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Magnetic field induced excess conductivity in $Y_{1-x}Ca_xBa_2Cu_3O_{7-y}$/Ag composite superconductors

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Abstract : Temperature dependent electrical resistivity has been studied in a set of $Y_{1-x}Ca_xBa_2Cu_3O_{7-y}$/Ag composite bulk samples. Partial doping of Ca and composite formation with Ag modifies the microstructure and enhances the conductivity of the system. These two aspects, one modifying the intragranular and the other modifying the intergranular properties of the YBCO superconductor have a profound influence on SCOPF region. In the present work fluctuation induced excess conductivity in presence of magnetic field has been calculated from ρ *vs* T data in the framework of Aslamazov and Larkin (AL) and Lawrence and Doniach (LD) theories. In addition to the 3D Gaussian fluctuation on approaching the transition temperature from higher side, we observed a genuine critical regime which is consistent with the predictions of the full dynamic 3D-XY universality class with model-E dynamics. Still closer to T_c^{mf}, when magnetic field is applied, evidence is found for a fluctuation regime beyond 3D-XY which can be interpreted as the occurrence of ultimate first order character of the superconducting transition. In contrast to the 3D-XY model, the samples showed a reduced critical exponent on application of magnetic field. We attribute the reduction of the critical exponent at higher magnetic field to an enlarged or extended critical region.

Keywords : Excess conductivity, SCOPF parameter, critical fluctuation.

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1. Introduction

Superconducting and normal state properties of high T_c superconductors (HTSC) are greatly modified due to sample inhomogeneity and weak link networks. In $YBa_2Cu_3O_{7-y}$ (YBCO) type HTSCs, for example, the carrier density can be enhanced either by increase of oxygen content in the sample or by on-site substitution with dopants of lower valence state introducing sample inhomogeneity. Ca-doping in particular has created much interest in the scientific community as this doping turns an oxygen deficient YBCO from insulator to superconductor. Partial doping of Ca^{2+} for Y^{3+} in $Y_{1-x}Ca_xBa_2Cu_3O_7$ ($Y_{1-x}Ca_x$-123) system, reduces the optimum (but unstable) oxygen stoichiometry for highest T_c in YBCO. The consequent T_c reduction however is compensated in part by the additional charge carriers introduced by Ca-doping [1]. Besides acting as the supplier of charge carriers, Ca-doping decreases normal state resistivity and grain size, and increases critical current density of the system [2]. Thus, this doping provides stable superconductivity in YBCO superconductors with reduced but stable oxygen stoichiometry even though it introduces microscopic inhomogeneity in the framework of YBCO structure. In the bulk granular cuprates, this kind of inhomogeneity is mostly confined to the grains. The intergranular Josephson junctions however influence the global superconductivity in granular YBCO sample. In this respect, composite formation of YBCO with such noble metals like Ag, Au have been shown to modify the weak link networks and enhance the conductivity and critical current density of the system without affecting the superconducting transition temperature T_c [3].

At the onset of transition from normal to superconducting state, simultaneous creation and destruction of cooper pairs takes place leading to fluctuation of superconducting order parameter. This gives rise to an excess conductivity called paraconductivity in

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the superconducting order parameter fluctuation (SCOPF) region. Far above T_c^{mf} (where the $d\rho/dT$ *vs* T is maximum), experiments can be interpreted in terms of Gaussian fluctuation of the order parameter [4]. Closer to T_c^{mf}, careful studies of specific heat [5], penetration depth [6], and electrical conductivity [7] are now consistently revealing effects of genuine critical fluctuations for transition belonging to the 3D-XY universality class. It has been shown that 3D-XY thermodynamics is robust even under weak to moderate magnetic fields [7,8].

The excess conductivity has been seen in conventional superconductors in a small temperature range above T_c^{mf} [9]. In cuprates, the SCOPF occurs over a considerable larger temperature range because of their very short coherence length (ξ ~10–12 A within the layers) and relatively large penetration depth (λ ~1000-2000 A in the layers) [10]. Based on Aslamazov and Larkin [11] and Lawrence and Doniach [12] theories, attempts have been made to ascertain dimensionality of the fluctuations and probe the dimensionality crossover when the system approaches the 3D superconducting state from 2D normal state. It is however a complicated process to determine the dimensionality of the order parameter fluctuations in cuprate superconductors, as the adjacent CuO_2 layers are weakly coupled. In bulk YBCO superconductor for example, this coupling, whether Josephson or proximity type is not clearly known. The sample inhomogeneity in the form of both intrinsic and extrinsic defects further complicates the situation. Grain boundaries, voids and cracks, in sintered samples in particular, destroy the superconducting order parameter, phase coherence and influence the paracoherent region. The crossover temperature from one region to another in particular has been shown to depend on the transition width, which in turn is dictated by intergranular coupling [13,14]. Therefore, on a fundamental level, the excess conductivity that arises due to SCOPF in a large temperature region above T_c, is influenced by the weak links across the grain boundaries [15].

