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Thermodynamic fluctuation analysis in $Tl_2Ba_2Ca_{1-x}Y_xCu_2O_{8+y}$ ($x = 0$–0.25) system

A Poddar†, P Mandal‡ and S Das†

† Saha Institute of Nuclear Physics, Block-AF, Sector-I Bidhannagar, Calcutta-700 064, India
‡ Max-Planck Institut für Festkörperforschung, 25 Martyrs Avenue, BP 166, 38042 Grenoble, Cédex, France

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Abstract. We report systematic conductivity fluctuation measurements above the mean-field transition temperature T_c for $Tl_2Ba_2Ca_{1-x}Y_xCu_2O_{8+y}$ ($x = 0$–0.25). The excess conductivity has been analysed in terms of the Aslamazov–Larkin, Lawrence–Doniach (LD) and Maki–Thompson (MT) models. The analysis of the LD fit shows that the system is highly anisotropic and the interlayer coupling J decreases with decrease in the holes in the system. In the under-doped region the dependence of T_c on J seems to be more comparable with that in the optimally doped region. Incorporating the MT term into the LD model fits our data over quite a high-temperature range. We have also observed that the MT contribution increases with decrease in the interlayer coupling strength.

1. Introduction

The extremely short-coherence-length, quasi-two-dimensional character as well as the high superconducting transition temperature T_c of the cuprate superconductors [1] are their most distinctive properties and these make the phenomenology quite different in certain respects from that of the conventional low-T_c superconductors. As a consequence of the short coherence length and quasi-two-dimensional nature of these systems there exists a broad temperature region over which the effects of thermodynamic fluctuation are manifested in several of their temperature-dependent properties such as resistivity [2, 3], susceptibility [4], specific heat [5], Hall effect [6] and magnetoresistance [7]. Since the interlayer coupling strength may be estimated [3] from the analysis of fluctuation-induced conductivity, it is quite interesting to determine the role of the interlayer coupling as T_c decreases with decrease in carrier concentration in the under-doped region of oxide superconductors. Recently, we have analysed [8] the excess conductivity of $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_{8+y}$ ($x = 0$–0.45) as a function of carrier concentration and observed that, in the low-hole-concentration region, T_c depends strongly on the interlayer coupling compared with that in the optimally and heavily doped region.

The fluctuation-induced enhancement in conductivity, termed excess conductivity or paraconductivity $\Delta\sigma$, was described in detail by Aslamazov and Larkin (AL) [9] and can be written as

$$\Delta\sigma_{AL} = A\varepsilon^{-\lambda} \tag{1}$$

where $\varepsilon = (T - T_c^{mf})/T_c^{mf}$ is the reduced temperature, λ is the critical exponent and A is a temperature-independent amplitude. For three-dimensional (3D) fluctuations, $A =$

$(e^2/32\hbar)[1/\xi(0)]$ and $\lambda = 0.5$ whereas, for two-dimensional (2D) fluctuations, $A = e^2/16\hbar d$ and $\lambda = 1.0$. $\xi(0)$ and d are the zero-temperature coherence length and the characteristic length, respectively, of a 2D system.

In high-T_c superconductors the coherence length along the c axis is much smaller than that in the a–b plane and such systems may be described as layered superconductors. For a layered superconductor with Josephson coupling between the adjacent layers, Lawrence and Doniach (LD) [10] calculated the fluctuation-induced conductivity $\Delta\sigma_{LD}$ of the layers which is given by

$$\Delta\sigma_{LD} = \frac{e^2}{16\hbar d}\frac{1}{\varepsilon}\left(1+\frac{J}{\varepsilon}\right)^{-1/2} \tag{2}$$

where $J = [2\xi_c(0)/d]^2$ is the interlayer coupling constant. From equation (2) it is seen that, for temperatures close to T_c^{mf}, $\Delta\sigma_{LD}(\varepsilon)$ diverges as $\varepsilon^{-1/2}$ (3D behaviour) while, for $T \gg T_c^{mf}$, $\Delta\sigma_{LD}(\varepsilon)$ diverges as ε^{-1} (2D behaviour). Thus the LD expression (equation (2)) predicts a crossover from 2D to 3D fluctuations; the crossover temperature is defined as

$$T_0 = T_c^{mf}(1+J) \tag{3}$$

and the slope of the $\ln(\Delta\sigma)$ versus $\ln\varepsilon$ curve at T_0 is -0.75.

It has been observed [11, 13] that for high-T_c oxide systems the experimental $\ln(\Delta\sigma)$ versus $\ln\varepsilon$ curve lies below the theoretical AL and LD expressions for $\Delta\sigma$ by a factor of C. For a c-axis-oriented $YBa_2Cu_3O_{7-\delta}$ thin film, Oh *et al* [11] obtained values of C lying within the range 3.0–7.0. These workers suggested that the factor C arises from a non-uniform current flow in their specimens caused by sample inhomogeneities on a submacroscopic scale. In our earlier communication [12] we also observed similar discrepancy between the theoretical and experimental $\ln(\Delta\sigma)$ versus $\ln\varepsilon$ curves of polycrystalline $TlBaCa_3Cu_3O_x$ samples. The estimated values of c lie in the range 10–31. Thus considering the factor C the modified AL and LD expressions for $\Delta\sigma$ of high-T_c oxide systems become

$$\Delta\sigma_{AL} = \frac{A}{C}\varepsilon^{-\lambda} = B\varepsilon^{-\lambda} \tag{4}$$

and

$$\Delta\sigma_{LD} = \frac{e^2}{16C\hbar d}\frac{1}{\varepsilon}\left(1+\frac{J}{\varepsilon}\right)^{-1/2} = D\frac{1}{\varepsilon}\left(1+\frac{J}{\varepsilon}\right)^{-1/2} \tag{5}$$

