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Analysis of Magnetic Properties and Critical Current Density of Tl-2234 High-Temperature Superconductor Using AC Magnetic Susceptibility Measurements

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Abstract

This study investigates the magnetic properties and critical current density of $Tl_2Ba_2Ca_3Cu_4O_{11+\delta}$ (Tl-2234) high-temperature superconductors through AC magnetic susceptibility measurements. Samples were synthesized using a one-step solid-state process with careful heat treatment protocols to minimize thallium evaporation. AC susceptibility measurements were conducted across temperatures ranging from 140 to 50K under various applied AC magnetic fields (0.5-8 mT). The analysis revealed a sharp transition primarily reflecting intragranular superconductivity, with the field dependence of the imaginary (χ'') component providing insights into flux dynamics within the grains. The imaginary component peaks shifted towards lower temperatures with increasing magnetic field strength, indicating enhanced magnetic field penetration and increased intra-grain pinning centers. Using Bean's critical-state model, with an estimated effective particle radius of $R \approx 44.5$ µm based on sieve size, the critical current density (J_c) was calculated from the peak positions in $\chi''(T)$ curves. The temperature dependence of J_c followed an empirical scaling relation, yielding a zero-temperature critical current density $J_c(0)$ of 3.6×10^5 A/cm² and a critical exponent of 1.26 ± 0.08, quantifying key superconducting parameters of the prepared material.

Keywords: AC magnetic susceptibility; Current density; Flux pinning; Magnetic properties.

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1. Introduction

High-Temperature Superconductors (HTS) have emerged as materials of substantial scientific significance, owing to their transformative potential in energy-efficient technological applications, encompassing power transmission infrastructure, Magnetic Resonance Imaging (MRI) systems, and magnetic levitation technologies [1,2]. Within the broader classification of HTS materials, copper oxide-based (cuprate) superconductors demonstrate remarkable characteristics at operational temperatures substantially exceeding the liquid nitrogen boiling threshold (77 K), conferring distinct practical advantages over conventional superconducting systems [3]. Specifically, thallium-based cuprates (TBCCO) stand out for their relatively high critical temperature (T_c), which can be achieved under ambient pressure [4]. Within this family, the compound (Tl-2234) has gained attention due to its unique structural features and promising superconducting properties. Structurally, Tl-2234 belongs to the Tl-1223 family and is characterized by alternating layers of TlO and CuO_2, which play a crucial role in its magnetic and electronic properties [5-8]. These layered structures, combined with various types of crystal defects such as oxygen vacancies and grain boundaries, significantly influence the material's flux pinning mechanisms and, consequently, its critical current density (J_c). The analysis of alternating-current magnetic susceptibility has established itself as a sophisticated analytical technique for elucidating the intricate magnetic phenomena exhibited by high-temperature superconducting materials. This technique provides essential information about flux pinning mechanisms, critical current densities, and intergranular coupling in these materials. The AC susceptibility response is characterized by both real (χ') and imaginary (χ'') components, offering complementary insights into magnetic shielding and energy dissipation processes, respectively. The temperature and field dependence of these components reveals crucial details about flux penetration, pinning strength, and energy dissipation [9,10]. Despite the extensive research on Tl-2234, a comprehensive understanding of the correlations between its structural characteristics, AC magnetic susceptibility behavior, and critical current density remains incomplete. This knowledge gap is particularly significant given the material's potential for practical applications in high-field and high-temperature environments. This study aims to investigate the temperature and magnetic field dependence of AC susceptibility components (χ' and χ'') in Tl-2234, while analyzing the correlation between structural defects and magnetic properties through higher harmonic susceptibility measurements.