The application of magnetic field in HTSC results in penetration of magnetic flux lines into the material and resistive transition shifts to lower temperature side. The tail part of the resistive transition is suggestive of the weak links between the grains and is extremely sensitive to magnetic field. The flux creep and flux flow model may essentially hold well if the magnetic flux enters into the material in the vortex state by bending due to the grain boundary or by crystallographic anisotropy [16]. The melting of the vortex lattice gives rise to long tail in the temperature dependence of the resistivity [17]. Application of magnetic field thus leads to the broadening of resistive transition region and enlargement of the critical region.

Within the framework of Ginzburg-Landau mean field approximation [18], temperature dependence of excess conductivity is expressed as [19]

$$\Delta\sigma(T) = 1/\rho(T) - 1/\rho_R(T), \qquad (1)$$

where $\rho(T)$ is the measured electrical resistivity and $\rho_R(T)$ is the regular resistivity obtained by linear extrapolation of the resistivity data from room temperature (RT) to $T_{c_{onset}}$, assuming that the resistivity follows a linear temperature dependence at temperatures well above T_c. This is a valid assumption in the present case.

Since cuprate superconductors present a layered structure, dimensionality behaviour of the fluctuation appears depending on the relative values of temperature-dependent-coherence length along the c-axis (ξ_c). In the vicinity of the critical temperature ($T - T_c^{mf} << T_c^{mf}$) the longitudinal paraconductivity can be presented in the general form as [15]

$$\Delta\sigma(T) = e^2/16\hbar d\varepsilon\left\{1 + [2\xi_c/d]^2\varepsilon^{-1}\right\}^{1/2}, \qquad (2)$$

where ε the reduced temperature defined as $(T - T_c^{mf})/T_c^{mf}$, $\xi_c(0)$ is the zero-temperature coherence length along c-axis and d is the effective separation of CuO_2 layers. In the vicinity of T_c^{mf}, $\xi_c(T) >> d$. So the eq. (2) is reduced to

$$(\Delta\sigma)_{3D} = \left[e^2/32\hbar\xi_c(0)\right]\varepsilon^{-1/2}. \qquad (3)$$

Again further from T_c^{mf}, $\xi_c(T) << d$, eq. (2) reduces to

$$(\Delta\sigma)_{2D} = \left[e^2/16\hbar d\right]\varepsilon^{-1}. \qquad (4)$$

According to the classical AL (Aslamazov and Larkin) theory, $\Delta\sigma$ shows a temperature dependence of the form [11]

$$\Delta\sigma = A\varepsilon^{-\lambda}, \qquad (5)$$

where λ is the Gaussian critical exponent, the parameter $A = e^2/32\hbar\xi_c(0)$ with $\lambda = 0.5$ in 3D, and $A = e^2/16\hbar d$ with $\lambda = 1.0$ in 2D. Thus, λ in the above expression depends on the dimension (D) of the system and defines the dimension of the system as $\lambda = 2 - D/2$.

The determination of $\Delta\sigma$ involves the determination of ρ_R for temperatures near T_c^{mf} by extrapolating the high temperature behaviour of ρ as

$$\rho_R(T) = \rho_0 + (d\rho/dT)T, \qquad (6)$$

where ρ_0 and $d\rho/dT$ are temperature independent parameters.

Realizing the importance of Ag in modifying granularity characteristics of HTSCs and the importance of the latter on the SCOPF parameters, the present study aims to analyze the effect of magnetic field in the SCOPF region, particularly in the critical region in a set of $Y_{1-x}Ca_x$-123/Ag composite samples.

2. Experimental

$Y_{0.9}Ca_{0.1}$-123 samples were prepared from the chemicals Y_2O_3, $BaCO_3$, $CaCO_3$ and CuO by standard solid-state reaction. For the preparation of Ag composite bulk sinters, Ag_2O was mixed with $Y_{0.9}Ca_{0.1}$-123 powder before the final stage of sintering. Throughly ground $Y_{0.9}Ca_{0.1}$-123 was mixed with Ag_2O with appropriate proportions to obtain ($Y_{0.9}Ca_{0.1}$-123)/Ag 10 wt.% composites. The samples were pressed into pellets, annealed at 920°C for 12 hrs and cooled to 400°C where they stayed for 12 hrs with oxygen flowing. Then the samples were cooled to room temperature at a rate of 2°C min^{-1}. The samples were characterized by *d-c* four-probe resistivity measurement using a computer controlled data acquisition system [20]. The temperature dependent resistivity (ρ *vs.* T) data were taken by varying magnetic field in the range from 0–1.1 kG. The field is applied in perpendicular direction to the measuring current. The data were taken during heating the sample to room temperature. The heating was confined to 3 K per minutes.