An additional indirect contribution to the excess conductivity arises from the scattering of the normal excitation by the fluctuating Cooper pairs and was estimated by Maki [14] and later modified by Thompson [15]. The direct acceleration of the fluctuation-induced superconducting pairs leads to the AL contribution. These superconducting fluctuations then decay into quasi-particles which in the presence of a weak pair-breaking effect may continue in a state of nearly equal and opposite momenta until they recombine to form fluctuating pairs. This indirect Maki–Thompson (MT) contribution is limited by strong inelastic scatterers and by the presence of pair-breaking interactions. The indirect MT contribution for layered superconductors was derived by Hikami and Larkin [16] and independently by Maki and Thompson [17] and, in the absence of magnetic field, is given as follows:

$$\sigma_{MT} = \frac{e^2}{8\hbar d\varepsilon(1-\alpha/\delta)}\ln\left(\frac{\delta}{\alpha}\frac{1+\alpha+(1+2\alpha)^{1/2}}{1+\delta+(1+2\delta)^{1/2}}\right) \tag{6}$$

where $\alpha = 2\xi_c^2(0)/d^2\varepsilon = J/2\varepsilon$, $\delta(= 16\xi_c^2(0)k_BT\tau_\phi/\pi^2 d\hbar)$ is the pair-breaking parameter and τ_ϕ is the phase relaxation time. Thus, when the factor C is taken into account, the total enhancement of the conductivity due to the presence of both LD and MT terms may be written as

$$\begin{aligned}\Delta\sigma &= \Delta\sigma_{\mathrm{LD}} + \Delta\sigma_{\mathrm{MT}} \\ &= \frac{e^2}{16C\hbar d}\left[\frac{1}{\varepsilon}\left(1+\frac{J}{\varepsilon}\right)^{-1/2} + \frac{2}{\varepsilon - J/2\delta}\ln\left(\frac{2\varepsilon\delta}{J}\frac{1+J/2\varepsilon+(1+J/\varepsilon)^{1/2}}{1+\delta+(1+2\delta)^{1/2}}\right)\right] \\ &= D\left[\frac{1}{\varepsilon}\left(1+\frac{J}{\varepsilon}\right)^{-1/2} + \frac{2}{\varepsilon - J/2\delta}\ln\left(\frac{2\varepsilon\delta}{J}\frac{1+J/2\varepsilon+(1+J/\varepsilon)^{1/2}}{1+\delta+(1+2\delta)^{1/2}}\right)\right].\end{aligned} \quad (7)$$

Experimentally the excess conductivity is determined by subtracting the normal conductivity σ_n from the experimental conductivity σ by using the expression

$$\Delta\sigma(\varepsilon) = \sigma - \sigma_n = \frac{1}{\rho(T)} - \frac{1}{\rho_n(T)} \quad (8)$$

where $\rho(\varepsilon)$ and $\rho_n(\varepsilon)$ are the measured and the normal-state resistivities, respectively, of the sample.

Thus, for the analysis of the excess conductivity, precise knowledge of the mean-field transition temperature T_c^{mf} and normal-state conductivity σ_n is necessary. The standard and mostly used procedures [3, 11, 18, 19] for the determination of T_c^{mf} are

(i) by extrapolating the linear part of the $(\Delta\sigma)^{1/\lambda}$ versus T curve near T_c.

(ii) from the best linear fit of $\ln(\Delta\sigma)$ versus $\ln\varepsilon$ data with T_c^{mf} as the free parameter and

(iii) by taking the temperature at which a sharp peak is observed in the $d\rho/dT$ versus T plot.

In the absence of any quantitative model for the determination of ρ_n for the oxide superconductors and following the observed linear T dependence of resistivity up to a very high temperature, most groups estimated ρ_n by fitting the linear high-temperature ($2T_c - 300$ K) resistivity data according to the expression

$$\rho_n(T) = a + bT \quad (9)$$

where a and b are constants.

It should be mentioned here that a poor choice of T_c^{mf} may result in a wrong conclusion regarding the dimensionality of the system. However, one can avoid this problem by using the derivative technique originally proposed by Testardi *et al* [20] and later used in [3]. From equations (1) and (8), one obtains

$$\Delta\sigma(\varepsilon) = \frac{1}{\rho(T)} - \frac{1}{\rho_n(T)} = A\varepsilon^{-\lambda}.$$

Differentiating this expression with respect to temperature and rearranging the terms give

$$\frac{1}{\rho^2}\frac{d\rho}{dT} - \frac{1}{\rho_n^2}\frac{d\rho_n}{dT} = \frac{\lambda(A)^{-1/\lambda}}{T_c^{mf}}\left(\frac{\rho_n-\rho}{\rho\rho_n}\right)^{-(1+1/\lambda)} = \frac{\lambda(A)^{-1/\lambda}}{T_c^{mf}}(\Delta\sigma)^{-(1+1/\lambda)}. \quad (10)$$

The exponent λ can be determined from the slope of the log–log plot of the left-hand side of equation (10) and $\Delta\sigma$.

