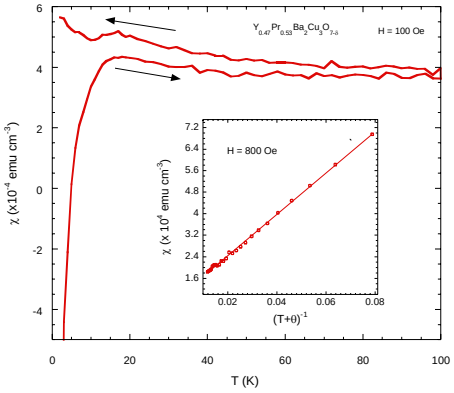

Fig. 4 Temperature T dependence of the dc-susceptibility χ a $Y_{0.47}Pr_{0.53}Ba_2Cu_3O_{7-\delta}$ single crystal for an applied field of 100 Oe. Inset: DC-susceptibility of the same sample at 800 Oe in a χ vs $(T-\theta_{CW})^{-1}$ plot.

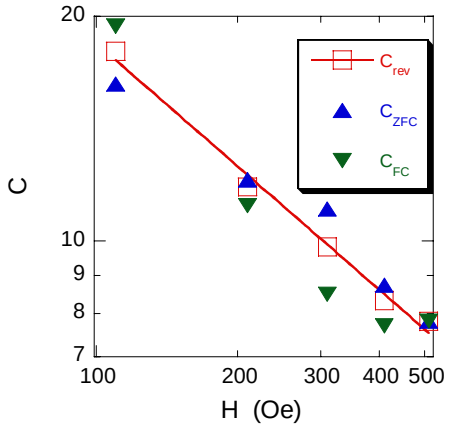

Fig. 5. Field dependence of the Curie Weiss constant C_{CW} for the reversible C_{rev}, ZFC C_{ZFC}, and FC C_{FC} susceptibilities. Notice that only C_{rev} obeys a power law vs field.

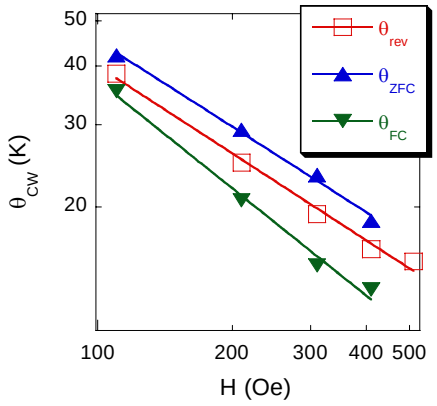

Fig. 6. Field dependence of the Curie Weiss temperature θ_{CW} for the reversible θ_{rev}, ZFC θ_{ZFC}, and FC θ_{FC} susceptibilities. Solid lines are linear fit with a power law.

The insertion of yttrium in $PrBa_2Cu_3O_{7-\delta}$ dilutes the Pr−Pr AFM exchange energy and releases holes introducing the long range Coulomb interaction which frustrates the AFM order due to the charge-spin coupling. It results a spin randomness, which in the presence of quenched disorder gives rise to a cluster spin glass [20]. The clusters are characterized by an antiferromagnetically ordered kernel and a border domain, the antidomain wall (ADW), over which the staggered magnetization changes the orientation. The frustration is collected within ADW's, hence, they carry a finite magnetic moment but also provide favourable channels for hole motion [21] and pairing. Superconductivity sets in for $x \le x_c$ when the burgeoning superconducting islands correlate their phases across the whole sample.

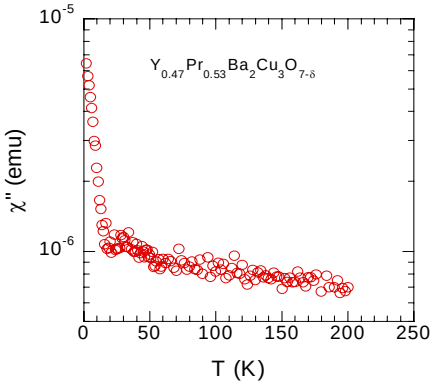

Fig. 7. Temperature T dependence of the imaginary part χ'' of the ac- susceptibility.

There is an interaction between the two coexisting phases, cluster glass and superconducting phase. In a pure glass system, the increase of the temperature reduces the charge correlation length. When it becomes smaller than the Debye length l_D the glass unfreezes [3]. The process is not trivial and is connected to the evolution of the complex energetic landscape of the glassy system and of the configurational entropy S_c. The latter is proportional to l_D^{-3}, hence decreases with

increasing temperature. Therefore, the number of the metastable states, which depends exponentially on S_c, decreases and the system evolves toward an extended viscous regime. A pure liquid state is obtained only when the vibrational motion in the potential minima becomes comparable to the charge correlation length. Finally, at even higher temperatures, the clusters themselves begin to disintegrate continuously, starting with those having low excitation energy, releasing the individual spins to the nascent paramagnetic sea up to creating a pure paramagnetic medium. Therefore, the viscous system consists of "floating" AFM clusters in a paramagnetic liquid. These clusters with an AFM kernel and moment carrying antidomain wall are equivalent to super-paramagnetic particles with an antiferromagnetic core and ferromagnetic magnetic shell [22].

The $M(T, H)$ depicts the evolution of the system through all available states. Starting from low temperatures and at fields up to 400 Oe, in ZFC regime, the response is first superconducting with diamagnetic susceptibility $\chi < 0$ (see Fig. 4). Above T_c, the superconducting contribution is limited only to the fluctuating diamagnetism whereas the contributions of the viscous liquid of clusters and free ion magnetic moments dominate at high T. The total magnetic moment reaches a maximum at T_G which is equivalent to the blocking temperature of the super-paramagnetic systems and further decreases in CW way. The moment of the moving clusters gives rise to a signal that dresses the ordinary paramagnetic response. This cluster signal is dependent on the warming/cooling protocol, hence, gives rise to a history dependent paramagnetism. In FC protocol, below T_G, $\chi_{FC} \sim T^{-\alpha}$ because the in the glass state ADW thickness increases with decreasing temperature due to the increase of the charge correlation length [3].

In conclusion, we find that the magnetic behavior of $Y_{0.47}Pr_{0.53}Ba_2Cu_3O_{7-\delta}$ could be explained starting from the evolution of superconducting and antiferromagnetic glass phases in interaction. An atypical hysteretic high-temperature regime was found to exist up to 200 K in relatively modest field. This regime consists of superconducting phase fluctuations and antiferromagnetic clusters interacting as viscous liquid phase or floating in a paramagnetic sea. We proposed a core-shell model to explain the irreversible contribution of the clusters.

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