Magnetoresistance Studies of Tl-1223 Phase Substituted by Scandium

R. Awad¹, I. H. Ibrahim¹², E. M. E. Mansour³, M. Roumie⁴ and A. Zein²

¹Physics Department, Faculty of Science, Alexandria University, Alexandria, Egypt.
²Physics Department, Faculty of Science, Beirut Arab University, Beirut, Lebanon.
³Chemistry Department, Faculty of Science, Beirut Arab University, Beirut, Lebanon.
⁴Accelerator Laboratory, National Council for Scientific Research, Beirut, Lebanon.

E mail: alialzein@gmail.com

Abstract
Superconducting samples of type TlBa$_2$Ca$_{2-x}$Sc$_x$Cu$_3$O$_{9-\delta}$ with $x$ = 0.0, 0.025, 0.05, 0.1, and 0.2 have been prepared via solid-state reaction technique. The effect of weak magnetic fields up to 4.4 kG on the electrical resistivity of the prepared samples has been studied to investigate the flux motion for this phase. The results reveal a slight shift in the superconducting transition temperature $T_c$ and an increase in the superconducting transition width $\Delta T$ with increasing magnetic fields. The experimental data fit well with the thermally activated flux creep model and the activation energy $U(B)$ shows a power law dependence on magnetic field as $B^{-\beta}$. Also, the transition width is related to the magnetic field according to the relation $\Delta T \propto B^n$. The values of $\beta$ and $n$ are strongly dependent on the Scandium-content. The temperature and magnetic field dependence of the activation energy $U(B, T)$ is found to be $U(B, T) \sim \Delta T B^{-\eta}$, where $\eta = \beta + n$. The critical current density $J_c(0)$ and the upper critical field $B_{c2}(0)$ are calculated, from the above measurements, as a function of Scandium-content. Finally the electronic thermal conductivity $\kappa_e$, estimated from Wiedemann-Franz law, is reported at different applied magnetic fields for the prepared samples.

Keywords: Scandium- activation energy- flux creep- Upper critical magnetic field.

1- Introduction
Polycrystalline high-temperature superconductors, HTSC's, exhibit large magnetoresistance in weak magnetic fields (tens of Oersteds) [1–2]. This effect can be used in devices that demand a sensitive electrical response for a weak magnetic field. In this sense, polycrystalline bulk HTSC's is more attractive materials than conventional magnetic sensors that utilize the Hall effect or the magnetoresistive (MR) effect in semiconductors [1]. The resistive transition of all HTSC's broadens in applied magnetic fields [3]. This broadening exists in the limit of zero current and is more pronounced in more highly anisotropic materials. The resistivity transition of HTSC's is characterized by two stages of transition [4]. At higher temperatures near the superconducting transition temperature $T_c$, superconductivity stabilizes in homogonous macroscopic regions of the sample and the magnetic field has a little effect on $T_c$. At lower temperature, closer to zero resistivity temperature, a long-range superconducting state with zero resistivity is achieved by means of percolation like process that controls the activation of weak link between grains. As the magnetic field increases, the weak links are affected and therefore the tail in resistivity measurements appears. There are some different models for interpretation of resistivity broadening under magnetic field such as thermally activated flux creep, phase slip, flux line melting and flux cutting ( curved flux lines) [5].

This work presents the effect of weak magnetic fields up to 4.4 kG on the electrical resistivity measurements of TlBa$_2$Ca$_{2-x}$Sc$_x$Cu$_3$O$_{9-\delta}$ substituted by scandium. The flux pinning activation energy and critical current density are estimated from the above measurements as a function of Sc-content.
2- Experimental technique:

Samples with the nominal composition of TlBa$_2$Ca$_{2-x}$Sc$_x$Cu$_3$O$_{9-\delta}$ ($0 \leq x \leq 0.2$) were synthesized by the conventional single step of solid-state reaction technique and characterized by means of X-ray powder diffraction, scanning electron microscope and energy dispersive X-ray [6].