The fluctuation-induced enhancement of conductivity in oxide superconductors including oriented polycrystalline thin films as well as single-crystal samples has been reported by many groups [2, 3, 11]. Experimental investigation [2, 11] regarding the dimensionality of the conductivity of $YBa_2Cu_3O_{7-\delta}$ revealed various conclusions, namely 2D, 3D as well as a crossover from 2D to 3D. In contrast, resistivity measurements on high-quality Bi–Sr–Ca–Cu–O and Tl–Ba–Ca–Cu–O materials [3, 4, 21] support clearly the 2D nature of fluctuations and also a crossover from 2D to 3D near T_c^{mf}. This result implies that the coupling between CuO_2 planes in Bi- and Tl-based superconductors is much weaker than in the 1:2:3 system. For $YBa_2Cu_3O_7$ it has been observed [22] that even in the absence of magnetic field the contribution of the MT term is not negligible and as a result the fluctuation conductivity extends to quite high temperatures. For this system the estimated phase relaxation time τ_ϕ was found to lie [7, 23, 24] in the range 10^{-13}–10^{-14} s at 100 K. From magnetoconductance and a zero-field excess-conductivity analysis of a $Tl_2Ba_2CaCu_2O_x$ thin film, Kim *et al* [25] reported a linear temperature dependence for the phase relaxation rate with $1/\tau_\phi \simeq (3.5 \pm 0.5) \times 10^{13}$ s^{-1} at 100 K. Ravi and Seshu Bai [26] fitted with excess-conductivity data of the Bi system to the sum of the LD and MT terms with τ_ϕ, $\xi_c(0)$ and d as free parameters and obtained $\tau_\phi \simeq 3.7 \times 10^{-16}$ s and 11.3×10^{-16} s at 100 K for the 85 K and 110 K phases, respectively, of the system.

In this paper we have investigated the role of interlayer coupling on the superconductivity of $Tl_2Ba_2Ca_{1-x}Y_xCu_2O_{8+y}$ (x = 0–0.25) from the systematic analysis of the AL, LD as well as the MT form for the fluctuation conductivity of the system.

2. Preparation and experimental details

Two different processes were followed for the preparation of $Tl_2Ba_2Ca_{1-x}Y_xCu_2O_{8+y}$ (x = 0–0.25) samples. The first process commonly known as matrix method has been described in our earlier report [27]. Using this method we have prepared x = 0–0.25 samples which are referred to hereafter as A1 (x = 0), A2 (x = 0), A3 (x = 0.1), A4 (x = 0.2) and A5 (x = 0.25). In the second process, appropriate amounts of Tl_2O_3, BaO_2, CaO, Y_2O_3 and CuO were finely mixed and pressed into pellets. The pellets were wrapped in Pt foil and then sintered at about 840 °C for 6–8 h in the presence of oxygen. After grinding, they were sintered again at 870–880 °C for about 5 h in flowing oxygen and then cooled to room temperature at the rate of 45 °C h^{-1}. The samples prepared following this method are referred to as B1 (x = 0), B2 (x = 0) and B3 (x = 0.2). It should be mentioned here that four $Tl_2Ba_2CaCu_2O_{8+y}$ (A1, A2, B1 and B2, i.e. x = 0) and two $Tl_2Ba_2Ca_{0.8}Y_{0.2}Cu_2O_{8+y}$ (B3 and A4, i.e. x = 0.2) samples show different superconducting transition temperatures, which might be due to the presence of different amounts of oxygen in each sample. In table 1, we have arranged all the samples in the order depending upon the superconducting transition temperature. X-ray powder diffraction pattern shows that all the samples possess a single phase. In table 1 we have presented the values of d (the average separation between two adjacent CuO_2 layers) corresponding to the c-axis lattice parameter of each sample.

The temperature-dependent resistivity of the samples was measured by the standard four-probe technique using silver paint contacts. A constant DC current (about 1–3 mA) from a Lake Shore constant-current source was passed through the sample and the voltage drop across the sample was measured with a Keithley (181) nanovoltmeter. For precise

Table 1. Values of T_c^{mf} (obtained from $d\rho/dT$), a, b, ρ(300 K) and d (obtained from the c-axis lattice parameters) for $Tl_2Ba_2Ca_{1-x}Y_xCu_2O_8$ ($x = 0$–0.25) samples.

Sample	T_c^{mf} (K) obtained from method (iii)	a (mΩ cm)	$b(10^{-3})$ (mΩ cm K^{-1})	ρ (300 K) (mΩ cm)	d (Å)
A1	104.7	0.9165	5.5514	2.58	14.69
A2	106.0	1.1439	6.4878	3.10	14.68
A3	103.5	2.2870	2.9926	3.19	14.67
B3	103.0	1.4613	2.8722	2.33	14.67
B1	100.2	0.7539	11.5960	4.25	14.66
B2	95.7	3.5129	15.6350	8.22	14.64
A4	96.5	6.8903	10.8466	10.14	14.65
A5	84.8	10.8602	37.2101	22.00	14.62

resistivity measurements, low-resistance contacts to the sample were made by annealing the sample with silver paste in the desired configuration at 300–400 °C for 1 hr in flowing oxygen. The sample temperature was measured with a chromel–alumel thermocouple placed very near to the sample. The average size of the samples is 9 mm × 3 mm × 1.0 mm and the geometry of the four-probe measurement, i.e. the average distances between the voltage and current pads are about 8 mm and about 6 mm, respectively.

