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1992 J. Phys.: Condens. Matter 4 7891

(http://iopscience.iop.org/0953-8984/4/39/004)

J. Phys.: Condens. Matter 4 (1992) 7891–7898. Printed in the UK

The effect of fluctuations on the electrical transport behaviour in $YBa_2Cu_3O_{7-x}$

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Received 15 April 1992, in final form 10 June 1992

Abstract. The excess conductivity behaviour of highly oriented $YBa_2Cu_3O_{7-x}$ thin films prepared by both coevaporation and laser ablation has been studied in detail in the reduced-temperature range $9 \times 10^{-4} < t < 1$. The excess conductivity in all the films studied was found to diverge sharply near T_c, in agreement with the conventional mean-field thoery. However, the detailed temperature dependence could not be fitted to either the power-law or the logarithmic functional forms as predicted by the theory. The excess conductivity of all the films was found to be exponentially dependent on the temperature over nearly three decades for $9 \times 10^{-4} < t < 10^{-1}$, in contradiction to the mean-field theory.

1. Introduction

The rounding of the superconducting phase transition has been conventionally attributed to fluctuations in the magnitude and lifetime of the order parameter. By considering fluctuations of magnitude less than the order parameter in the Ginzburg–Landau (GL) theory, a critical temperature region was predicted in which the GL theory will not be valid. For temperatures greater than the critical limit the excess contribution $\Delta\sigma$ to electrical conductivity is estimated using the mean-field value of the order parameter in the time-dependent GL (TDGL) theory. The excess conductivity $\Delta\sigma$ is defined as $\sigma_{\rm exp} - \sigma_{\rm calc}$ where $\sigma_{\rm exp}$ is the experimentally observed electrical conductivity and $\sigma_{\rm calc}$ the conductivity due to normal-electron scattering alone in the absence of superconducting fluctuations. It is found to follow a power-law-type temperature dependence given by

$$\Delta\sigma(T) \propto (t)^{-\eta} \tag{1}$$

where t is the logarithm of the reduced temperature given by $\ln(T/T_c^{\rm mf}) \simeq [(T - T_c^{\rm mf})/T_c^{\rm mf}]$ for $t < 1$ ($T_c^{\rm mf}$ is the mean-field transition temperature) and η a constant which depends on the dimensionality of conduction [1]. For three-dimensional (3D) conduction it is $\frac{1}{2}$ and for two-dimensional (2D) conduction it is 1. Hence the temperature dependence of $\Delta\sigma$ has been extensively studied in order to determine η and, thus, the dimensionality of order parameter fluctuations, the nature

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of contributions to excess conductivity and also to estimate the coherence length. In the case of $YBa_2Cu_3O_7$, equation (1) has been extensively used to determine the dimensionality of $\Delta\sigma$ in the range $10^{-3} < t < 1$ and it has been reported to be 3D, 2D and quasi-2D crossing over to a 3D behaviour close to T_c [2]. Recently, however, it was found that $\Delta\sigma$ does not exhibit the classical power-law dependence on t in the mean-field regime but instead has a logarithmic dependence [3]. The deviation from normal behaviour of the specific heat of an untwinned single crystal was also found to exhibit a logarithmic temperature dependence in the range $10^{-4} < t < 10^{-1}$ by Regan *et al* [4]. The contribution of fluctuations to diamagnetic susceptibility in bulk pellets on the other hand was found to obey the predictions of conventional GL theory [5]. Howson *et al* [6] have studied the variation in thermoelectric power up to T_c in single crystals and found that it exhibits an anomalous peak near T_c because of the presence of 3D divergent fluctuations. This large body of experimental results clearly indicates that the phenomenon of fluctuations and the length of the critical region in the oxide superconductors is still not completely understood.

In the present work we report the systematic study of $\Delta\sigma$ in highly oriented $YBa_2Cu_3O_{7-x}$ thin films prepared by two different techniques: coevaporation and laser ablation. The study of fluctuation effects requires the background or normal-state contribution to the overall conductivity to be accurately determined. Hence the normal-state behaviour was analysed in terms of both the linear metallic conduction phenomenon and the more recent resonating-valence-bond (RVB) model [7]. It was found that the film with the lowest room-temperature resistivity follows metallic conduction behaviour while the other films follow the RVB model. The temperature variation in $\Delta\sigma$ for all the films was found to deviate completely from that predicted by the conventional GL-based models.

