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An investigation on the processing and characterization of (thallium(0.5-x) lead(0.5-x) europium(2x)) strontium(2-y) barium(y) calcium(2) copper(3) oxygen(z) high-temperature superconductor to determine the optimum barium concentration and study the effects of doping europium in thallium and lead sites

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An Investigation on the Processing and Characterization of $(\mathrm{Tl}_{0.5-x}\mathrm{Pb}_{0.5-x}\mathrm{Eu}_{2x})\mathrm{Sr}_{2-y}\mathrm{Ba}_{y}\mathrm{Ca}_{2}\mathrm{Cu}_{3}\mathrm{O}_{z}$ High Temperature Superconductor to Determine the Optimum Ba Concentration and Study the Effects of Doping Europium in Tl & Pb Sites

A Dissertation

Submitted

In partial Fulfillment

of the Requirements for the Degree of

Doctor of Industrial Technology

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Dedicated to

My wife JIN, GUANG

and to

My sons JU, REX BEN and JU, ROY BEN

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An Abstract of a Dissertation

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May 1996

Abstract

This research focused on $(Tl_{0.5-x}Pb_{0.5-x}Eu_{2x})Sr_{2-y}Ba_yCa_2Cu_3O_z$ compound to determine the optimum y (Barium concentration), and then studied the effects of doping Eu in Tl, Pb sites. Studies on optimum y give rise to a high quality superconducting material as a target material for sputter deposition of thin films for electronics applications. Studies on doping Europium (Eu) is a new investigation and might lead to better understanding of the structural properties of the superconductor.

The experiment was conducted in three steps as follows:

1. Optimizing the sample fabrication process by determining material sintering temperature in the furnace and sintering time in the air inside the Pt crucible.

2. Set Tl/Pb = 1, and determine the optimum y in $(Tl_{0.5}Pb_{0.5})Sr_{2-y}Ba_yCa_2Cu_3O_z$ by trying different y, starting from y = 0.1 to 1.0.

3. Determine the effect of Eu doping in Tl, Pb sites by experimenting different doping concentrations for Eu, starting from 1% to 10%.

Samples were prepared in two steps: precursor Sr-Ba-Ca-Cu-O were synthesized first, starting from high purity BaO, CaO, CuO and SrO. The second step was to add the appropriate amount of PbO and Tl_2O_3 (or Eu_2O_3 in case of Eu doping) to form the superconducting compound. Each sample was heat

treated at 900°C for 24 hours twice and the pellets were made under the pressure of 15,000 PSI.

The $(Tl_{0.5}Pb_{0.5})Sr_{2-y}Ba_yCa_2Cu_3O_z$ superconductors with y varying from y = 0 to y = 1.0 were processed and sintered in air inside a Pt crucible at 880°C for 10 min. Sintered samples were characterized using the ac susceptibility and x-ray diffraction measurements.

From the AC susceptibility measurements, the critical temperature of the samples (Ba = 0.0 to Ba = 1.0) is between 100°K and 120°K. Sample with Ba doping of 0.75 showed the highest T_C of 118°K indicating the dominance of 1223 phase in this superconducting compound. The Barium concentration of 0.75 also shows higher magnitude of χ' indicating higher volume fraction of superconductivity.

The powder x-ray diffraction measurements indicated that the strongest peak for the 1223 phase is the (105) peak close to $2\theta = 30°$. At lower concentration of Ba below 0.5, the intensity of (105) peak is slowly increasing and then above 0.5, the intensity of the 1223 peak improves, indicating the growth of significant 1223 phase in the superconductor. The sample is predominantly 1212 phase for Barium concentration below 0.5.

Other studies of importance in this research is the doping of Europium in Tl and Pb sites. Europium being a rare-earth metal offers the possibility of magnetic studies using Mössbaur spectroscopy. This gives an unique opportunity to study the structure of TlPbSrBaCaCuO superconductor precisely. As Eu concentration in the superconductor increases, the T_C decreases gradually. With the maximum doping fraction of 2x = 0.1, the T_C dropped close to 100°K from 118°K for 2x = 0.0. The magnitude of real part of susceptibility is higher than $1x10^{-3}$ (emu/gm) for Eu = 0, and decreases to $3x10^{-4}$ (emu/gm). A notable observation in the Eu doping superconducting samples is the paramagnetic nature of the samples above the critical temperature.

