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Possible weakly first-order superconducting transition induced by magnetic excitations in the YBCO system: A fluctuation conductivity study

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ABSTRACT

Fluctuation conductivity is experimentally studied in the genuine critical region near the superconducting transition of $YBa_2Cu_3O_{7-\delta}$, $YBa_2Cu_{2.985}Fe_{0.015}O_{7-\delta}$ and $Y_{0.95}Ca_{0.05}Ba_2Cu_3O_{7-\delta}$ single crystal samples. Two fluctuation regimes where the electrical conductivity diverges as a power-law of the reduced temperature were systematically observed. In the first regime, farther from the critical temperature T_c, the transition behaves as predicted by the thermodynamics of the three dimensional-XY (3D-XY) universality class characteristic of a second-order phase transition. In the asymptotic regime closer to T_c a power-law regime characterized by a much smaller exponent is observed. The smallest value ever reported for the fluctuation conductivity exponent in the high-T_c superconductors is obtained for the Fe- and Ca-doped systems. We suggest that the regime beyond 3D-XY is a crossover towards a weakly first-order transition induced by internal magnetic excitations.

1. Introduction

One of the most distinctive properties of the high temperature cuprate superconductors (HTCS) is the rather short and anisotropic coherence length, ξ, of the Ginsburg–Landau (GL) superconducting order parameter (SOP) when compared with that observed in conventional superconductors. In the HTCS, the component $\xi_c(0)$ is, in general, much smaller than the c lattice parameter. Therefore, the topology of the SOP becomes strongly dependent on the crystalline structure, chemical ordering and defect characteristics of these materials [1]. An important consequence of the small ξ is the occurrence of large regions around the critical temperature T_c dominated by thermal fluctuations in the electrical conductivity [2–4].

Thermal fluctuations have also been observed near T_c of the HTCS systems in properties such as the specific heat, magnetic susceptibility, thermal expansion, Hall effect, and others [5]. Far above T_c, the thermal fluctuation phenomena have been interpreted in terms of a scenario described by the Gaussian approximation to the GL theory [2–4,6]. Closer to T_c, detailed measurements of equilibrium [7,8] and transport [3,4,9] properties of the HTSC revealed the existence of a genuine critical fluctuation regime which is described by the three-dimensional-XY (3D-XY) universality class. Particularly, in the electrical conductivity this regime is identified by the critical exponent $\lambda_{CR} \sim 0.33$ [4,9–11]. This value is expected to occur in the particular case where the dynamics is given by the model-E, in the classification of Hohenberg and Halperin [12]. However, electrical conductivity measurements in $YBa_2Cu_3O_{7-\delta}$ (YBCO) single crystals [9,11,13,14] and YBCO/Au composite thin films [15] reveal the existence of fluctuation regimes beyond 3D-XY. These "supercritical" (S-CR) regimes are characterized by exponents smaller than λ_{CR}. The physical origin of the S-CR fluctuations is still unclear, but the low values observed for the characteristic exponent $\lambda_{S\text{-}CR}$ suggest that the ultimate character of the YBCO superconducting transition in the HTCS is of the weakly first-order (WFO) type. Some scenarios were proposed with the aim of understanding the mechanism that could drive the superconducting transition of the HTCS into the WFO class [9,16,17].

In this work we study experimentally the effects of chemical impurities in the in-plane fluctuation conductivity of YBCO single

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crystals. In addition to a reference pure sample, YBCO crystals were grown having 0.5% of the Cu atoms substituted by Fe, or 5% of the Y atoms replaced by Ca. Particular attention is given to the effects of these chemical substitutions on the critical and S-CR fluctuation regimes. The obtained results furnish evidences in favor of the WFO scenario for describing the ultimate character of the superconducting transition in the studied crystals.

Previous results on X-ray, Mössbauer spectroscopy and electrical resistivity reported for $YBa_2(Cu_{1-x}Fe_x)_3O_{7-\delta}$ with $x < 0.02$, show that the substitutional Fe atoms are located at the Cu(1) sites (chain sites) [18–20] and T_c remains practically unaltered [21]. Besides, the Fe substitution causes an increase of the oxygen content in the YBCO structure [22]. This fact justifies the observation of local magnetic ordering in $YBa_2(Cu_{1-x}Fe_x)_3O_{7-\delta}$ polycrystalline samples [23]. Nuñez et al. [24] showed that in the low substitution limit, the uniformly distributed impurities stabilize in the low-spin Fe^{3+} state with an effective magnetic moment of 2.18 μ_B. In Ref. [25] authors show that polycrystalline samples of $YBa_2(Cu_{1-x}Fe_x)_3O_{7-\delta}$ with $x > 0.01$ undergo a magnetic transition at a characteristic temperature T_m ($T_m < T_c$) that increases with x without suppressing the superconducting state. The fact that a magnetic ordering is induced by replacing only 1% of the Cu atoms by Fe leads authors in Ref. [24] to suggest the existence of spin fluctuations with significant amplitude in the Cu sites of YBCO.

One of the relevant effects produced by the Ca substitution in YBCO superconducting properties is a marked reduction of the critical temperature [26–32]. Several hypotheses have been put forward to explain the role of the Ca atoms in this effect [27–30, 33–35]. In particular, Hatada et al. suggest that the $T_c(x)$ depression in $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ is associated with the enhancement of the antiferromagnetic correlations between Cu-spins located in adjacent Cu(2)–O superconducting planes [31]. These authors claim that, in a scenario of oxygen underdoping, in $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ samples, the c crystallographic lattice parameter of YBCO increases [31]. This augmentation, however, is accompanied by a reduction of the distance between adjacent Cu(2)–O superconducting planes, [31,33–35] which in turn strengths the antiferromagnetic correlations between the in-plane Cu(2) spins along the c-axis crystallographic direction [31,33–35].