2. Experimental methods

The thin films of $YBa_2Cu_3O_{7-x}$ were deposited onto $SrTiO_3$ (100) substrates by two methods: coevaporation and laser ablation. In the coevaporation, Y and Cu were electron beam evaporated while BaF_2 was resistively evaporated onto a cold substrate in an oxygen ambient. The thickness of the as-deposited film is 0.5 μm. The films were later annealed at 850 °C in wet flowing oxygen to form the superconducting phase. In the case of laser ablation the film (0.3 μm thick) was deposited onto a heated substrate in an oxygen atmosphere from a sintered $YBa_2C_3O_7$ target. Structural characterization of the films was done by scanning electron microscopy and large-angle x-ray diffraction. The DC transport behaviour as a function of T was studied by the standard four-probe method. Thin Au wires attached to the film surface by In solder were used as leads for electrical characterization. These provided very-low-resistance ohmic contacts to the film surface. In the transition region, data points were taken at 0.2 K intervals to facilitate accurate analysis. The normal-state conductivity σ_{calc} for $T < 110$ K was determined by extrapolation of the regression-fitted data in the range $110 \text{ K} < T < 180$ K.

3. Results

The microstructure of the two coevaporated films (C1 and C2) were completely different although they were deposited and annealed under apparently identical conditions.

Film C1 has long cylindrical grains of about 0.25 μm diameter. The grains are highly oriented with their a–b plane along the film plane. Film C2, however, has a basket-weave-type grain morphology with an aspect ratio of about 16 in the film plane. The x-ray diffraction spectrum shows that the film has both c-axis- and a–b-axis-aligned grains along the film normal. The a–b-axis-aligned grains, however, were restricted to the top surface of the film [8]. The laser-ablated film (L) also shows a highly oriented cylindrical grain morphology similar to that of film C1. The film has a mirror-like smooth surface morphology, indicating that the surface roughness is much less than those of the coevaporated films.

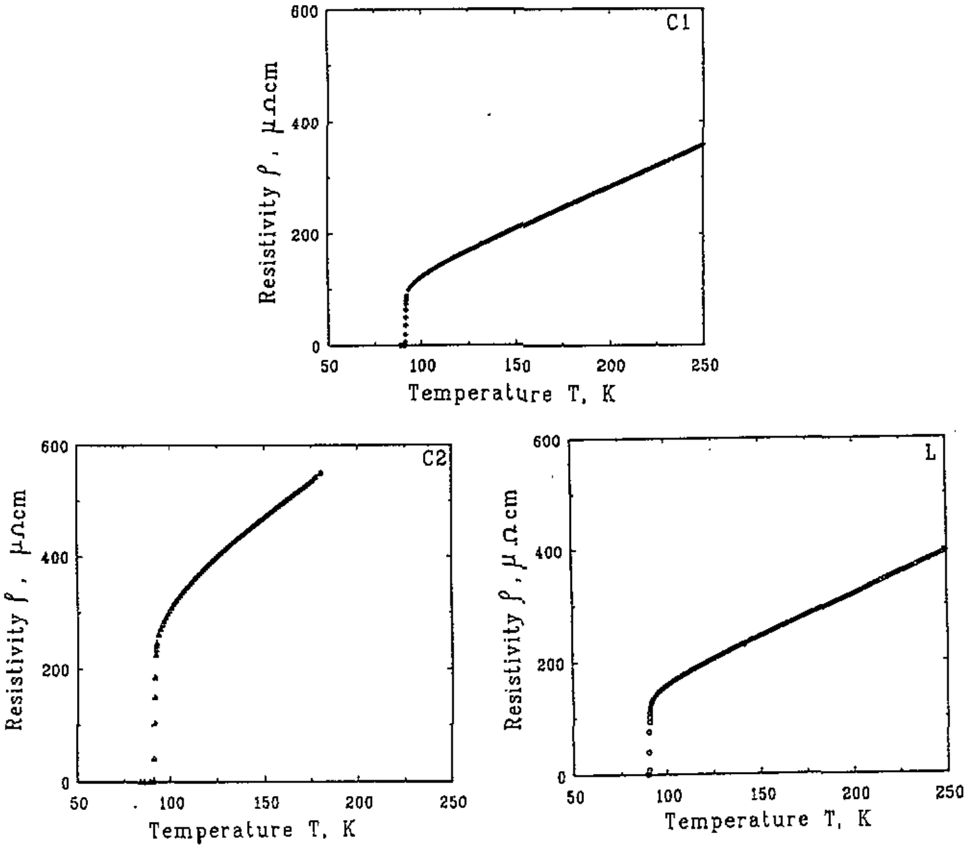

Figure 1. The variation in the resistivities ρ of the three films with temperature T.