3. Results and discussion

The temperature dependence of resistivity for $Tl_2Ba_2Ca_{1-x}Y_xCu_2O_{8+y}$ samples (A1, A4 and A5) are shown in figure 1. All the samples show a linear variation in the resistivity above 200 K. To estimate the normal-state resistivity ρ_n we consider the following procedure. We choose different sets of random (ρ, T) data points from the temperature interval 200–300 K and, for each set, the values of a and b are determined from the best linear fit of equation (9). Final a- and b-values are then obtained by taking simple algebraic averages of a and b corresponding to each set. The values of a, b and ρ(300 K) for each sample are presented in table 1.

For all the samples we observed a single sharp peak in the $d\rho/dT$ versus T curve. Representative plots of $d\rho/dT$ as a function of temperature for samples A2 and B1 are shown in figure 2. A similar sharp peak in the $d\rho/dT$ versus T plot has also been observed in the 110 K phase Bi system by Ravi and Seshu Bai [26] and in the Tl-2:2:1:2 system by Duan *et al* [21]. These workers identified the temperature corresponding to the sharp peak in the $d\rho/dT$ versus T curve with the mean-field transition temperature. Following this we have also identified the temperature T_c at which a single sharp peak is observed in the $d\rho/dT$ versus T curve with the mean-field transition temperature T_c^{mf} and restrict our analysis in the mean-field regime of the Gaussian fluctuation above T_c^{mf}. The T_c^{mf} obtained from the $d\rho/dT$ versus T curve is presented in table 1.

Figure 3(a) shows the variations in $\ln(\Delta\sigma/\sigma_0)$ with $\ln\varepsilon$ for samples A1, A3, B1 and B3 and figure 3(b) those for samples A4 and A5. To compare the nature of the temperature dependence of enhanced conductivity with theoretical predictions we fit the observed excess conductivity using the AL theory (equation (4)). During the fit, B, λ and T_c^{mf} are treated as free parameters. Figure 3 shows close agreement between the experimental points and the AL theory (solid lines) in a wide temperature range. The values of the fitted parameters together with the temperature region of the best fit are listed in table 2. The values of C presented in the table were estimated from the values of d (table 1) and the fitted parameters

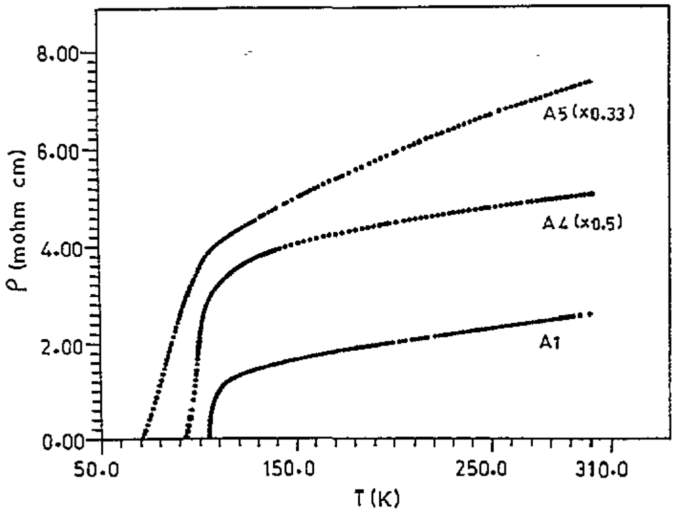

Figure 1. The variation in resistivity ρ with temperature T of $Tl_2Ba_2Ca_{1-x}Y_xCu_2O_{8+y}$ for $x = 0$ (A1), 0.20 (A4) and 0.25 (A5) samples.

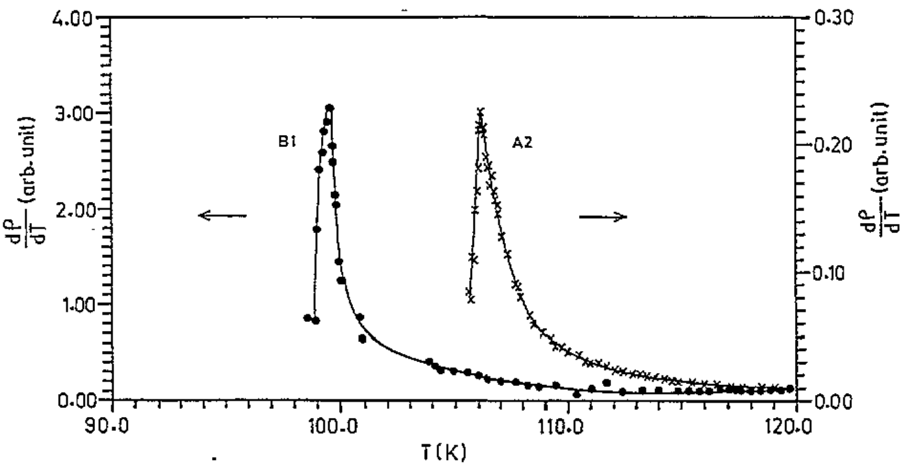

Figure 2. Plot of $d\rho/dT$ versus T for samples A2 ($x = 0$) and B1 ($x = 0$).