CHAPTER I

INTRODUCTION

Recently, thallium compound superconductors have gained special attention among the new high temperature superconductors. Thallium-lead-strontium-calcium-copper-oxide (Tl-Pb-Sr-Ca-Cu-O) based superconducting compounds offer the advantages of high critical temperatures and superior superconducting properties (Sheng & Hermann, 1988). Thin films of thallium based compounds have potential impact on electronics applications due to their high critical temperature (T_c) that can be as high as 120°K, high critical current density (J_c) which is greater than 10^6 A/cm^2 at 77°K, and low microwave surface resistance (R_s) with values of almost two orders of magnitude lower than Cu at 77°K and 10 GHz (Subramanyam, Ju, Shand, Zhang, & Shi, 1995).

Recently, Dr. G. Subramanyam and others including the author at the University of Northern Iowa have performed research on synthesis and characterization of $(Tl_{0.5}Pb_{0.5})Sr_2Ca_2Cu_3O_z$ (1223) bulk superconductors (Subramanyam et al., 1995). Synthesis of Tl-1223 phase bulk superconductors for sputtering targets was studied using three different methods. The first method used a stoichiometric starting compound of $(Tl_{0.5}Pb_{0.5})Sr_2Ca_2Cu_3O_z$ (1223), in the second the Ca/Sr ratio was varied and in the third partial Ba doping for Sr sites was used. The stoichiometric starting compound of 1223 after high temperature heat treatments yielded only the lower T_c phase, namely, the $(Tl_{0.5}Pb_{0.5})Sr_2CaCu_2O_z$ (1212) superconductor. Increasing the Ca/Sr ratio to 1.5 yielded mixed phase samples with the promotion of the 1223 phase, but with a significant amount of 1212 phase. Samples with Ba doping in Sr sites have yielded the highest T_c and a dominant 1223 phase.

The samples were processed in Platinum crucibles at various temperatures to obtain the optimum processing conditions. All the samples required an additional amount of Tl and Pb to compensate for loss during high temperature heat treatments. X-ray diffraction, vibration sample magnetometer, and ac susceptibility measurements were used for sample analysis.

Based on past experiments, further exploration on Tl-Pb-Sr-Ca-Cu-O based superconductors were made. This research was focused on finding optimum y, as well as studying the effect of Eu substitution for Tl and Pb in the $(Tl_{0.5-x}Pb_{0.5-x}Eu_x)Sr_{2-y}Ba_yCa_2Cu_3O_z$ compound to obtain the best superconducting properties.

Statement of Problem

The problem of this study is to determine the optimum Barium concentration (y) in the **$(Tl_{0.5-x}Pb_{0.5-x}Eu_{2x})Sr_{2-y}Ba_yCa_2Cu_3O_z$** high temperature superconducting compound to obtain superior superconducting properties (high critical temperature T_c and superconducting volume fraction) and phase purity. In addition, substitution of Eu (Europium) for Tl and Pb would be investigated to get optimum Europium concentration (x). Eu is a rare earth metal with interesting magnetic properties. Doping Eu for Tl & Pb sites would allow Mossbaür Spectroscopy studies to determine the crystal structure and might also help to understand the mechanism of superconductors.

The Barium concentration and Europium concentration in this Tl compound are independent variables. The dependent variables are superconducting properties which include critical temperature (T_C), superconducting volume fraction (Vsc) and current density (J_C), and phase purity.

Statement of Purpose

The main purpose of this research was to optimize the Ba concentration in the Tl-Pb-Sr-Ba-Ca-Cu-O compound to produce high quality 1223 phase high temperature superconducting compound. The high quality superconductor would be used for fabrication of superconducting thin films of thickness less than 1 µm on substrates such as $LaAlO_3$ (lanthanum aluminate).

