A New Method For Determining The Lower and Upper Critical Magnetic Field in Tl-2234 Superconductor

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ABSTRACT
The purpose of this paper is to discuss and calculation the critical fields of type-II superconductors. In order to develop a physical basis for the concept of the critical field, particularly the intermediate state. The Tl$_3$Ba$_2$Ca$_x$Cu$_{2+y}$O$_{6-δ}$ (Tl-2234) Superconducting sample was prepared under normal pressure by a one step of solid-state reaction technique. The offset transition temperature of the Tl-2234 sample without applied magnetic field is 105 K, whereas, the offset transition temperature drops to 38.97 K with 12 T applied magnetic field. By using the resistance measurements as function of temperature at high different magnetic fields, we investigated a new method for calculation of the critical magnetic field. The upper critical field $B_c2$ and the lower critical field $B_c1$ and the critical magnetic field $B_c$ of anisotropic magneticsuperconductors are calculated. In addition, the Ginzburg-Landau parameter $(κ)$ and the coherence length $(ξ(0))$ were estimated from $B_c$. The results of our calculations agree in agreement with experimental data for (Tl-2234) Superconductor and the experimental values well with theoretical findings.

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INTRODUCTION

Much attention has been focused on the lower- and upper-critical fields in type-II superconductors under magnetic fields. It is important quantity since they give the most direct information about the microscopic parameters like the superconducting coherence length and its anisotropy within the superconducting state. So far, $B_c$ of Tl$_3$Ba$_2$Ca$_x$Cu$_{2+y}$O$_{6-δ}$ and other high-T$_c$ materials has been determined mainly from resistance transition curve (Thopart et al., 2000; Celik et al., 2008). One characteristic of a superconductor that it has “zero resistance”, i.e., a superconductor will lose its electrical resistance and carry current without heat loss if it is cooled to a certain temperature, critical temperature $T_c$. The current flowing in the zero resistance superconductor is called “supercurrent”. The other characteristic of a superconductor is that on cooling below $T_c$, it excludes an external magnetic field from penetrating into its interior, which is known as the “Meissner Effect”. But the magnetic field will penetrate the sample at some high field. In fact, any applied magnetic field can enter a finite distance, the penetration depth $λ_0$ into the surface layer of a superconductor. All superconductors can be divided into two classes, Type I and Type II, based on the ratio of $λ$ to another fundamental superconductivity parameter, the coherence length $ξ(0)$ which is the interaction distance between paired electrons. This ratio, $κ=λ/ξ(0)$, is called the Ginzburg-Landau parameter. If $κ<1/√2$, the superconductor is Type I. If $κ>1/√2$, the superconductor is Type II. The most fundamental characteristic distinction/definition for Type I and Type II superconductors is the sign of the interface energy between the normal and superconducting domains. Type I has a positive interface energy, and Type II has negative interface energy. Type I superconductors do not let the magnetic flux penetrate into its interior until the magnetic field reaches a critical field $H_c$. Above $H_c$, the magnetic field penetrates the entire sample, the Type I superconductor becomes a normal state conductor. Pure element superconductors mainly belong to the Type I. The negative normal/superconducting interface energy allows the Type II superconductor to occupy as much interfacial area as possible. When a magnetic field $H$ is applied, the Type II superconductor is in the Meissner state and no magnetic flux from entering its interior until $H$ reaches a lower critical magnetic field $H_{c1}$. Above $H_{c1}$, the magnetic flux has penetrated into the Type II SC and the material is in a mixed normal and superconducting
state between $H_1$ and $H_2$. With increasing field above $H_1$, the superconductivity decreases until at another field $H_2$. Above $H_2$, the Type II superconductor becomes normal state. The penetrated magnetic flux consists of discrete quanta called fluxons. Each fluxon has a value of $2.1 \times 10^{-15}$ Wb and is composed of normal state core with a radius of $\xi$ and a vortex of supercurrent with a radius of $\lambda$ (Fossheim and Sudbø, 2005; Poole Jr et al., 2007).

Type-II superconductor materials exhibit different magnetic properties from type-I superconductors. When such a superconductor, type-II, is in a magnetic field, the free energy can be lowered by causing domains of normal material containing trapped flux to form with low-energy boundaries created between the normal core and the surrounding superconducting materials. When the applied magnetic field exceeds a value referred to as the lower critical field, $B_{c1}$, magnetic flux is able to penetrate in the quantized units by forming cylindrically symmetric domains called vortices. For applied fields slightly above $B_{c1}$, the magnetic field inside a type-II superconductor is strong in the normal cores of the vortices, decreases with distance from the cores, and becomes very small faraway. For much higher applied magnetic fields the vortices overlap and the field inside the superconductor becomes stronger everywhere (Li et al., 2006; Nakai et al., 2009). Eventually, when the applied field reaches a value called the upper critical fields $B_{c2}$, the material becomes normal. Type-II superconductors also have zero resistance, but their perfect diamagnetism occurs only below the lower critical field $B_{c1}$. The superconductor used in partial applications, which have relatively high transition temperature, carry large currents and often operate in large magnetic fields, are all of type-II.