2. Experimental procedures

Several pure YBCO, $YBa_2Cu_{2.985}Fe_{0.015}O_{7-\delta}$ (YBCO-Fe) and $Y_{0.95}Ca_{0.05}Ba_2Cu_3O_{7-\delta}$ (YBCO-Ca) twinned single crystals were grown by the self-flux method [36,37]. Selected single crystals were submitted to an additional oxygenation process at $T = 450\,^{\circ}C$ for ten days in order to achieve the optimal oxygen content. The crystal dimensions are (2.20 × 0.80 × 0.040) mm^3 for the YBCO sample, (1.50 × 0.36 × 0.035) mm^3 for the YBCO-Ca sample and (0.90 × 0.69 × 0.010) mm^3 for the YBCO-Fe sample.

The studied crystals were characterized by X-ray diffraction (XRD). The XRD data were collected with 0.02° steps at 1°/min in the angular range $10° \leq 2\theta \leq 100°$. The spectra showed only the (00l) lines as expected for well oriented samples. The values found for the $\boldsymbol{c}$-axis lattice parameter are: $\boldsymbol{c} = 11.66(\pm 0.01)$ Å for YBCO; $\boldsymbol{c} = 11.68(\pm 0.01)$ Å for YBCO-Ca, and $\boldsymbol{c} = 11.71(\pm 0.01)$ Å for YBCO-Fe. These values are in agreement with those reported in the literature [22,26,30,35,36].

The in-plane resistivity $\rho(T, H)$ was measured as a function of the temperature, in the presence of magnetic field, with a low-frequency *ac*-current technique which employs a lock-in amplifier as a null detector. A variable decade transformer was used in a compensating electric circuit [4]. Four inline electrical contacts were painted with silver paste on one of the sample's surfaces oriented parallel to the crystallographic *ab* plane. The applied current density intensity was kept in the range of $J \leq 1.5$ A cm^{-2}. Constant magnetic fields ($H \leq 500$ Oe) were applied parallel to $\boldsymbol{J}$. This field–current configuration was adopted to significantly reduce the effect of Lorentz force on the fluctuation conductivity results. Temperatures were measured within an accuracy of 2 mK using a Pt sensor. The $\rho(T, H)$ data were recorded as the temperature was increased or decreased in rates never exceeding 0.02 K/min. A large number of closely spaced experimental points were recorded in order to allow the numerical determination of the temperature derivative of the resistivity, $d\rho/dT$, in the temperature range encompassing T_c.

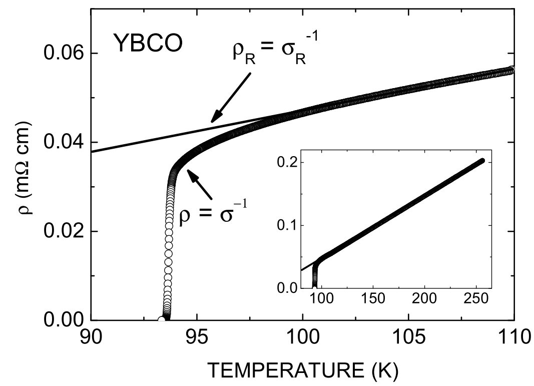

Fig. 1. Resistivity vs T for the pure YBCO sample in zero applied magnetic field. The regular term $\rho_R(T)$ is obtained from extrapolating to low temperatures the linear high-temperature behavior. The inset shows the resistivity in the whole measured temperature interval.

3. Method of analysis

We analyze the contribution of thermal fluctuations to $\sigma(T, H)$, $\{\sigma = \rho^{-1}\}$ by assuming that the fluctuation conductivity, $\Delta\sigma(T, H)$, diverges as a power law of the type

$$\Delta\sigma(T, H) = A\varepsilon^{-\lambda}, \tag{1}$$

where $\varepsilon = [T - T_c(H)]/T_c(H)$ is the field dependent reduced temperature, λ is the critical exponent, and A is a constant amplitude. The excess conductivity, $\Delta\sigma$, is estimated by subtracting the regular term $\sigma_R(T, H) = 1/\rho_R(T, H)$ from the measured conductivity $\sigma(T, H) = 1/\rho(T, H)$, that is,

$$\Delta\sigma = \sigma - \sigma_R = 1/\rho - 1/\rho_R. \tag{2}$$

As can be seen in the Fig. 1, the regular term ($\sigma_R = \rho_R^{-1}$) is obtained by extrapolating the high-temperature behavior [$\sigma_R(T) = 1/(a + bT)$, where a and b are constants] to temperature below T_c.

In the analysis of the fluctuation conductivity results, we adopt a method analogous to that applied by S. Kouvel and M.E. Fischer [38] to study critical phenomena in magnetic transitions. According to this method, we numerically determine a quantity identified as the logarithmic derivative of conductivity, $\chi_\sigma(T, H)$ which is defined as [4]

$$\chi_\sigma = -\frac{d}{dT}\ln\Delta\sigma. \tag{3}$$

Then, from Eqs. (1) and (3), we obtain

$$\chi_\sigma^{-1} = \frac{1}{\lambda}(T - T_C). \tag{4}$$

As previously shown [4,9], the identification of regimes described by straight lines in plots of $[\chi_\sigma(T, H)]^{-1}$ versus temperature allows the simultaneous determination of T_c and the critical exponent λ. The main sources of uncertainty in our data analysis come from the criteria adopted to estimate $\sigma_R(T)$ and from the numerical calculation of temperature derivatives, once