Significance of the Study

The **Tl-Pb-Sr-Ca-Cu-O** compound superconductors have proven to be superior for electronic applications among the high temperature superconductive materials available (Sheng & Hermann, 1988), because of its lower intrinsic defect structure and strong flux pinning resulting in higher critical current density. The main advantages of the Tl-Pb-

Sr-Ca-Cu-O system are the strong flux pinning associated with single Tl layer structures (Tkaczyk et al., 1992).

This research focused on $(Tl_{0.5-x}Pb_{0.5-x}Eu_{2x})Sr_{2-y}Ba_yCa_2Cu_3O_z$ compound to determine the optimum y (content of Barium), and then studied the effects of doping Eu in Tl, Pb sites. Studies on optimum y give rise to a high quality superconducting material as a target material for sputter deposition of thin films for electronics applications. These studies will lead to high T_C, higher volume fraction of superconductivity, and phase pure superconductors.

Studies on doping Europium (Eu), to the author's knowledge has not been investigated to date. This would be a new investigation and might lead to better understanding of the structural properties of the superconductor. Since the trace of Eu could be detected by the study of magnetic ordering using techniques such as Mössbaur Spectroscopy, it was hoped that basic information on the superconducting mechanism may be understood by studying the doping effects. Once a process was established for fabricating good quality bulk superconductors, it would be used to fabricate thin film superconductors at the University of Northern Iowa.

Research Questions

1. What is the optimum y in $(Tl_{0.5-x}Pb_{0.5-x}Eu_{2x})Sr_{2-y}Ba_yCa_2Cu_3O_z$ needed to obtain the superconducting properties (susceptibility, magnetization, critical temperature (T_C),

critical current density (J_C) and volume fraction of superconductivity).

2. How does varying y affect the superconducting properties, T_C, volume fraction of superconductivity, and phase purity?

3. What are the characteristics of the structural properties of $(\mathrm{Tl}_{0.5-x}\mathrm{Pb}_{0.5-x}\mathrm{Eu}_{2x})\mathrm{Sr}_{2-y}\mathrm{Ba}_y\mathrm{Ca}_2\mathrm{Cu}_3\mathrm{O}_z$ to be studied using x-ray diffraction?

4. What are the effects of doping Eu for Tl & Pb in $(\mathrm{Tl}_{0.5-x}\mathrm{Pb}_{0.5-x}\mathrm{Eu}_{2x})\mathrm{Sr}_{2-y}\mathrm{Ba}_y\mathrm{Ca}_2\mathrm{Cu}_3\mathrm{O}_z$ on the superconducting properties (susceptibility measurements-magnetization, critical temperature, volume fraction of superconductivity and phase purity).

Definition of Terms

Critical Temperature, T_C:

When resistivity of superconducting samples are measured, " . . . T_C is the temperature where the resistivity has decreased to half its value at the onset temperature (T_{CO}). The onset temperature for superconducting phenomena is the level where the resistivity versus temperature curve just begins to depart from the normal to superconducting behavior . . . " (Doss, 1989, p. 56). When the magnetic susceptibility measurements are performed, T_C is defined as the temperature at which the magnetic susceptibility starts to be negative, as the sample is being cooled.

Critical Current Density, J_c:

"The current density that, when exceeded, causes the material to "quench", that is, lose its superconducting properties, is J_c" (Doss, 1989, p. 67). Typically, the J_c is taken as the critical current density that gives rise to an electric field of 1 μv/cm.

Critical Thermodynamic Magnetic Field, Hc:

The superconductor's free energy is utilized in the expulsion of flux. " . . . Once the free energy can no longer be expanded without destroying the superconducting state, the material 'quenches', that is, converts to the normal state of finite dc resistance" (Doss, 1989, p. 65). The critical Magnetic field H_{c2} is the magnetic field above which the material becomes non-superconducting.

Meissner Effect:

Meissner effect refers to the phenomenon that magnetic fields are expelled from the superconductor when the superconductors are well below critical temperature T_c and critical magnetic field H_c. Meissner effect is the criterion to judge if the a compound or an alloy is superconducting.

Two-Fluid Models:

The current transport in the superconducting state is a mixture of paired electrons and unpaired electrons. This model of superconducting behavior is called the Two-Fluid

Model. It reflects that not all conduction electrons participate in the superconducting behavior.