The electrical resistivity of the samples was measured using a conventional four–probe technique in the temperature range $18 \leq T \leq 300$ K with a closed- cryogenic system. The samples used for resistivity measurements had dimensions of about $1.5\times0.2\times0.3$ cm$^3$ and the connection of the copper leads with the samples was made using a silver paint. The temperature of the samples is monitored by a Kp-Au 0.07 at. % Fe thermocouple and stabilized with the aid of a temperature controller to within $\pm 0.1$ K. The resistivity versus temperature measurements were performed at different applied dc magnetic fields ($0.29, 0.5, 1.2, 2.34, 3.53$ and $4.4$ kG) at a constant driving current of 1 mA. The magnetic field was applied normal to the direction of the driving current and was generated with an electromagnet.

3- Results and discussion

![Graph of electrical resistivity vs. temperature](attachment:figure1.png)

Figure 1 The variation of the electrical resistivity with temperature at different applied magnetic field values for: (a) TlBa$_2$Ca$_2$Cu$_3$O$_{9-\delta}$ and (b) TlBa$_2$Ca$_{1.8}$Sc$_{0.2}$Cu$_3$O$_{9-\delta}$.
Figures 1a and 1b show the temperature dependence of the electrical resistivity measurements under various applied magnetic fields \( B \) for \( \text{TlBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{9-\delta} \) and \( \text{TlBa}_2\text{Ca}_{1.8}\text{Sc}_{0.2}\text{Cu}_3\text{O}_{9-\delta} \), respectively. A slight shift in the superconducting transition temperature \( T_c \) is observed as the applied magnetic field increases. These results are just a consequence of the fact that being very close to \( T_c \), the change between the normal and the superconducting resistivity is very small. This means that only a small fraction of the carriers are superconducting [7]. Also, this behavior could be attributed to the strong intragrain pinning energy, which does not allow any vortex motion near \( T_c \) [8]. As the temperature is lowered, the electrical resistivity is more sensitive to the applied magnetic field and the resistivity tail appeared, indicating that at lower temperatures more carriers are superconducting so the change between the zero field and the applied field resistivity is greater [9] or associated to the weak links between the grains. It is clear, from the figures that, the zero resistivity temperature \( T_0 \) is affected by the applied magnetic fields, showing a shift to considerably lower temperature as the applied magnetic fields increase.

**Figure 2** The variation of the activation energy \( U(B) \) with the applied magnetic fields for \( \text{TlBa}_2\text{Ca}_{2-x}\text{Sc}_x\text{Cu}_3\text{O}_{9-\delta} \), \( x = 0.0, 0.05, 0.1 \) and \( 0.2 \).

**Figure 3** The variation of transition width (\( \Delta T = T_c - T_0 \)) as a function of the applied magnetic field for \( \text{TlBa}_2\text{Ca}_{2-x}\text{Sc}_x\text{Cu}_3\text{O}_{9-\delta} \), \( x = 0.0, 0.025, 0.05 \) and \( 0.1 \).
In the low current limit, the resistivity behavior in the tail part can be expressed by the following exponent relation [10]:

$$\rho(T, B) = \rho_0 \exp \left( \frac{-U}{k_B T} \right)$$

(1)

where $k_B$ is the Boltzmann constant, $U$ is the flux pinning activation energy and $\rho_0$ is a prefactor independent on the applied magnetic fields. The Arrehineus plot between $\ln(\rho)$ versus $(1/T)$, for all prepared samples, at various applied magnetic fields shows nonlinear relation over all range of temperature. This result indicates that $U$ depends not only on the applied magnetic field but also on the temperature. The magnetic field and temperature dependence of $U$ can be represented by the following relations [5]:

$$\frac{\rho}{\rho_p} = \left[ I_0 \left\{ \frac{\gamma(B,T)}{2} \right\} \right]^{-2}$$