B. The table shows that the values of λ are close to 1.0, suggesting the two-dimensional nature of superconductivity in this system. It is also seen from figure 3 that the slope of the experimental $\ln(\Delta\sigma/\sigma_0)$ versus $\ln\varepsilon$ curve decreases as the temperature T approaches T_c^{mf}. Moreover, we have observed that, in the low-temperature region, the shape of the curve is sensitive to the choice of T_c^{mf}. In order to avoid any wrong choice for the estimation of T_c^{mf} we have also determined the critical exponent using the derivative technique (equation (10)). The exponent obtained from the log–log plot of the left-hand side of equation (10) and $\Delta\sigma$ is found to be in close agreement with that obtained from the AL fit. For samples A3 and A4, the variation in $\ln[(1/\rho^2)(d\rho/dT) - (1/\rho_n^2)(d\rho_n/dT)]$ with $\ln(\Delta\sigma)$ is shown in figure 4.

It has already been mentioned that for layered superconductors the LD theory is more

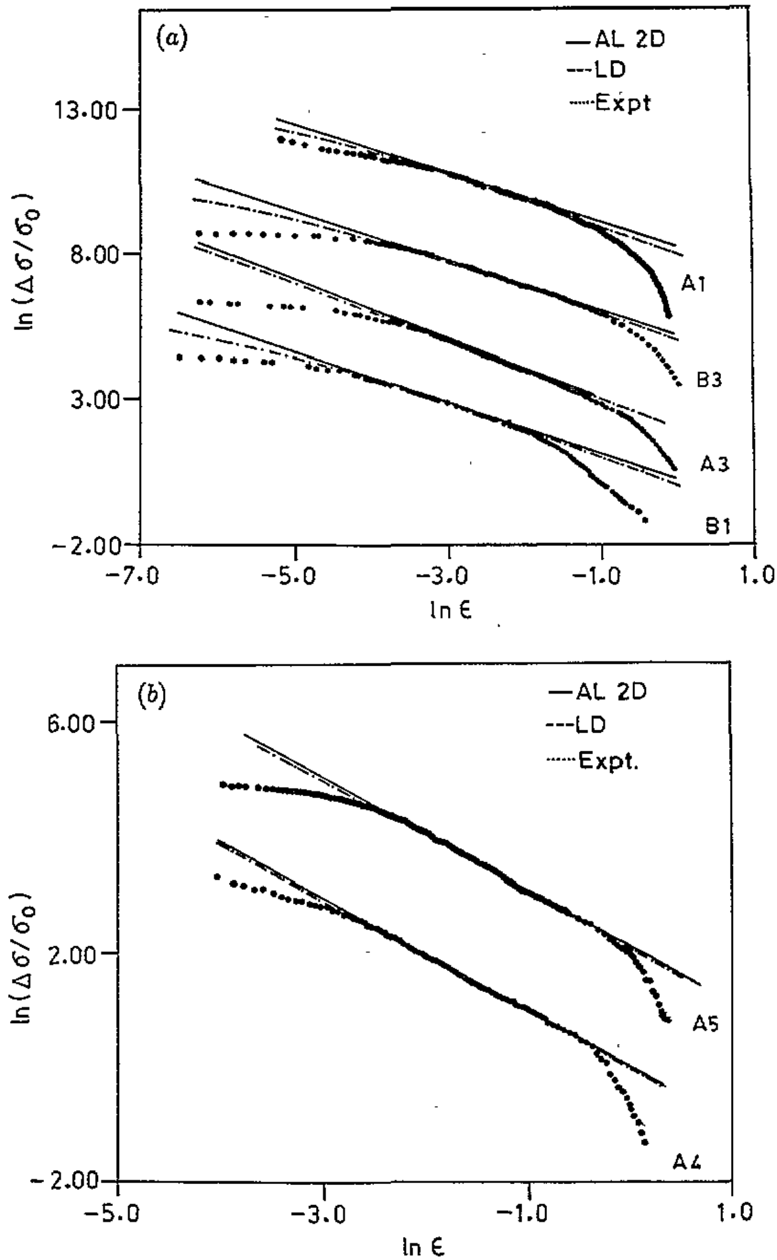

Figure 3. (*a*) Plot of $\ln(\Delta\sigma/\sigma_0)$ versus $\ln\varepsilon$ for $Tl_2Ba_2Ca_{1-x}Y_xCu_2O_{8+y}$ with $x = 0$ (A1), 0.1 (A3), 0 (B1) and 0.2 (B3) samples, σ_0 being the room-temperature conductivity. To accommodate all the curves in one figure we have added 8, 2 and 5 to the values of $\ln(\Delta\sigma/\sigma_0)$ for samples A1, A3 and B3, respectively. (*b*) The variation in $\ln(\Delta\sigma/\sigma_0)$ against $\ln\varepsilon$ for $Tl_2Ba_2Ca_{1-x}Y_xCu_2O_{8+y}$ for $x = 0.2$ (A4) and 0.25 (A5) samples. σ_0 is the room-temperature conductivity. We have added 2 to the value of $\ln(\Delta\sigma/\sigma_0)$ for sample A5 to include its data in the scale of sample A4.

appropriate for describing the nature of fluctuation than is the AL theory. For this reason

Table 2. AL-fit parameters and the temperature ranges of the fit.