Magnetic Susceptibility:

The magnetic susceptibility represents the magnetic response of the material to an applied field H.

Tl-based Compounds:

All of the Tl-based compounds can be described by the general formula, $Tl_mA_2Ca_{n-1}Cu_nO_{2n+m+2}$, where m = 1 or 2; n = 1 to 5; A = Ba or Sr. For convenience, the names of these compounds are abbreviated as 2223 for $Tl_2Ba_2Ca_2Cu_3O_{10}$, where each number denotes the number of Tl, Ba(Sr), Ca and Cu ions per formula, respectively. The compounds with m = 1 and m = 2 are usually referred to as single and double Tl-O layered compounds, respectively (Parkin et al., 1988; Toradi et al., 1988).

Crystal Structure:

A crystal structure has a structural regularity because covalent bonds maintain a specific number and orientation of neighbors for each atom and repeats the pattern in three dimension (Vlack, 1964).

CHAPTER II

REVIEW OF LITERATURE

Research Background

In the 19th century, it was known that the resistivity of metallic electrical conductors decreased as they were cooled to lower temperatures. It was predicted that the metallic resistivity could be decreased to zero at low enough temperatures. However, the cryogenic technology had not been developed, and it was impossible to cool samples to low enough temperatures to test the prediction of the zero resistance (Doss, 1989).

It was not until 1908 that Kamerlingh H. Onnes successfully prepared the first liquefied helium, which broke through the scientific barrier of studying superconductivity. With this new technology, Onnes experimented with mercury (Hg) and found a rapid decrease in the resistance at 4.25°K, with the level becoming almost zero below 4.2°K. Later, he tested other metallic elements, such as indium (superconducting at 3.4°K), tin (superconducting at 3.7°K) and lead (superconducting at 7.2°K) (Dahl, 1992; Doss, 1989).

In 1933, Walker Meissner and Robert Ochsenfeld discovered that magnetic flux was expelled from superconductors operating well below critical temperature T_C and critical magnetic field H_C. This phenomenon is the famous "Meissner effect" which was a breakthrough in the understanding of superconductivity. Another important development in 1930s was the "two-fluid" model for modeling the electron flow pattern in superconducting material (Dahl, 1992; Doss, 1989).

During the period from approximately 1930 to 1950, a lot of work was done by researchers in their attempts to understand the mechanism of superconductors. To explain the Meissner effect, F. and H. London presented their work of exploring new relationships between the superconducting current flow and the magnetic field (Doss, 1989).

Superconductors were classified either as Type I or Type II. Type I is used to specify the pure metallic superconductors. For type I supercoductor there is only one critical magnetic field H_c. If external magnetic field is greater than H_c the superconducting material will become normal immediately. Type II is used to specify the alloy superconductors. For type II superconductor there are two critical magnetic fields H_{c1} and H_{c2}. Even if the external magnetic field is over H_{c1} the superconducting behavior is still existing and gradually decreasing until magnetic field reach to H_{c2}. Superconducting material is in a mixed state between H_{c1} and H_{c2}. The first oxides found to be superconducting were NbO and TiO. The type II materials were proved to be capable of transporting enormous currents while operating in high magnetic fields (Buckel, 1991; Dahl, 1992; Doss, 1989). The age of oxide superconductors started in the early 1960s (Table 1). Generally oxides are not considered as having good metallic properties except ReO_3 and RuO_2. However, the first oxides found to be superconducting were NbO and TiO. These oxides may be viewed merely as metals which have dissolved some oxygen (Sleight, 1988).

Table 1

History of Oxide Superconductors

Compound	Tc (°K)	Date discovered
TiO, NbO	1	1964
$SrTiO_{3-x}$	0.7	1964
Bronzes		
A_xWO_3	6	1965
A_xMoO_3	4	1969
A_xReO_3	4	1969
Ag_7O_8X	1	1966
$LiTi_2O_4$	13	1974
$Ba(Pb,Bi)O_3$	13	1975
$(La,Ba)_2CuO_4$	35	1986
$YBa_2Cu_3O_7$	95	1987
Bi/Sr/Cu/O	22	1987
Bi/Sr/Ca/Cu/O	90	1987
Tl/Ba/Ca/Cu/O	122	1988
K/Ba/Bi/O	30	1988

Note. From "Chemistry of High-Temperature Superconductors" by A. W. Sleight, 1988, Science, 242, p. 1519.