3. Results and discussion

The phase formation and quality of the samples have been reported in our previous publication in which XRD spectrum shows the separate peaks for metallic Ag and SEM supports the segregation of Ag at the grain boundaries of $Y_{0.9}Ca_{0.1}$-123 grains [21].

Figure 1 shows the temperature dependence of resistivity (ρ *vs* T) behaviour of the sample with varying magnetic field up to 1.1 kG in constant measuring current density of 1 A cm^{-2}. The ρ *vs* T characteristics show a decrease of T_{c0} (R = 0), T_c^{mf} (where the $d\rho/dT$ *vs* T is maximum as shown in Figure 2) and increase of superconducting transition width (ΔT_c) with magnetic field. The ΔT_c is the full width at half maximum (FWHM) of the peak of $d\rho/dT$ *vs* T plot (Figure 2).

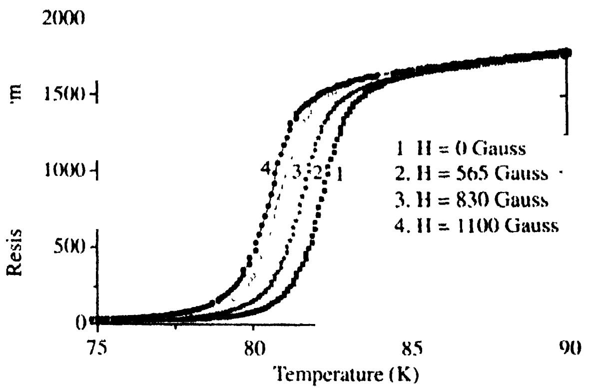

Figure 1. Temperature dependent resistivity of $Y_{1-x}Ca_xBa_2Cu_3O_{7-y}$/Ag composite bulk sample at different magnetic fields.

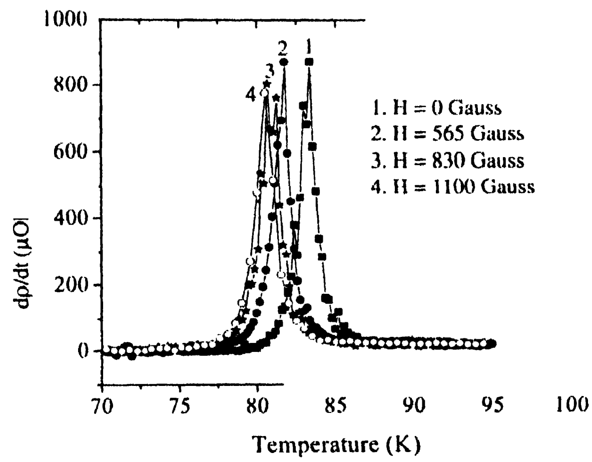

Figure 2. Temperature derivate of resistivity of the sample near T_c.

The decrease of T_{c0} with magnetic field suggests that it affects grain boundaries (Josephson junctions) more than the grains (Abrikosov junctions). It also leads to the resistive broadening near the mean field region in presence of magnetic field, which is clearly evidenced in Figure 2. The influence of the magnetic field on the inter-granular coupling is due to thermally activated flux creep or flux flow behavior, which accounts for the increase of ΔT_c at higher magnetic field. T_c^{mf} represents a lower limit for observing fluctuation regimes in normal phase. It can also be a good approximation for critical temperature in studies of conductivity fluctuation in the mean field region. As magnetic field increases, T_c^{mf} also moves to lower temperature side due to reduction of supercurrent which is evident from Figure 2. The disordering of vortices in the *ab*-plane of the layered structure of YBCO, due to thermal fluctuations, reduces the supercurrent [22].