The superconducting transition does not occur at a fixed temperature but at a temperature ranging between the onset critical temperature $T_{c1,\text{onset}}$ and the zero resistance temperature $T_{c1,\text{offset}}$. The width of this range $\Delta T = T_{c1,\text{onset}} - T_{c1,\text{offset}}$ is called the transition width and it is affected by the driving current and applied magnetic field (Abou-Aly et al., 2000; Khosroabadi et al., 2003; Yıldırım et al., 2013). This transition width may reaches a value ranging between 40 K and 50 K which is much larger than in the case of low $T_c$ superconductors (Lee et al., 1989). This is a general problem in all high temperature superconductors, where the resistance decreases rapidly about an order of magnitude for a few Kelvin and then the resistance decreases smoothly to zero as the temperature reaches to zero resistance temperature (resistance noise). The amplitude of the resistance noise in all ceramic superconductors is very different when the same resistance value is obtained either by increasing or by decreasing the applied magnetic field. The effect of magnetic field on the electrical resistance measurements is a rich source of information about the upper critical field (Schilbe et al., 2003), activation energy (Awad et al., 2001) and the vortex dynamics in typeII superconductors (Ogale et al., 1995). Usually, above the transition temperature $T_c$ (ordinary normal state), both small and moderate applied magnetic fields do not affect the electrical resistance data. But below the transition (superconducting state), the presence of magnetic field shifts the transition temperature downward and broadens the transition width. Resistance versus temperature measurements of the samples was performed for temperature range of 10-130K under various applied magnetic field ranging from 0 T to 12 T by Cryocooler and superconducting magnet from CRYOGENIC Industries.

The rest of this paper is organized as follows. In the first section, we concentrate on the transition width ($\Delta T$) dependence of (B) and obtain expressions valid in the vicinity of the critical temperature $T_c$. In Section 4, we derive the expressions for the upper and lower critical magnetic field and outline the Ginzburg–Landau theory. Our results for Tl-2234 sample is presented in Section 4 and discussed in the light of available experimental data.

The aim of this work is the study of the effect of applied magnetic field (up to 12 T) on the transition behavior in Tl-2234 superconductor, which would allow describing the critical properties of individual superconductors.

**Experimental Technique:**

Samples with the nominal composition of Tl$_2$Ba$_2$Ca$_2$Cu$_2$O$_{6+\delta}$ were synthesized by the conventional single step of solid-state reaction technique. Stoichiometric amounts of highly pure Tl$_2$O$_3$, BaO$_2$, CaO, and CuO were mixed using an agate mortar to make fine powder which was sieved in 64μmsieve to obtain a homogeneous mixture. The powder was pressed into the form of a disc of dimensions 1.5 cm diameter and 0.2 cm thickness. Then the samples were wrapped in a silver foil, in orderto reduce thallium evaporation during the preparation. In order to protect the furnace from possible hazardous effects, the sealed quartz tube of diameter 1.5 cm. And offlength 15 cm was put in a closed, stainless steel protecting tube. Finally, the sealed tube was placed horizontally in a furnace and heated at a rate of 4°C/min to 820°C, and held at this temperature for four hours, then werecooled to room temperature with a rate of 0.5°C/min. The electrical resistivity of the prepared samples was measuredby the conventional four-probe technique from room temperatureredown to zero resistivity temperature via a closed cryogenicrefrigeration system employing helium gas as a workingmedium. The samples have the shape of parallelepipeds of approximate dimensions15x 2 x 3 mm$^3$ and the connections of the copper leads with the sample were made using a conductivesilver paint, and a constant current of 2mA is passed through the sample during resistivity measurements in four probe method, provided from a Keithley2400 current source, was used to avoid heating effects on the samples. We use Keithley 2400 SourceMeter as the DC bias.
source because it can be operated either in voltage or current source mode. In the whole experiments, the stable magnetic field up to 12 T was applied by LakeShore Superconducting Magnet System and the temperature of the sample was controlled precisely within 1 mK. During the cooling down process, both the Keithley SourceMeter and Lakeshore temperature controller are controlled by the LabVIEW software, It is used to display, record, and store the data.