The modern high-Tc superconductors began in April 1986 with reporting of superconductivity in lanthanum-barium copper oxide at 30°K (Bednorz & Müller, 1986). Late in 1986, Paul Chu and his colleagues demonstrated that, under very high pressure, La-Ba-Cu-O showed the superconductivity behavior above 40°K. On Jan. 29, 1987, the team at the University of Alabama/Huntsville discovered the famous Y-Ba-Cu-O superconductor $YBa_2Cu_3O_7$ which was called the 1-2-3 compound (Wu et al., 1987).

Since the famous 1-2-3 superconductor was discovered, a lot of research have been conducted on superconductors with higher T_c and better properties. Zhengzhi Sheng and Allen Hermann reported their success with a Thallium compound (Tl-Ca-Ba-Cu-O) that showed an onset of superconducting behavior at about 120°K, and zero dc resistance at 106°K (Sheng & Hermann, 1988). The thallium compound was considered to be the potential superconductor of the future (Raveau et al., 1989). The Tl-Pb-Sr-Ca-Cu-O superconductor has showed the best properties for electronic applications among the high T_c superconductors. This is the goal of this researcher to further explore thallium-lead-strontium-calcium-copper-oxide (Tl-Pb-Sr-Ba-Ca-Cu-O) based superconducting compounds, to synthesize bulk single phase superconductors.

Characteristics of Superconductors

All superconductors exhibit zero (dc) electrical resistivity and Meissner effect below a critical transition temperature, T_c. The transition from normal conduction to superconduction is determined not only by the temperature but also by the magnetic field strength (H) at the surface of the samples. There is a critical magnetic field H_c, which, when exceeded, will destroy superconductivity. The current density (J_c) is another important factor to be considered. If a current density (J_c) above the critical value is transported through the superconductor, this also destroys the superconductivity. These three parameters, that is, temperature, magnetic field and current density are closely related in determining the state of the superconductor (Buckel, 1991; Burns, 1992).

The Meissner effect is considered the better criterion to judge if a material is superconducting, because it is difficult to measure absolute zero resistivity. The essential point of the Meissner effect is that when the external H field is lowered below a critical level, the superconductor expels the internal magnetic field. Therefore the reports of superconductivity in new compounds or alloys must meet the Meissner effect criterion before it could be accepted (Buckel, 1991). The type I materials must exclude all of an applied magnetic field to remain superconducting. The systems have a tendency to seek their lowest energy state. For superconductor, the lower energy state is the superconducting state. As temperature is lowered farther below T_c, the difference in energy between the superconducting and the normal state increases. If a magnetic field is applied, energy is required from the superconductor for the expulsion of flux. The type II materials are able to retain their superconductive characteristics in rather intense magnetic fields (Burns, 1992).

The current flow in superconductors is comprised of electron pairs, instead of individual electrons in the normal metallic state. The two-fluid model, which explains electron current flow patterns in superconductivity, describes the superconducting state as a mixture of paired electrons (responsible for superconducting current) and unpaired electrons (responsible for normal current) (Buckel, 1991).

Applications of Superconductivity

The high temperature superconductivity (particularly T_C = 90-125 K) makes it possible to expand its applications, because of the lower cost of liquid Nitrogen (77K) compared to liquid Helium (4°K). The first reported applications for High-T_C superconducting materials are in electronics such as SQUIDs (Superconducting Quantum Interference Devices) operation. A SQUID device must be biased so that small changes in flux will result in the critical current of the weak link being exceeded.