The variation in the resistivities ρ with temperature of the three films C1, C2 and L shows a sharp transition into the superconducting state (figure 1). The transition width, defined as the width at half-maximum of the temperature-derivative curve, is about 0.4 K for all the films. The parameters that are important in any systematic study of $\Delta\sigma$ are

(i) determination of the mean-field transition temperature T_c^{mf} and
(ii) determination of the background or normal-state transport contribution to the overall conductivity.

It has been shown that the inflection temperature in the ρ–T curve can be approximated as the mean-field transition temperature [9, 10]. This criterion was used in the determination of T_c^{mf} for the films and is given in table 1.

Table 1. The zero-resistance temperature $T_c(0)$, mean-field transition temperature T_c^{mf} and the transition width $\Delta T_c(0)$ to zero-resistance state of the three films.

Sample	$T_c(0)$ (K)	T_c^{mf} (K)	$\Delta T_c(0)$ (K)
C1	91.1	91.5	0.4
C2	90.5	91.4	0.5
L	90.2	90.5	0.3

4. Discussion

The low-temperature superconductors are of strong-coupling BCS type and their normal-state behaviour can be estimated using a linear temperature dependence for ρ based on the conventional electron scattering mechanisms. However, the scattering mechanisms in the normal state of the oxide superconductors are not clearly known. Both the Fermi-liquid-based models which predict a linear temperature dependence and the non-Fermi-liquid-based models are currently used to fit the normal-state transport behaviour [11]. According to the non-Fermi-liquid-based RVB model, the charge carriers are assumed to be confined to the Cu–O a–b planes of the crystal, thus leading to metallic conduction behaviour along the planes and phonon-activated hopping conduction in between the planes. The overall resistivity $\rho(T)$ in such an hypothesis is given by an expression of the form

$$\rho(T) = aT^{-1} + bT \tag{2}$$

where a and b are temperature-independent constants. In the present analysis, the ρ–T data of all the films were fitted to both the linear temperature dependence of the type $\rho(T) = \rho(0) + bT$ and the RVB dependence, equation (2), using the least-squares regression-fitting routine in the range 110 K $< T <$ 180 K. It was found that the best fit to film C1 was the linear relation while that for films C2 and L was the RVB-type relation. However, only the linear coefficient b of the two films C2 and L is in reasonable agreement with that predicted by the RVB model. The values of a in equation (2), 442.1 and 925.8 $\mu\Omega$ cm K, are orders of magnitude lower than the predicted values. The linear temperature coefficients b for the three films are 0.186 $\mu\Omega$ cm K^{-1}, 0.294 $\mu\Omega$ cm K^{-1} and 1.584 $\mu\Omega$ cm K^{-1}, respectively. These values are well within the average values observed for single crystals [2, 3] and indicate the phase purity of the films. Using the simple Drude relation for metallic conduction given by $\rho(T) = (3\pi\hbar/2)/e^2k_F^2l$ where l is the quasi-particle mean free path, e the electron charge, $\hbar$ the Planck constant and $k_F \simeq 4.46 \times 10^7$ cm^{-1} [12] the Fermi wavevector, the 'metallic parameter' k_Fl can be estimated for the three films. These were found to be 17, 7 and 14 for films C1, C2 and L, respectively, at 120 K, indicating that all the three films are very much on the metallic side of the Ioffe–Regel limit. This clearly shows that the microscopic conduction mechanism in these materials above T_c is not completely understood, although a mathematical fit to the RVB model can be obtained. Recently, on the basis of mid-infrared phonon spectroscopy [13] and transport [14] studies on polycrystalline pellets it has been reported that the contribution from fluctuations persists up to temperatures as high as $2T_c$. This corresponds to the 'fluctuation onset' temperature, indicating that the lifetime of the superconducting fluctuations is finite and large even at $2T_c$, for which there is no direct experimental evidence at present.