Sample	Range of fit (K)	T_c^{mf} (K)	C	λ	χ^2
A1	109.0–137.0	104.70	4.92	0.88	2.35×10^{-3}
A2	107.2–119.0	106.05	7.55	0.86	2.91×10^{-2}
A3	107.0–138.0	103.50	13.10	1.05	2.32×10^{-4}
B3	106.0–142.0	102.97	8.22	0.89	1.45×10^{-3}
B1	101.5–116.5	99.99	10.94	0.86	2.72×10^{-3}
B2	97.1–106.3	95.60	18.56	0.96	3.33×10^{-3}
A4	98.0–139.2	96.00	25.20	0.97	1.68×10^{-2}
A5	92.5–147.1	84.99	43.10	1.01	2.063×10^{-2}

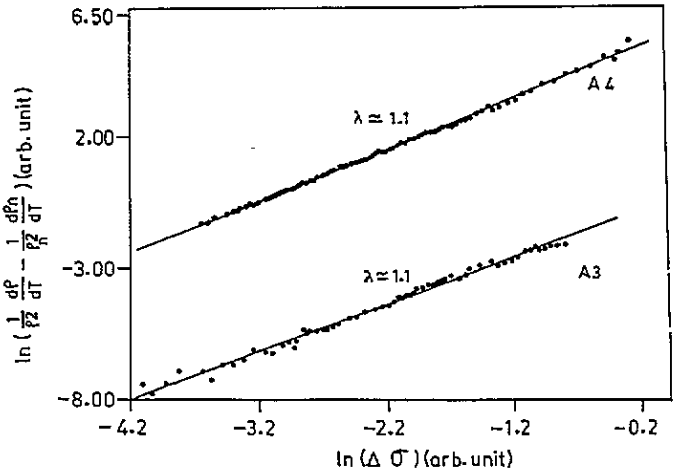

Figure 4. Plot of $\ln[(1/\rho^2)(d\rho/dT) - (1/\rho_n^2)d\rho_n/dT]$ against $\ln(\Delta\sigma)$ for samples A3 ($x - 0.1$) and A4 ($x = 0.2$).

we have also fitted the experimental excess conductivity with the LD theoretical predictions (equation (5)) taking $D(= e^2/16C\hbar d)$, J and T_c^{mf} as free parameters. The LD fit, shown by dashed lines in figure 3, is done over the same range of temperatures as in the case of the AL fit. Various fitted parameters together with the 2D-to-3D crossover temperature T_0 obtained from equation (3) are presented in table 3. It should be mentioned here that T_c^{mf} determined using method (iii) (table 1) and that obtained from method (ii) for AL and LD fits (tables 2 and 3) are close to each other. From table 3 it is seen that the sum of the squared deviation χ^2 for the LD fit is smaller than that for the AL fit (table 2). Thus, although good fits to the experimental data are obtained for both AL and LD models, the LD model provides a better fit than does the AL theory. In our earlier communication [27] we reported that the superconducting transition temperature of $Tl_2Ba_2Ca_{1-x}Y_xCu_2O_{8+y}$ (x = 0–0.6) decreases with a reduction in the carrier density, i.e. with increase in Y content x. This result, together with that in table 3, suggests that the interlayer coupling J decreases with reduction in the carrier concentration in the system.

From the excess-conductivity analysis of the $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_{8+y}$ (x = 0–0.35) system [8] we observed that both the interlayer coupling J and $\xi_c(0)$ decrease with the reduction in carrier concentration in the system. Moreover, in the low-hole-concentration region, T_c depends strongly on J compared with that in the optimally and heavily

Table 3. Parameters obtained from LD fit and 2D–3D crossover temperature T_0 estimated from equation (3).

Sample	T_c^{mf} (K)	C	$\xi(0)$ (Å)	$J \times 10^{-3}$	T_0 (K)	χ^2
A1	104.70	4.79	1.19	26.50	107.51	3.37×10^{-4}
A2	105.98	7.45	0.70	9.33	106.97	7.85×10^{-3}
A3	103.00	12.50	1.01	19.10	105.00	5.41×10^{-3}
B3	102.00	7.66	1.21	27.31	104.82	1.08×10^{-3}
B1	100.00	10.78	0.88	14.50	101.50	2.63×10^{-3}
B2	95.60	18.58	0.53	5.16	96.09	1.81×10^{-3}
A4	96.00	23.43	0.38	2.72	97.26	2.13×10^{-2}
A5	85.00	42.25	0.33	2.02	85.17	2.01×10^{-2}

doped regions. Following this result we expect a similar dependence of T_c on J in $Tl_2Ba_2Ca_{1-x}Y_xCu_2O_{8+y}$. For this we have plotted T_c^{mf} as a function of J (figure 5). The figure shows that in the under-doped region the superconducting transition temperature appears to decrease with increasing interlayer coupling J. To obtain a clear picture of the dependence of T_c on J in the under-doped region of Tl-2:2:1:2, one needs to analyse the excess conductivity of more samples with higher yttrium content x. Unfortunately, there is a limitation in the preparation of these samples. First, samples with a higher yttrium content are multiphase and, second, the superconducting transition width for these samples is so much broadened that it is not possible to obtain a consistent picture from the excess-conductivity analysis.

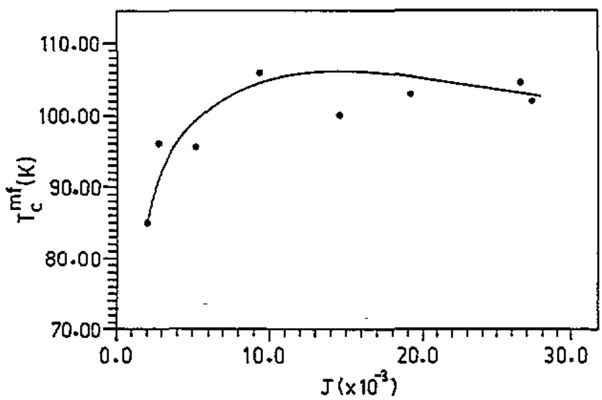

Figure 5. The dependence of T_c on the interlayer coupling J of $Tl_2Ba_2Ca_{1-x}Y_xCu_2O_{8+y}$ ($x = 0$–0.25).