RESULTS AND DISCUSSION

The temperature dependence of the zero-field resistivity of the sample, measured in dc mode using a 2 mA bias current, is shown in Fig. 1. The resistance dependence on temperature for the sample has two stages of transition in the superconducting state. The first stage (high temperature region) starts at the onset temperature \( T_{\text{c onset}} = 120 \, \text{K} \) and shows a rapid decrease in the resistance, about an order magnitude for a few Kelvin. In the second stage (low temperature region) the resistance smoothly decreases to zero as the temperature approaches \( T_{\text{c}} \). This is believed to be due to the very short coherence length \( \xi \approx 10^{-7} \, \text{Å} \) and the granular structure of the high temperature superconductors (Tinkham, 1988). Our first observation is that B has no effect on the first stage of the transition, because, as one can see in Fig. 1., the curves obtained for resistance at different values of magnetic field \( B = 0, 0.25, 0.5, 1, 2, 4, 6, 8 \) and 12 T overlap in the high temperature region. This could also contain some information concerning spatial distribution of “strong” superconducting grains and “weak” superconducting boundaries. In the second stage we observe, for the higher value of the magnetic field, that the transition width \( \Delta T \) is enlarged. by increasing the applied magnetic field (Kameli et al., 2008).

The resistance has been estimated considering the above reported dimensions for the sample. The sample is metallic down to 130 K where saturation in the resistance starts to appear. The critical temperature, defined as the temperature where the resistance R is less than \( 10^{-5} \, \Omega \) is equal to \( T_{\text{c onset}} = 105 \, \text{K} \) and the superconducting transition width defined as the difference between the temperatures measured at 90% and 10% of the normal state resistance, is \( \Delta T_{\text{c}} = 15 \, \text{K} \). Resistive transitions, normalized with respect to \( R_{\text{on}} \), the value of the resistance just before the superconducting transition, in external magnetic fields up to 12 T are shown in Fig. 2. The effect of the applied field is the broadening of the resistance transitions, suggesting the presence of dissipation phenomena similar to those observed in conventional HTS. The \( R-T \) behavior above \( T_{\text{c onset}} \) is nearly independent of the applied field. For intermediate values of the magnetic field the \( T_{\text{c onset}} \) value decreases from 105 K to around 38.97 K. Fig. 2. Shows the temperature dependence of the resistance of the sample for different magnetic fields up to 12 T. The resistance starts to drop at the temperature of 120 K and then vanishes below 105 K in zero magnetic fields. The resistance exhibits the linear behavior above \( T_{\text{c}} \), and the residual resistance ratio (RRR) is also obtained, where \( \text{RRR} = R(292K)/R(T_{\text{c}}) = 1.98 \), which means that the scattering becomes large at the onset temperature, and demonstrating the good quality of the present sample. The onset of the transition temperature, \( T_{\text{c}} \), decreases very slowly with increasing magnetic field. However, the \( T_{\text{c}} \) (R = 0) decreases significantly, and the \( \Delta T_{\text{c}} (T_{\text{c}} (90\%) - T_{\text{c}} (10\%)) \) broadens simultaneously.

The broadening of the resistance transition region in a magnetic field over a wide temperature region below \( T_{\text{c}} \) is one of the intriguing properties of high \( T_{\text{c}} \) superconductors and has recently become an experimentally well-established phenomena. The broadening is caused by energy dissipation related to the thermally activated flux creep (Triscone et al., 1990; Lee et al., 2005).

![Fig. 1: The temperature dependence of resistance for Tl2Ba2Ca4Cu4O10+δ at different applied magnetic fields.](image)

In the previous section, as we saw in the second stage, we observe for the higher value of the magnetic field, the transition region is enlarged. So, the transition width \( \Delta T \) as function of the magnetic field was plotted in Fig. 3 for the sample Tl2Ba2Ca4Cu4O10+δ.
Fig. 2: Normalized resistance versus temperature graph under various magnetic field under 130K for Tl₂Ba₂Ca₂Cu₄O₆+δ sample.

Fig. 3: The transition width $\Delta T$ versus the applied magnetic field for Tl₂Ba₂Ca₂Cu₄O₆+δ sample.

Some researchers (Tampieri et al., 1998; Khosroabadi et al., 2003) found that the difference between the intragranular ($T_{\text{cg}}$) and intergranular ($T_{\text{cj}}$) transition temperatures as a function of magnetic field has the same behavior as we found (Figs. 3). The experimental data in figure is well fitted to the exponential equation (empirical relation) in the form

$$\Delta T = \Delta T(0) \exp \left( \frac{B}{B_c} \right)^n$$

(1)

where $\Delta T(0)$ is the transition width at zero applied magnetic field, $n$ is a critical exponent of magnetic field and $B_c$, as we see later, is the geometrical mean of the critical magnetic fields. For our studied sample, we found that $n$ is closed to $(1/3)$. The inserted chart in figure (3) represent the linear fit of the equation (1), with the form

$$\ln \Delta T(B) = \ln \Delta T(0) + \left( \frac{B}{B_c} \right)^{\frac{1}{3}}$$

(2)

where the slope of these straight lines equal to $\left( \frac{1}{B_c} \right)^{\frac{1}{3}}$. The value of the critical magnetic field $B_c$ is evaluated to be 1.52 T. It is clearly, from the Fig. 3, which the transition width $\Delta T(B)$ is divided into two zones, the lower limit zone and the upper limit zone. The lower limit zone where the transition width increases rapidly by small increasing of the magnetic field. In this zone this behavior may be related to “weak” superconducting boundaries. In the upper limit zone the transition width increases smoothly by increasing of the magnetic field, this is may be due to “strong” superconducting grains.