Figure 3 shows the plot of ln $\Delta\sigma$ *vs* ln ε in absence of magnetic field with exponent λ_{3D} = 0.47 at the higher

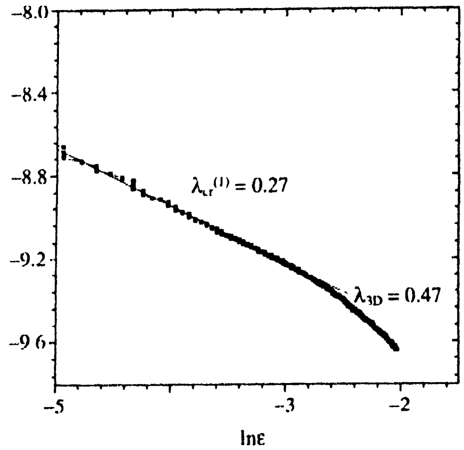

Figure 3. The plot of ln $\Delta\sigma$ *vs.* ln ε at magnetic field H = 0 Gauss.

temperature side. As λ = 0.5 [eq. (3)] indicates a 3D behavior of the order parameter fluctuation [23], we attribute this region with λ_{3D} = 0.47 to 3D Gaussian fluctuation. Decreasing the temperature further, we observed a cross over from the 3D Gaussian to the power law regime characterized by a small exponent $\lambda_{cr}^{(1)}$ = 0.27. This indicates the breakdown of mean field description for fluctuation of conductivity in our sample.

Figures 4, 5 and 6 show the variation of ln $\Delta\sigma$ with ln ε in different magnetic field ranging from 565 G to 1.1 kG respectively. Both the 3D and genuine critical fluctuation regime with exponents λ_{3D} and $\lambda_{cr}^{(1)}$ are still present. But λ_{3D} decreases from 0.47 to 0.30 and $\lambda_{cr}^{(1)}$ decreases from 0.27 to 0.19 with increase of magnetic field from 0 to 1.1 kG. In addition to these regions, we observe another narrow regime in presence of magnetic

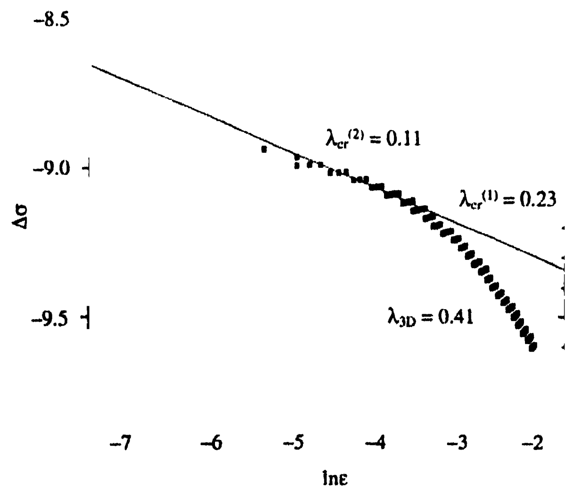

Figure 4. The plot of ln $\Delta\sigma$ *vs* ln ε at magnetic field H = 565 Gauss.

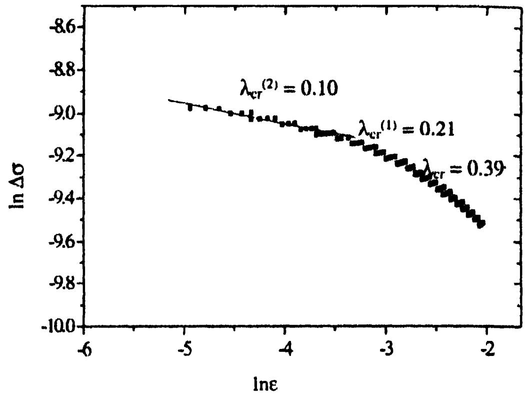

Figure 5. The plot of ln $\Delta\sigma$ *vs.* ln ε at magnetic field H = 830 Gauss

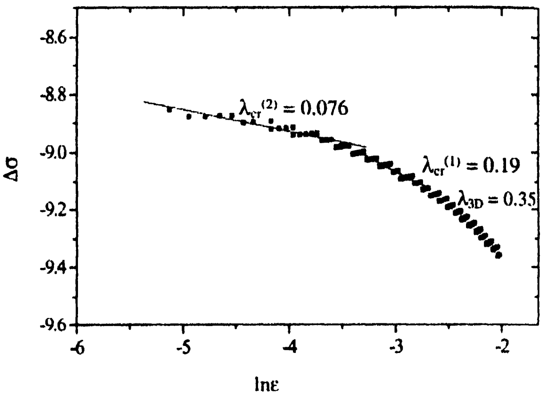

Figure 6. The plot of ln $\Delta\sigma$ *vs* ln ε at magnetic field H = 1.1 k Gauss

field where the exponent $\lambda_{cr}^{(2)}$ decreases from 0.11 to 0.076 with increasing magnetic field from 565 G to 1.1 kG. A similar result has been found in YBCO single crystals [24] and in thin films of YBCO/Au composites [25].