From figure 3 it is seen that at high temperatures the experimental data lie below the predictions of AL and LD theory. This type of discrepancy between the theory and experiment has also been observed, [2, 3, 18] for $YBa_2Cu_3O_{7-\delta}$, $Bi_2Sr_2CaCu_2O_{8+y}$ and $Tl_2Ba_2CaCu_2O_{8+y}$ systems. A possible explanation of the above discrepancy is as follows. Fluctuations ψ can be treated within the framework of the Ginsberg and Landau (GL) theory [2] as long as they are small and vary slowly over the scale of the zero-temperature coherence length. At a temperature T, the spatial correlation of the fluctuation extends roughly over a distance equal to the coherence length $\xi(T)$. As T increases, $\xi(T)$ decreases, and consequently fluctuations vary rapidly on the scale of the size of a Cooper pair. As a result short-wavelength fluctuations make a significant contribution to the enhancement of conductivity. Thus, at high temperatures ($T > 2T_c$), it is necessary to introduce a high-momentum cut-off in order to remove the short-wavelength fluctuations for which the GL theory is not valid.

In the mean-field regime, we have also fitted the excess conductivity considering both the LD and the MT forms for the fluctuations (equation (7)). During the fit we have used the value of J obtained from the LD fit (table 3) and treated D, T_c^{mf} and δ as free parameters. Following Kim *et al* [25] and Ravi and Seshu Bai [26], we have also assumed δ to be temperature independent, i.e. $\tau_\phi \sim 1/T$. The solid lines in figure 6 represent the LD+MT (equation (7)) fit to the experimental excess conductivity for samples A1, B3 and A5. Using the value of J (table 3) and the fitted value of δ we have estimated the phase relaxation time τ_ϕ at 100 K from the expression for the pair-breaking parameter δ. The best-fit parameters together with the temperature region, the sum of the squared deviations χ^2 and the estimated values of τ_ϕ at 100 K obtained for samples A1, B3 and A5 are presented in table 4. It is observed (table 4) that the inclusion of the MT term fits our data over a larger temperature range than for the AL and LD fits (table 3). Moreover, the χ^2-value for the LD+MT fit is much smaller than those for the AL or for the LD fit. It shall be pointed out that the MT contribution to the fluctuation conductivity is more for sample A5 having a small interlayer coupling J than for the samples A1 and B3 with a large value of J. The values of τ_ϕ (table 4) for our polycrystalline $Tl_2Ba_2CaCu_2O_{8+y}$ samples are found to lie in the range $(1.3–1.8)\times10^{-15}$ s and this is smaller by a factor of 10 than that obtained by Kim *et al* ($\tau_\phi \simeq 3 \times 10^{-14}$ s). A similar discrepancy in τ_ϕ for sintered [28] ($\tau_\phi \leqslant 3 \times 10^{-14}$ s) and c-axis-oriented thin films [7] ($\tau_\phi \simeq 1 \times 10^{-13}$ s) of $YBa_2Cu_3O_{7-x}$ has also been observed by Matsuda *et al*. They also pointed out that the effect of a preferred orientation of the c axis is crucial for the determination of τ_ϕ.

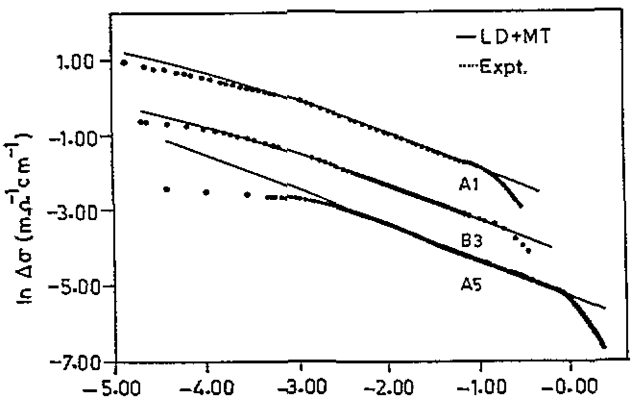

Figure 6. Variation in $\ln(\Delta\sigma)$ versus $\ln\varepsilon$ for $Tl_2Ba_2Ca_{1-x}Y_xCu_2O_{8+y}$ for $x = 0$ (A1), 0.2 (B3) and 0.25 (A5) samples. We have added 1 to the value of $\ln(\Delta\sigma)$ for sample A1 for clarity of the curves.

So far, measurements on paraconductivity [2, 3, 11] have only been analysed using the AL and LD models which are appropriate to an s-wave superconductor and the possible implication of non-s-wave pairing has not been investigated. Yip [29] studied theoretically

Table 4. The list of parameters obtained from the fit to the LD+MT expression (equation (5)) and corresponding values of τ_ϕ at 100 K for samples A1, B3 and A5.

Sample	Range of fit (K)	T_c^{mf} (K)	C	τ_ϕ (100 K) (s)	χ^2
A1	110.14–146.44	104.70	5.02	1.6×10^{-15}	8.81×10^{-4}
B3	106.15–152.00	102.94	7.24	1.8×10^{-15}	3.60×10^{-4}
A5	94.90–165.00	84.99	26.70	1.3×10^{-15}	3.12×10^{-4}

the effects of impurities on paraconductivity in an unconventional superconductor and pointed out that the AL contribution would be essentially unchanged while the MT contribution would not be present. Thus, if the comments of Yip are correct and the MT term is present only in s-wave superconductors [30], then this is a crucial issue as many theorists argue for an unconventional pairing mechanism [31, 32] in high-T_c systems.