The fitting functions and fitting parameters of the transition width dependence of magnetic field for lower limit zone and the upper limit zone are given in Eq. (3), solved together with the conventional boundary conditions.

$$\begin{cases} 
\Delta T(B) = m_1B + C_1 \text{ (lower limit zone)} \\
\Delta T(B) = m_2B + C_2 \text{ (upper limit zone)}
\end{cases}$$

(3)

From the lower and the upper limit zone, where they fitted to linear equation and the slope equal

$$\text{slope} = \left( \frac{d\Delta T}{dB} \right) \approx \frac{\Delta T}{B}$$

(4)
for the lower limit zone (in the linear region), when
\[ B \rightarrow B_{c1}, \text{ so } \Delta T \rightarrow \Delta T(0) \text{ and slope } = (\text{slope})_{\text{lower}}. \] Substituting in Eq. (4) we find
\[ \Delta T(0) = m_2 B_{c1} + C_1 \]
\[ (\text{slope})_{\text{lower}} = \frac{\Delta T(0) - c_1}{B_{c1}} \Rightarrow (\text{slope})_{\text{lower}} = \frac{\Delta T(0)}{B_{c1}} \]
and
\[ B_{c1} = \frac{\Delta T(0)}{(\text{slope})_{\text{lower}}} \]
thus
\[ B_{c1} = 0.268T \]
For the upper limit zone (at the beginning when the transition width \( \Delta T \) versus magnetic field becomes straight line)
\[ \text{When } B \rightarrow B_{c2}, \text{ so } \Delta T = T_c T_0 = T_c, \text{ where } T_0 \rightarrow 0 \text{ and slope } = (\text{slope})_{\text{upper}}. \]
Substituting this condition to Eq (4) we find
\[ (\text{slope})_{\text{upper}} = \frac{T_c}{B_{c2}} \Rightarrow B_{c2} = \frac{T_c}{(\text{slope})_{\text{upper}}} \]
and thus
\[ B_{c2} = 150T \]
Comparing Eq. (6) with The BCS equation \( B_{c2} = 1.83 T_c \), we note that the two equations have the same form, and the difference between them is the constant and this constant 1.83 equivalent to \((\text{slope})^{-1}_{\text{upper}}. \) The upper critical magnetic field \( B_{c2} \) in Eq.(6) doesn't depend only on the \( T_c \) but also depends on the chosen region, which determines the \((\text{slope})_{\text{upper}}. \) If we chose this region away from the \( B_c \) (the geometrical mean of critical magnetic fields), then the determination of \( B_{c2} \) will be more precisely.
According to GL theory (Ginzburg and Levanyuk, 1958), the GL parameter \( \kappa \) calculated using equation
\[ B_{c2} = \sqrt{2} B_c \kappa \] (Buckel and Kleiner, 2004; Kim et al., 2007; Aly et al., 2010; Raza et al., 2013)This leads the value of GL parameter \( \kappa \) is found to be 69.8.
The coherence length \( \xi \) is a difficult quantity to measure directly, and it is thus often calculated from upper critical magnetic field at the temperature \( T=0K. \) The relationship between coherence length and the upper critical magnetic field at \( T = 0K \) is as follows:
\[ \xi(0) = \frac{\Phi_0}{2 \pi B_{c2}} \]
\( \Phi_0 \) is the quantum of flux with a magnitude \( 2.07 \times 10^{-15} \text{ Tm}^2. \) The coherence length of the temperature \( T = 0K, \) is 1.48nm for the sample Ti-2234. From the definition \( (\lambda = K \xi) \) of the Ginzburg–Landau parameter
The penetration depth \( \lambda, \) is 103.3nm. These values \( (\xi, \lambda) \) are good approximations to the experimentally determined values for typical high temperature superconductors.

**Conclusion:**
In summary, we have determined the upper and the lower critical magnetic field of Ti-2234 sample by measuring the electrical resistance as a function of temperature at different applied magnetic field. The upper and lower critical magnetic field obtained from sample can be well scaled with its superconducting transition \( T_c. \) The two important parameters are estimated on the basis of the normal state and superconducting state properties. We have obtained the \( \kappa \) and \( \xi \) values for a Ti-2234 sample. Our result matches well with the experimental results available in the literature.

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