The critical exponent for fluctuation of conductivity is given by [26]

$$\lambda = \nu\,(2 + Z - D + \eta), \tag{7}$$

where ν is the coherence length critical exponent, Z is the dynamical exponent, η is the exponent for the order parameter correlation function. Renormalization group calculations for 3D-XY model gives ν = 0.67 and $\eta \cong 0$ [27]. Further, the model-E theory developed by Hohenberg and Halperin [28] for superfluid transition predicts that Z = 3/2. This yields a critical exponent $\lambda_{cr}^{(1)}$ = 0.33 for fluctuation conductivity. The theoretical prediction of this value is close to our experimentally determined critical exponent $\lambda_{cr}^{(1)}$. This scaling is therefore an indication of genuine critical fluctuation.

The additional regime characterized by the small exponent $\lambda_{cr}^{(2)}$ is observed beyond 3D XY-E scaling regime, corresponding to the exponents varying from 0.11 to 0.076 with increase of magnetic field from 565 G to 1.1 kG. This regime, which appears in the presence of magnetic field, may be associated with a weakly first-order superconducting transition still closer to T_c^{mf} indicating the cross over to type-I behaviour. This regime can be interpreted on the basis of scaling analysis for a first order transition [24]. This analysis predicts the coherence-length critical exponent $\nu = 1/D$, which, for $D = 3$, would corresponds to approximately one-half of its value for the 3D-XY model. Then, keeping the 3D XY-E values for z and η in eq. (7), $\lambda = 0.16$, which is in agreement with the experimental value for $\lambda_{cr}^{(2)}$ observed in our case.

The exponents characterizing different fluctuation regimes decrease with increase of magnetic field (Table 1). The lower values of exponents in higher magnetic

Table 1. Characteristics of $Y_{1-x}Ca_xBa_2Cu_3O_{7-y}$ 10 wt.% (x = 0.1) superconductors in the SCOPF region.

Field (Gauss)	T_c^{mf} (K)	ΔT_c (K)	λ_{3D}	$\lambda_{cr}^{(1)}$	$\lambda_{cr}^{(2)}$	$\xi(0)$ (Å)
0	83.35	1	0.45	0.27	-	3.00
565	81.75	1.5	0.38	0.23	0.11	2.92
830	80.65	1.6	0.35	0.21	0.10	2.83
1100	80.5	2.5	0.30	0.19	0.076	2.82

field are probably due to the fact that critical fluctuation dominates at higher magnetic field. It can be clearly seen that the superconducting phase is suppressed to lower temperature with increasing magnetic field (Figure 1). In addition, the magnetic field weakens the intergranular Josephson junctions, which is responsible for increasing ΔT_c. The increase in ΔT_c has been shown to influence the various critical regions and the 3D fluctuation conductivity [15] as is evidenced in our case.

Using the expression for Ginzberg number $\varepsilon_G = (T_G - T_c^{mf})/T_c^{mf}$, where T_G is the limit of the GL (Ginzberg-Landau) theory, we have estimated the GL number to be 0.065 for our system. GL coherence length, is calculated from the relation [29]

$$\varepsilon_G^{YBCO}/\varepsilon_G^{YCBCO} = [\xi(0)^{YCBCO}/\xi(0)^{YBCO}]^4, \quad (8)$$

where $\varepsilon_G^{YBCO} = 0.0038$ [7], $\xi(0)^{YBCO} = 6.1$ Å [30]. This relation gives us the GL coherence length ξ (0) in our system for different magnetic fields (Table 1). The estimated value is significantly lower than the reported value for a grain aligned YBCO. Lower value of the GL coherence length suggests that application of magnetic field reduces the coupling strength of the superconducting grains and increases the granularity character of the sample. This agrees with the report of Vieira *et al* [29].

4. Conclusion

Temperature dependent electrical resistivity study in a set of $Y_{1-x}Ca_xBa_2Cu_3O_{7-y}/Ag$ composite bulk samples has revealed the profound influence of the magnetic field on the superconducting order parameter fluctuation around the transition temperature. When the superconducting state is approached from above, we first observed a 3D Gaussian fluctuation regime. Bellow this regime a full dynamic 3D-XY fluctuation regime with exponent $\lambda_{cr}^{(1)}$ is clearly identified. In the presence of magnetic field when we approach further towards T_c^{mf}, a new scaling regime is observed within the critical region corresponding to a power law with a smaller exponent $\lambda_{cr}^{(2)}$. This gives the concept of a weakly first order normal to superconducting transition. The GL coherence length estimated in the Ca-doped YBCO/Ag composite system decreases with increase of magnetic field suggesting the reduction of coupling strength of the superconducting grains and increase of granularity character of the sample at higher magnetic fields.

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