We have presented the results for the excess conductivity of $Tl_2Ba_2Ca_{1-x}Y_xCu_2O_8$ ($x = 0$–0.25) and fitted these data to the existing theories of the AL, LD and MT contributions to the fluctuation conductivity. In the under-doped region we have observed a correlation between T_c and the interlayer coupling strength J obtained from the LD fit. The addition of the MT term to the LD model fits the data over a larger temperature range. The estimated phase relaxation time $(1.6$–$3.0)\times10^{-15}$ s at 100 K suggests strong pair-breaking effects in our samples. We observed that the MT contribution increases with decrease in J. The presence of the MT term indicates that the pairing mechanism in $Tl_2Ba_2CaCu_2O_8$ is probably conventional s wave type.

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References

[1] Iye Y 1989 *Studies of High Temperature Superconductors* vol 2, ed A Narlikar (New York: Nova Science) p 199
Batlogg B 1991 *Phys. Today* June p 44
[2] Frietas P P, Tsuei C C and Plaskett T S 1987 *Phys. Rev.* B **36** 833
Friedmann T A, Rice J P, Gianpintzakis J and Ginsberg D M 1988 *Phys. Rev.* B **39** 4258
[3] Kim D H, Goldman A M, Kang J H, Gray K E and Kampwirth R T 1989 *Phys. Rev.* B **39** 12 275
Poddar A, Mandal P, Das A N, Ghosh B and Choudhury P 1989 *Physica* C **159** 231
[4] Lee W C, Kleem R A and Johnston D C 1989 *Phys. Rev. Lett.* **63** 1012
Mandal P, Poddar A, Das A N, Ghosh B and Choudhury P 1990 *Physica* C **169** 43
[5] Inderhees S E, Salamon M B, Rice J P and Ginsberg D M 1991 *Phys. Rev. Lett.* **66** 232
[6] Iye Y, Nakamura S and Tamegai T 1989 *Physica* C **159** 616
Rice J P, Giapintzakis J, Ginsberg D M and Morchel J M 1991 *Phys. Rev.* B **44** 10 158
[7] Matsuda Y, Hirai T, Komiyama S, Terashima T, Bando Y, Iijima K, Yamamoto K and Hirata K 1989 *Phys. Rev.* B **40** 5176
[8] Mandal P, Poddar A and Das S 1994 *J. Phys.: Condens. Matter* **6** 5689
[9] Aslamazov L G and Larkin A I 1968 *Phys. Lett.* **26A** 238
[10] Lawrence W E and Doniach S 1971 *Proc. 12th Int. Conf. on Low Temperature Physics (Kyoto, 1970)* ed E Kanada (Tokyo: Keigaku) p 361

[11] Oh B, Char K, Kent A D, Naito M, Beasley M R, Geballe T H, Hammond R H and Kaptulnik A 1989 *Phys. Rev.* B 37 7861
[12] Poddar A, Mandal P, Das A N, Ghosh B and Choudhury P 1989 *Physica* C **159** 231
[13] Ausloos M and Laurent Ch 1988 *Phys. Rev.* B **37** 611
[14] Maki K 1968 *Prog. Theor. Phys.* **39** 897
[15] Thompson R S 1970 *Phys. Rev.* B **1** 327
[16] Hikami S and Larkin A I 1988 *Mod. Phys. Lett.* B **2** 693
[17] Maki K and Thompson R S 1989 *Phys. Rev.* B **39** 2767
[18] Poddar A, Mandal P, Das A N, Ghosh B and Choudhury P 1989 *Physica* C **161** 567
[19] Ravi S and Seshu Bai V 1992 *Solid State Commun.* **83** 117
[20] Testardi L R, Reed W A, Hohenberg P C, Hammerle W H and Breenert G F 1969 *Phys. Rev.* **181** 800
[21] Duan H M, Kichl W, Dong C, Cordes A W, Saced M J, Viar D L and Hermann A M 1991 *Phys. Rev.* B **43** 12925
[22] Ausloos M, Gillet F, Laurent Ch and Clippe P 1991 *Z. Phys.* B **84** 13
[23] Overend N and Howson H A 1992 *J. Phys.: Condens. Matter* **4** 9615
[24] Winzer K and Kumm G 1991 *Z. Phys.* B **82** 317
[25] Kim D H, Gray K E, Kampwirth R T and McKay D M 1991 *Phys. Rev.* B **43** 2910
[26] Ravi S and Seshu Bai V 1992 *Mod. Phys. Lett.* B **6** 1857
[27] Poddar A, Mandal P, Das A N, Ghosh B and Choudhury P 1991 *Phys. Rev.* B **44** 2757
[28] Matsuda Y, Hirai T and Komiyama S 1988 *Solid State Commun.* **68** 103
[29] Yip S-K 1990 *Phys. Rev.* B **41** 2612
[30] Balian R and Werthamer N R 1963 *Phys. Rev.* **131** 1553
[31] Monien H, Pines D and Takigawa M 1991 *Phys. Rev.* B **43** 258
Monthoux P and Pines D 1992 *Phys. Rev.* B 47 6069
[32] Levi B G 1993 *Phys. Today* May 17, and references therein