2. Experimental Methods

2.1 Sample Preparation

The synthesis of $Tl_2Ba_2Ca_3Cu_4O_{11+}\delta$ was initiated through the careful selection of high-purity precursor materials, including thallium oxide (Tl_2O_3), barium peroxide (BaO_2), calcium oxide (CaO), and copper oxide (CuO). The preparation followed a standard one-step solid-state process, where stoichiometric quantities of these precursors were thoroughly mixed using an agate mortar. To ensure homogeneity, the resulting powder was passed through an 89 µm sieve. The homogeneous powder mixture was subsequently compressed into a disc-shaped pellet with dimensions of diameter 1.5 cm and thickness 0.2 cm.

2.2 Heat Treatment

To minimize thallium evaporation during the heat treatment, the sample was carefully wrapped in silver foil. For safety considerations and to protect the furnace from potential hazardous effects, the wrapped sample was placed in a sealed quartz tube (diameter 1.5 cm × length 15 cm), which was then enclosed within a protective stainless steel tube. The heat processing methodology consisted of positioning the hermetically sealed specimen in a horizontal orientation within a furnace chamber, followed by controlled heating at a 6°C/min gradient until achieving 811°C. The specimen was held isothermally at this temperature for a duration of six hours, after which controlled cooling was implemented at 0.5°C/min until reaching ambient temperature. Superconducting characteristics were further optimized through supplementary heat treatment in ambient atmosphere at 500°C.

2.3 Magnetic Measurements

Magnetic Characterization Magnetic measurements were conducted using a LakeShore Superconducting Magnet System integrated with a helium cryostat. AC magnetic susceptibility components (real and imaginary) were recorded across AC field amplitudes from 0.5 mT to 8 mT, spanning a temperature range of 50 -140 K. Sample preparation involved mechanical grinding and size classification through an 89 µm mesh to ensure homogeneous particle distribution. It is important to clarify that the 89 µm mesh was used for particle size classification to ensure homogeneity of the sample, and this mesh size represents the maximum size of particles that were allowed to pass through, effectively setting an upper limit on the particle size in the measured sample.

3. Results and Discussion

3.1 AC Magnetic Susceptibility Analysis

The complex AC magnetic susceptibility ($\chi = \chi' + i\chi''$) serves as a powerful analytical tool for investigating high-temperature superconductors, particularly in granular systems where both components (χ' and χ'') exhibit distinct temperature and field amplitude dependencies [11]. AC susceptibility measurements have gained popularity due to their simplicity, cost-effectiveness, and high sensitivity, enabling sophisticated quantitative analysis [12]. When exposed to a periodic field, $H_a(t) = h \sin(2\pi\nu t)$, the sample exhibits a magnetic moment in opposition to the applied field, expressed as[13]:

$$\chi(h,\nu) = \chi' + i\chi'' \quad (1)$$

Where ν is the frequency. The real component (χ') approximates zero-field-cooling susceptibility in DC measurements, quantifying the superconductor's shielding response, while the imaginary component (χ'') reflects energy dissipation. In polycrystalline high-Tc materials, temperature-dependent AC susceptibility reveals superconducting transitions. The real component (χ') shows a sharp drop associated primarily with the onset of intragranular superconductivity. While distinct intergranular coupling effects can sometimes be observed in bulk polycrystalline samples, in powder samples such as the current sample, where intergranular connections are inherently weaker or disrupted, any signature of a separate intergranular transition is often masked or convoluted with the dominant intragranular response [14]. The χ'' curve also shows predominantly one peak, typically associated with losses within the grains. Figure 1 presents the temperature dependence of both real and imaginary components of AC magnetic susceptibility for the Tl-2234 sample under varying alternating magnetic fields. The sample demonstrates a well-defined superconducting transition, characterized by a sharp response with minimal broadening in the transition region.