(2)

where $I_0$ is the modified Bessel function and $\rho_p$ is the resistivity at temperature $T_c$ and $\gamma$ is the barrier height $[\gamma(B,T) = U(B,T) / k_B T]$ [11], from this $U(B,T)$ can be written in the form:

$$U(B, T) = a B^{-\beta} (1 - \frac{T}{T_c})^q$$

(3)

where $a$, $\beta$ and $q$ are constants and their values will be determined as a function of Sc-content later. To eliminate the temperature of $U(B)$, we calculate it from the slope of the linear part of the relation between $\ln(\rho)$ versus $1/T$. The variation of the flux pinning activation energy with the applied magnetic fields for TlBa$_2$Ca$_{2-x}$Sc$_x$Cu$_3$O$_{9-\delta}$, $x = 0.0, 0.05, 0.1$ and $0.2$ is shown in figure 2. $U(B)$ drops rapidly as the applied magnetic fields increase up to $1.1$ kG and then it decreases slowly (nearly plateau) as the applied magnetic fields reach $4.4$ kG and the data are fitted as a power law relation; $U(B) \sim B^{-\beta}$. This behavior is quite similar to that observed in all high-temperature superconductors [12]. It is clear that $U(B)$ increases as $x$ increases from $0$ to $0.05$ and then it decreases. The dependence of $U(B)$ on substitution-content is not easy to discuss due to the dependence of $U$ on $T_c$ which differs with Sc-content. Similar behavior was found by Paulius et al. [13] in YPr-123 system. They have attributed this increase to the local suppression of the superconducting order parameter in the vicinity of Pr ions. On the other side they related the reduction of $U(B)$ with substitution-content to three factors which are the decreasing of $T_c$, decreasing in the thermodynamic critical field $H_c$ [14] and the contraction of distance between the pinning centers.

The temperature dependence of $U$ can be determined from the magnetic field dependence of transition width $\Delta T$. The variation of transition width ($\Delta T= T_c - T_0$) as a function of the applied magnetic field is shown in figure 3. The transition width data are well fitted according to a power law scaling relation as $\Delta T \propto B^n$.

Using the above equations and many mathematical treatments, discussed by Mohammed et al. [15], we can find the dependence of $U$ on both $T$ and $B$:

$$U(B,T) \sim \Delta T B^{-\eta}; \quad \eta = \beta + n.$$  

(4)

The calculated values of $\beta$, $n$, $\eta$ and $q$ are listed in table 1 as a function of Sc-content.

| Sc-content | $\beta$ | $n$ | $\eta$ | $q$ |
|------------|---------|-----|--------|-----|
| 0          | 0.653   | 0.195 | 0.8489 | 3.3399 |
| 0.025      | 0.544   | 0.178 | 0.7229 | 3.0612 |
| 0.05       | 0.378   | 0.190 | 0.5691 | 1.9827 |
| 0.1        | 0.463   | 0.193 | 0.6561 | 2.3977 |
| 0.2        | 0.694   | 0.219 | 0.9132 | 3.1660 |

Table 1: Variation of $\beta$, $n$, $\eta$ and $q$ with Sc-content $x$ for TlBa$_2$Ca$_{2-x}$Sc$_x$Cu$_3$O$_{9-\delta}$
It is clear that the values of $\beta$, $n$ and $\eta$ decrease with increasing the Sc-content up to $x = 0.05$ and then they increase. It is well known that the value of $\beta$ is strongly dependent on the orientation of magnetic field with respect to the basal plane and the range of applied magnetic field [16-17]. The values of $q$, determined using equation 4, are in the range of 1.9 – 3.6 for our samples. In some other works the value of $q$ have been derived to be 3/2 [16], 2 [17] and 1.53-2.8 [18]. Also, we notice that $q$ increases with the increase of Sc-content, confirming with the results of Shakeripouer et al. [19] for Gd-123 doped by Ca and Pr.

The magnetic field dependent parameter $C(B)$ is defined to be equal to $A(B)/B$, where $A$ is a constant that depends on the magnetic field and $B$ is the applied magnetic field, is calculated from the barrier height $\gamma(B,T)$ [11]. This parameter is proportional to the activation energy and related to the critical current density at zero temperature $J_c(0)$ by the relation [20]:

$$C(B) = \frac{J_c(0)h d^2}{ek_B T_c},$$

where $d$ is the average grain- size, which determined from scanning electron microscope (1-4$\mu$m). The calculated values of $J_c(0)$ for TlBa$_2$Ca$_{2-x}$Sc$_x$Cu$_3$O$_{9-\delta}$ with $x = 0.0$, 0.05 , 0.1 and 0.2 is plotted as a function of the applied magnetic field in figure 4. As the applied magnetic field increases, the critical current density decreases as a power law, $J_c(0) \approx B^m$. Also, it is clear that the critical current density enhances as Sc-content increases till $x$ around 0.05 and then it decreases. The enhancement of $J_c(0)$, for $0 \leq x \leq 0.05$, is probably due to the lattice defects produced from the partial substitution of Ca$^{2+}$ ion by Sc$^{3+}$ ion that enhances the flux pinning. Whereas the suppression of $J_c(0)$, for $x > 0.05$, is attributed to the increase of porosity and non superconducting phase as Sc-content increases.