The excess conductivity $\Delta\sigma(T) = \sigma_{\mathrm{exp}}(T) - \sigma_{\mathrm{calc}}(T)$ determined using the relation $\rho(T) = \rho(0) + bT$ for film C1 and equation (2) for films C2 and L was found to diverge sharply as T approaches T_c^{mf}. This is in qualitative agreement with the conventional theory which predicts a divergence of the magnitude of the order parameter fluctuations at T close to T_c^{mf}. The critical temperature region t in which the TDGL theory is not applicable can be estimated using typical values for $YBa_2Cu_3O_7$; $T_c^{\mathrm{mf}} = 91$ K, the zero-temperature upper critical field $H_{c2}(0) = 674$ T and the GL parameter $K = 200$ [15], and therefore t is found to be about 2×10^{-2}. For $t \geqslant 2 \times 10^{-2}$, $\Delta\sigma(T)$ can in principle be determined using equation (1) (power-law dependence of $\Delta\sigma$ on t). Although the rate of decay of the fluctuating superconducting pairs is explicitly considered in obtaining equation (1), their effect on the quasi-particle conductivity is not considered. An additional term has been proposed to equation (1) by Maki and Thompson (as quoted by Skocpol and Tinkham [16]) to account for the effect of fluctuations on the quasi-particle conductivity and it was found to be four times equation (1) in the case of 3D conduction and $(e^2/8\hbar d)[(t-\delta)^{-1}\ln(t/\delta)^{-1}]$ for 2D conduction, where δ is the pair-breaking parameter and d the film thickness. The criterion for 2D conduction is $d/\xi(T) \ll 1$ where $\xi(T)$ is the superconducting coherence length. In the present case, even $d/\xi(0)$ for all the three films is much greater than unity and hence 2D conduction can be completely ruled out. The addition of an extra term to equation (1) changes only the magnitude of $\Delta\sigma(T)$, leaving the power-law temperature dependence intact. In the present work, however, $\Delta\sigma(T)$ for all the three films determined from the experimental data does not show a power-law dependence on t as predicted in the range $9 \times 10^{-4} < t < 4 \times 10^{-1}$; this can be clearly seen in figure 2. It has a continuously changing curvature which has been observed earlier. However, the previous reports have inferred changes in the dimensionality of electrical transport on the basis of linear fits to small portions of the curve [2].

The above method of analysis relies on the accurate determination of T_c^{mf}. In the case of oxide superconductors, the fluctuation effects on the conductivity are spread over a large temperature range compared with the conventional superconductors because of their extremely short coherence length ξ and the high value of $T_c(0)$. Hence the accurate determination of T_c^{mf} is difficult. An alternative method of analysing the fluctuation effects which does not depend on T_c^{mf} has been used in the case of Tl–Ba–Ca–Cu–O thin films and single crystals [17, 18]. According to this method, equation (1) can be rewritten as

$$[\Delta\sigma(T)]^{-1/\eta} = D^{-1/\eta}[(T - T_c^{\mathrm{mf}})/T_c^{\mathrm{mf}}] \tag{3}$$

where the constant D is given by $e^2/32\hbar\xi(0)$ for 3D conduction and $e^2/16\hbar d$ for 2D conduction. Differentiating and rearranging equation (3) gives

$$\ln[-\mathrm{d}(\Delta\sigma)/\mathrm{d}T] = \ln(D^{-1/\eta}/T_c) + (1 + 1/\eta)\ln(\Delta\sigma). \tag{4}$$

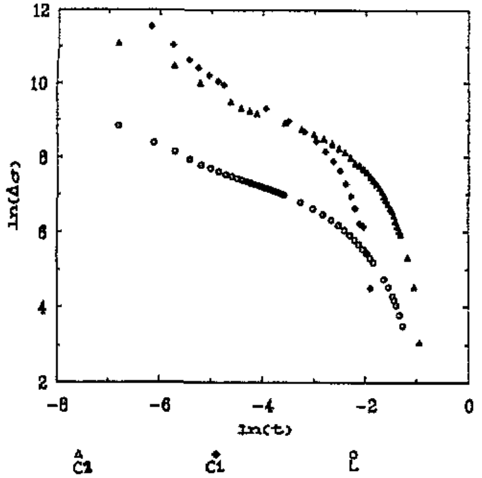

Figure 2. The excess conductivity $\Delta\sigma$ as a function of the reduced temperature $t = \ln(T/T_c^{mf})$ for all three films. It can be seen that a 'power-law' fit is not feasible except in small ranges of t. Δ, C2; ♦, C1; ○, L.

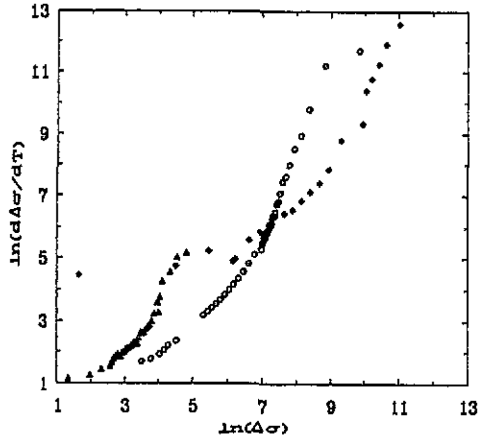

Figure 3. The excess conductivity $\Delta\sigma$ data represented according to equation (4), which is independent of T_c^{mf}. The presence of continuous curvature in the whole temperature range indicates a clear deviation from power-law behaviour.