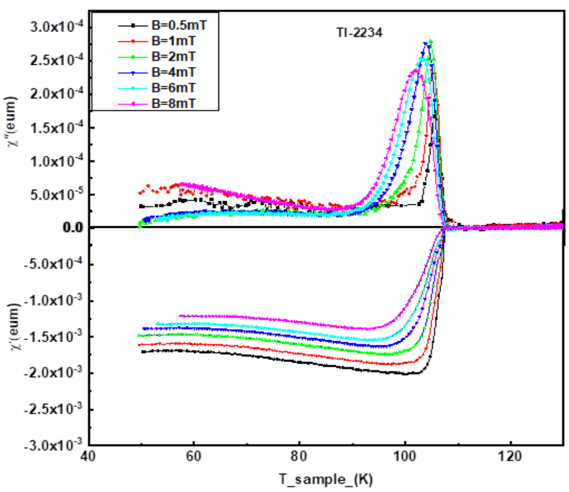

Figure 1. AC magnetic susceptibility vs. temperature for the powder sample Tl-2234 under different AC field amplitudes.

The real component exhibits modest field-dependent broadening across the transition. Notable features in the data include systematic shifts of the χ'' peaks toward lower temperatures with increasing alternating magnetic field amplitude. This behavior can be attributed to two primary mechanisms: first, the enhanced magnetic field strength within the grains leads to increased activation of intra-grain pinning centers, resulting from a reduction in the effective superconducting volume fraction. Second, the progressive shift of the penetration temperature (T_p) to lower values with increasing field strength correlates with an expansion of the frozen volume fraction at inter-grain boundaries, consequently diminishing the overall superconducting phase fraction. These observations provide insights into both the intra-grain and inter-grain magnetic response of the Tl-2234 system. After presenting the AC susceptibility data, it is important to discuss the observed critical temperature (T_c) value of approximately 107 K, as shown in Figure 1. This value is somewhat lower than the commonly reported T_c range of 113-116 K for optimally doped and single-phase Tl-2234 compounds, as documented in the literature [15,16]. Several factors may explain this discrepancy. First, the oxygen stoichiometry in Tl-based cuprates is highly sensitive and can significantly impact superconducting properties, including T_c. Even minor deviations from the optimal oxygen content can lead to a reduction in Tc. Second, as noted in the introduction, Tl-based superconductors are susceptible to phase inhomogeneity and intergrowth of different Tl-Ba-Ca-Cu-O phases. The presence of minor secondary phases or intergrown members of the homologous series, even if not easily detectable by bulk techniques like AC susceptibility, can influence the overall superconducting behavior and potentially lower the observed Tc. Additionally, the method used to determine Tc can affect the measured value. In this study, Tc was identified based on the onset of the diamagnetic transition in the real part of the AC susceptibility (χ'). Other techniques, such as resistivity measurements or alternative criteria applied to susceptibility data, might yield slightly different T_c values. While the obtained T_c is slightly lower than expected, it still falls within a reasonable range for Tl-2234-based materials.

3.2 Critical Current Density

The imaginary component of AC magnetic susceptibility (χ'') provides valuable information about energy dissipation mechanisms in materials, including eddy current losses, magnetic hysteresis, and magnetic flux dynamics under alternating fields [17]. Quantitative analysis of χ''(T) curves, particularly the position and field dependence of their maxima, enables determination of the critical current density (J_c) in superconducting materials. For measurements conducted at low frequencies, the temperature and field evolution of AC susceptibility is conventionally interpreted through critical-state models [18]. These theoretical frameworks describe the distribution of penetrated supercurrents, which flow at a density equivalent to the material's critical current density (J_c). A fundamental assumption in these models is that J_c depends solely on the local internal field (H_i) [11]. Bean's critical-state model, which simplifies the analysis by assuming J_c is Hi-independent, offers a direct methodology for extracting J_c values from temperature-dependent χ" measurements [19]. Within this framework, χ" exhibits a maximum when the applied field achieves complete penetration of the superconducting volume, specifically at the point where shielding currents match the sample's maximum current-carrying capacity [20]. As illustrated in Figure 2, the temperature dependence of critical current density can be systematically calculated from these χ" maxima. When an external magnetic field fully penetrates the entire volume of a superconductor, a condition is reached where the shielding currents become equivalent to the maximum carrier capacity (determined by hole concentration) or the bulk critical current density.