Recently, there has been a lot of progress in the development of high-T_C SQUIDs. It has been demonstrated that SQUIDs have sufficient sensitivity for applications such as in the measurement of magnetic fields and nondestructive testing. Khare and Chaudhari (1994) reported, "a bicrystal junction high-T_C direct current-SQUID has been operated successfully at 77°K in a portable battery operated microcooler" (p. 2353). Multilevel high T_c SQUIDs, suitable for digital circuit applications, have been fabricated and tested (Forrester, Davidson, Talvacchio, Gavaler, & Przybysz, 1994). $YBa_2Cu_3O_{7-x}$ based monolithic integrated dc SQUID magnetometers, have been fabricated and characterized (Higenkamp, Brons, Soldevilla, Ijsselstijin, & Flokstra, 1994).

Koch, Rozen, Sun and Gallagher (1994) have invented a three superconducting quantum interface device gradiometer (TSG) that uses three SQUID magnetometers and a novel feedback method to measure magnetic field gradients. One SQUID is the reference, while the other two are sensors. The feedback loops for the sensors measure the differences between the magnetic field at the reference location and those at sensor locations. The voltage difference between the two sensor SQUID outputs divided by the distance between two sensors represents the average magnetic field gradient.

Superconducting Josephson circuits are another good example of applications. They are ideal for very large scale integrated neural networks, because of their ultra-high-speed operation and very low power dissipation. An artificial neural network is a candidate for intelligent information processing. It is comprised of many neuron elements, which are connected to one another. Mizugaki, Nakajima, Sawada and Yamashita (1994) presented superconducting synapse circuits, which allow digitally variable synaptic weights. Each bit of the synapse circuit consists of 2^m ($m = 0, 1, 2, \ldots$) double-junction SQUIDS. "This is the first successful implementation of such an active synapse into superconducting neural network" (Mizugaki et al., 1994, p. 1712).

Superconducting magnets with different size and geometry have already been produced. They are used in scientific research, in high energy and solid state physics. They are also going to be used for controlled fusion, superconducting motors, energy storage facilities, and the magnetic suspension of trains (Buckel, 1991).

High-T_c Superconductors (HTS) with high current density (> 10^6 A/cm^2) can be applied in computer-chip manufacturing. Progress is being made in the area of manufacturing hybrid combinations of high-temperature superconductors and semiconductors (Doss, 1989).

With continued research, more and more applications are being explored in different fields. It will be possible to use superconductors to transmit electrical power, which can offer advantages in terms of low power losses. Superconductors will also provide an opportunity in measurement technology to increase the sensitivity by using its magnetic characteristics. Superconducting thin film transmission lines will have an impact for electronic circuits and in high speed computers (Buckel, 1991).

Thallium-Barium-Copper Oxide and Related Compounds

Thallium System Superconductor

During the last few years, high-temperature superconductivity of thallium compounds has gained particular attention following the reports of the onset of superconductivity near 120°K in oxides containing copper, alkaline earth, and thallium. The new series of thallium compounds were represented as $Tl_2Ba_2Ca_{n-1}Cu_nO_{4+2n}$, where n is the number of CuO sheets stacked consecutively (Eibschütz et al., 1988; Qiu & Shih, 1988). Table 2 presents the Thallium-copper-oxides summarized by Professor Greenblatt (1989).

A great deal of work has been devoted to the research of new superconductive cuprates since the beginning of 1987. Other dissertation research on Tl-system related topic are summarized in Table 2. Raveau, Michael, Martin and Hervieu (1989) stated, "besides the '123' family and the La_2CuO_4 -type series, two new series of cuprates appear to be most promising, namely the bismuth and the thallium compounds, which exhibit the highest critical temperatures (108°K for bismuth and 125°K for thallium)" (p. 257).

Table 2

Superconducting Thallium-Copper-Oxides

Compound	T_c (°K)
$Tl_2Ba_2CuO_6$	0-90
$Tl_2Ba_2CaCu_2O_8$	110
$Tl_2Ba_2Ca_2Cu_3O_{10}$	115-125
$Tl_2Ba_2Ca_3Cu_4O_{10}$	104-110
$TlBa_2Cu_5O_5$	metallic
$TlBa_2CaCu_2O_7$	50-90
$TlBa_2Ca_2Cu_3O_9$	110-117
$TlBa_2Ca_3Cu_4O_{11}2$	120
$TlBa_2Ca_4Cu_5O_{13}$	110
$(Tl,Bi)Sr_2CuO_5$	metallic
$(Tl,Bi)Sr_2CaCu_2O_7$	90
$(Tl,Bi)Sr_2Ca_2Cu_3O_9$	120
$(Tl,Pb)Sr_2CuO_5$	0-20
$(Tl,Pb)Sr_2CaCu_2O_7$	80-85
$(Tl,Pb)Sr_2Ca_2Cu_3O_9$	115
$TlSr_2CaCu_2O_7$	100