The dimensionality η can be deduced from the slope of the $\ln[-\mathrm{d}(\Delta\sigma)/\mathrm{d}T]$ versus $\ln(\Delta\sigma)$ plot and using equation (4). Figure 3 shows $\Delta\sigma$ plotted according to this modified scheme. It can be clearly seen that even this alternative methodology which is independent of T_c^{mf} does not give conclusive evidence for the dimensionality of conduction in these films. Even according to this modified scheme of analysis the data exhibit a continuous curvature in the whole range and the dimensionality can be inferred only by fitting small portions of the excess conductivity. The recent electrical transport, mid-infrared phonon spectrum and heat capacity studies have clearly shown that the 'onset' temperature for fluctuations can be as high as about $2T_c$. The onset of fluctuations in the electrical transport behaviour has been attributed to the quasi-2D Maki–Thompson correction factor which has a logarithmic temperature dependence [3, 14]. The $\Delta\sigma$-values in the present work, however, could not be fitted satisfactorily to a logarithmic temperature dependence. The $\Delta\sigma(T)$ data are replotted as shown in figure 4 and it can be clearly seen that $\Delta\sigma(T)$ has an exponential dependence on t: $\Delta\sigma(T) \propto \exp(t^{-\alpha})$ where α is the slope in the range $9 \times 10^{-4} < t < 10^{-1}$ for all the three films. This clearly illustrates two important points.

(i) The TDGL theory underestimates the critical temperature region by at least an order of magnitude.

(ii) The mean-field approximations are not valid in the case of $YBa_2Cu_3O_{7-x}$.

The underestimation of the critical region has been attributed to the large contribution of the higher-order fluctuation corrections [19]. In the critical region, $\Delta\sigma$ is predicted to diverge as t goes to 0 with a temperature dependence similar to that in the mean-field region but with an exponent different from η based on the 3D XY model [20]. The results of the present work, however, cannot be understood even according to these models.

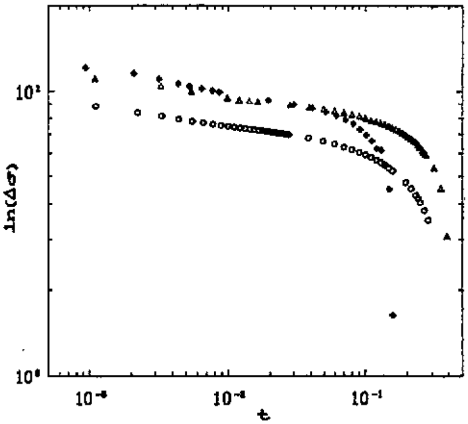

Figure 4. Log-normal plot of $\Delta\sigma$ versus t, showing the exponential temperature dependence of $\Delta\sigma$ for all the films in the range $9 \times 10^{-4} < t < 10^{-1}$.

5. Conclusions

The macroscopic and microscopic properties of the oxide materials in their superconducting state are being extensively studied. Many models have already been proposed to explain these properties. However, the normal-state behaviour remains the least studied to date. The only phenomenological model that has been proposed to explain the normal-state electron transport behaviour is the RVB model. The results of the present work indicate that it is insufficient to explain the transport behaviour above T_c. The linear and hopping coefficients obtained for the two films which obey the RVB expression for conductivity are much lower than the values predicted by the model.

The excess conductivity in the mean-field region of the three films studied does not obey the temperature dependence predicted by the TDGL theory. Even though such a behaviour has been observed before, the results are still fitted to the TDGL theory and the dimensionality of the electrical transport determined. However, we find that the excess conductivity is better represented by an exponential relation and that there is no model at present, macroscopic or microscopic, which can explain this type of behaviour. Hence the dimensionality of electrical transport is still inconclusive. The onset temperature for fluctuations observed in the present work, $t \simeq 0.1$, agrees with that observed by Regan *et al* [4], indicating that the length of the critical region is larger than that predicted by the theory. However, the functional dependence on temperature is found to be different.

Acknowledgments

SV wishes to acknowledge the US National Research Council for an Associateship at Lewis Research Center, NASA, and MAS acknowledges NASA for the support through grant NAG-440751. The authors acknowledge Professor J Scofield for the use of his evaporation system, J D Warner for the laser-ablated film, and T Harris and G Kitchen for their assistance. Critical reading of the manuscript by Dr Padma Satish is gratefully acknowledged.

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