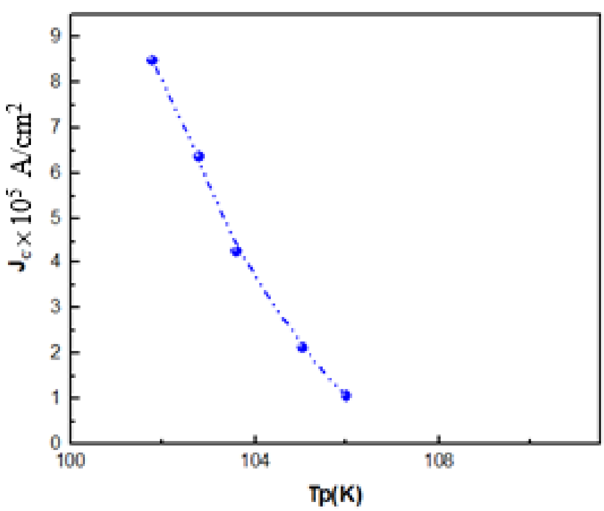

Figure 2. Temperature dependence of critical current density for the Tl-2234.

At the characteristic temperature T_p, corresponding to the maximum in the imaginary component of magnetic susceptibility (χ"), the penetration field H_p achieves complete sample penetration, reaching the center of the specimen. This penetration process can be quantitatively described using Bean's critical state model, which establishes a mathematical relationship between the critical current density J_c at the penetration temperature T_p and the penetration field H_p. This relationship has been well documented in previous studies [21-23], providing a fundamental framework for understanding the field penetration dynamics in superconducting materials.

$$J_c(T) = \frac{H_p}{R} \qquad (2)$$

Equation (2) employs the parameter 'R', which is defined as the radius pertinent to the sample geometry. For cylindrical samples subjected to an axial magnetic field, 'R' directly corresponds to the cylinder radius, as detailed in reference [24]. However, for powder samples, R necessitates interpretation as an effective average particle radius, as established in reference [25]. In this study, the effective radius R for the powder sample was estimated based on the 89 µm sieve used for particle size classification and homogenization. This mesh size represents the maximum particle diameter allowed to pass. Therefore, an effective average particle radius of R ≈ 44.5 µm (half the maximum diameter) was utilized in the J_c calculations using Equation (2). Subsequently, utilizing Equation (3), the critical current density was evaluated based on the peak position observed in the imaginary component (χ'') of the magnetic susceptibility. The critical current density J_c was calculated from the peak position of the imaginary part of AC magnetic susceptibility χ'' (Eq. 1). The temperature dependence of critical current density for the studied sample demonstrates strong agreement with the scaling relation expressed in equation (3) [14]:

$$J_c(T) = J_c(0)\left[1 - \frac{T}{T_c}\right]^{\alpha} \qquad (3)$$

Here, $J_c(0)$ represents the critical current density at absolute zero temperature (0 Kelvin) and α is the critical exponent. The critical current density value was obtained from the peak position. The solid line in Figure (3) shows the correlation between the experimental critical current density measurements as a function of temperature and equation (3) and the fitting parameters $J_c(0)$ and α were found to be $J_c(0) = 3.6\times10^5$ A/cm^2 and $\alpha = 1.26 \pm 0.08$. The study findings align well with prior research on thallium-based superconductors, highlighting the competitive superconducting characteristics of the Tl-2234 phase.