![Figure 4 The values of $J_c(0)$ for TlBa$_2$Ca$_{2-x}$Sc$_x$Cu$_3$O$_{9-\delta}$ with $x = 0.0$, 0.05 , 0.1 and 0.2 as a function of applied magnetic field.](image)

The variations of electronic thermal conductivity, determined from Wiedemann-Franz law, with the temperature at different applied magnetic fields for TlBa$_2$Ca$_{2-x}$Sc$_x$Cu$_3$O$_{9-\delta}$ with $x = 0$ and 0.2 are shown in figures 5 and 6, respectively. It is clear that the electronic thermal conductivity decreases rapidly up to $T=115$ K and then it tends to plateau as the temperature increases as shown in the inset of figures 5 and 6. This behavior is consistent with the previous results of thermal conductivity of the high-temperature superconductors [21]. This sharp increase in $\kappa_e$ may be due to the rounding of the electrical resistivity that we observe above $T_c$ in high-temperature superconductors. This rounding was explained according to thermodynamics fluctuations of the superconducting order parameter and there are two processes that contribute to the thermodynamics fluctuations. The first process is due to the
acceleration of superconducting pairs created in the thermal nonequilibrium with finite life time [22]. The second process results from the scattering of normal-state quasi particles with the superconducting pairs [23]. Also, the electronic thermal conductivity, at certain temperature below 115 K, decreases as the magnetic field increases, whereas for T >115 K, the magnetic field doesn’t affect the thermal conductivity. This decrement in $\kappa_e(T)$ is probably due to the presence of vortices that acts as additional scattering centers and electronic expiations, which are more still plentiful [24].

Figure 5 The variation of $\kappa_e$ with the temperature at different applied magnetic fields for TlBa$_2$Ca$_{2-x}$Sc$_x$Cu$_3$O$_{9-\delta}$ with x = 0. (The linear part in the temperature range 127-149.5 K is shown in the inset).

Figure 6 The variation of $\kappa_e$ with the temperature at different applied magnetic fields for TlBa$_2$Ca$_{2-x}$Sc$_x$Cu$_3$O$_{9-\delta}$ with x = 0.2. (The linear part in the temperature range 132-152 K is shown in the inset).

4- Conclusion
Magnetoresistance studies of TlBa$_2$Ca$_{2-x}$Sc$_x$Cu$_3$O$_{9-\delta}$ system with x = 0.0, 0.025, 0.05, 0.1, and 0.2 showed that the magnetic field has a little effect in the first stage of transition whereas it enlarged
the transition width in the second stage of transition. This enlargement was related to the applied magnetic field according to the formula \( \Delta T \sim B^{n} \). The flux pinning energy \( U(B) \), determined from Arrhenius relation, increased until \( x = 0.05 \), beyond which it decreased. Magnetic field and temperature dependence of the flux pinning energy have been obtained as \( U \sim B^\beta T (1 - T/T_c) ^{\eta} \). The temperature and magnetic field dependence of the flux pinning energy can be represented as \( U \sim \Delta T B^{-\eta} \), \( \eta = \beta + n \). The critical current density enhanced until \( x = 0.05 \), indicating that the lower content of Sc acted as a source of flux pinning. The reduction in the critical current density for \( x > 0.05 \) was attributed to the increase of secondary phases. The calculated electronic thermal conductivity showed a temperature independent behavior above the superconducting transition temperature and it increased sharply at temperatures around \( T_c \). The applied magnetic field depressed the electronic thermal conductivity below \( T_c \) and didn’t affect it above \( T_c \).

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