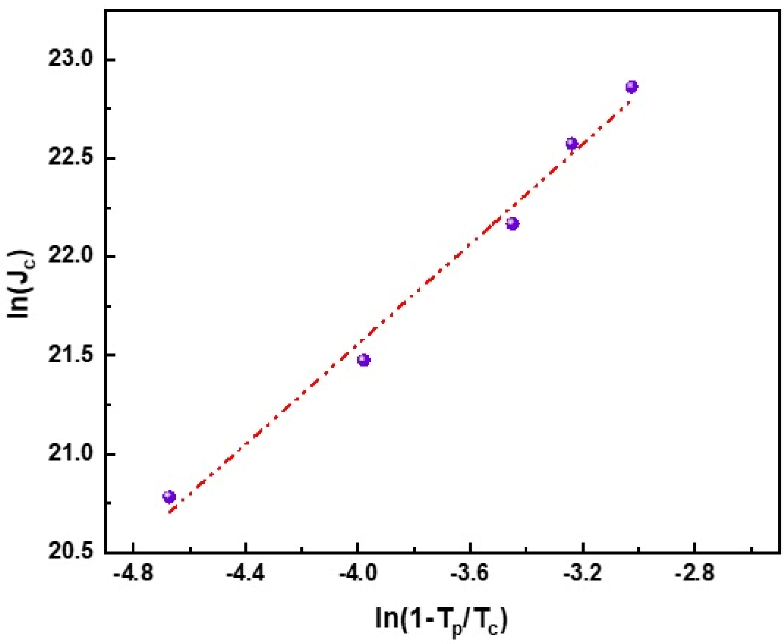

Figure 3. ln $J_c(T)$ vs.ln (1-T/T_c) for the Tl-2234 sample. The slope of line determines the value of 'α' specific to sample.

The observed critical current density $J_c(0) = 3.6\times10^5$ A/cm^2 suggests the material is of good quality with strong pinning centers, which is typical for well-prepared thallium-based superconducting systems. The estimated critical current density at zero temperature, $J_c(0)$, for the Tl-2234 sample falls within the range of 10^5 to 10^6 A/cm², consistent with the expected values for high-quality thallium-based superconductors. Reference [26] reports similar values for thallium cuprate superconductors, though measured at 77 K. In contrast, the $J_c(0)$ estimate in this study is extrapolated to 0 K, demonstrating that the obtained values align with the known high critical current properties of these materials, even at absolute zero. The measured $J_c(0) = 3.6\times10^5$ A/cm^2 is consistent with other thallium cuprate systems, such as Tl-2223 with $J_c(0)= 3.0\times10^6$A/cm^2 [27] and Tl-1223 with $J_c(0) \approx 2.8\times10^5$ A/cm^2. Additionally, The critical exponent $\alpha= 1.26 \pm 0.08$ further demonstrates consistency with established flux creep models for high-temperature superconductors, supporting both the Anderson-Kim flux creep model predictions and the expectations of collective creep theory (where the typical α range is 1.0 - 1.5) [28-31].

4. Conclusion

The AC magnetic susceptibility measurements revealed behavior dominated by the intragranular superconducting transition in the Tl-2234 system, as evidenced by the sharp drop in the real component (χ') and the characteristics of the imaginary component (χ''). The sharp transition observed in the susceptibility curves, accompanied by minimal broadening in the transition region, indicates good sample quality and homogeneity inferred from the magnetic response. The systematic shift of χ'' peaks toward lower temperatures with increasing alternating magnetic field strength provides evidence for enhanced magnetic field penetration effects and pinning within the grains. This behavior is attributed to an increase in intra-grain pinning centers, resulting from the synthesis and heat treatment process. Application of Bean's critical-state model to the AC susceptibility data, using an estimated effective particle radius of $R \approx 44.5$ µm, enabled successful determination of the critical current density (J_c), assumed to primarily reflect the intragranular current carrying capacity in this powder sample. The temperature dependence of J_c followed the empirical scaling relation with remarkable precision, yielding a zero-temperature critical current density $J_c(0)$ of 3.6×10^5 A/cm² and a critical exponent of 1.26 ± 0.08. These findings contribute significantly to understanding the relationship between the magnetically inferred microstructural features and the magnetic properties in Tl-2234 superconductors. Future research directions could focus on optimizing synthesis conditions to further enhance the critical current density and investigating the effects of various dopants on the magnetic properties and pinning mechanisms within this system.

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