Note. From "Chemistry and Superconductivity of Thallium-based cuprates" by M. Greenblatt, S. Li, L. E. H. Mcmills, & K. V. Ramanujachary, 1988.

Table 3

Summary of Tl-System Related Dissertation Research

#1	Investigations on the (Tl,Pb,Bi)Sr_2CuO_5 superconductors and $Ba_{2-x}K_xPbO_4$ oxides.
Year	1993
By	Pan, Min-Hsueh
Sch.	The State University of New Jersey-New Brunswick
#2	Sputtered Tl-Ba-Ca-Cu-O superconducting thin films.
Year	1993
By	Wu, Lixue
Sch.	Wichita State University
#3	Thallium-based high temperature superconducting thin films for microwave electronics applications.
Year	1993
By	Subramanyam, Gurunathan
Sch.	University of Cincinnati
#4	Electronic transport properties of single crystal thallium-2201 superconductors.
Year	1992
By	Yandrofski, Robert Michael
Sch.	Harvard University
#5	Novel synthesis of Tl-Ba-Ca-Cu-O superconductors.
Year	1992
By	Bayya, Shyam Sundar
Sch.	Alfred University
#6	The phase development and microstructure of thin-film superconductors in the Tl-Ba-Ca-Cu-O system.
Year	1992
By	Lanham, George Michael
Sch.	University of California, Santa Barbara
#7	Magnetic penetration depth in Tl-based superconducting single crystals.
Year	1991
By	Ning, Hong
Sch.	University of South Carolina

(table continues)

#8 Resistive anomalies in a distributed point contact proximity effect junction: A study of the high T_c superconductor $Tl_2Ba_2Ca_2Cu_3O_{10}$.
Year 1991
By Holcomb, Matthew James
Sch. Stanford University

#9 Synthesis, characterization, and thermoelectric power study of thallium-based high Tc superconductors.
Year 1991
By Xin, Ying
Sch. University of Arkansas

#10 Transport properties of high temperature superconductor TlBaCaCuO thin films.
Year 1989
By Kim, Dong Ho
Sch. University of Minnesota

Note. Information was obtained from Dissertion Abstracts International.

Qiu (1988) stated, "This new [thallium] system, along with the other system of Bi-Ca-Sr-Cu-O, is especially interesting because of the possible improved thermal stability over the Y-Ba-Cu-O compounds" (p. 1122). Using a multilayer thermal evaporation method, they prepared the thin films of the Tl-Ca-Ba-Cu-O compound in their laboratory on conventional alumina substrates. The prepared films showed a resistance transition at about 120°K with zero resistance temperature of 85°K after a prolonged treatment at 660°C (Qiu & Shih, 1988).

It is reported that thallium cuprates differ from the bismuth compounds by the existence of numerous extended defects which affect both rock salt-type layers and oxygen deficient copper layers. Based on their research, Raveau et al. (1989) announced, "the synthesis of new high T_C superconductive cuprates with a bidimensional character is favored by the presence of two species, namely lone pair cations and oxidizing alkaline earth cations, but that experimental conditions play an important role in the synthesis of new phase" (p. 257).

The bidimensional character of the structure necessary for superconductivity occurred at the synthesis of superconductive bismuth cuprates, "is also confirmed by the fact that thallium strontium cuprates and even bismuth '2223' cuprates are stabilized by the introduction of lead into the matrix in agreement with the existence for both cations, Bi^{III} and Pb^{II}, of the $6s^2$ lone pair" (Raveau et al., 1989, p. 262).

Tl-Ca-Ba-Cu-O has a variety of superconducting phases, and exhibits higher T_C and better superconducting properties than 1-2-3 compounds. The phase formation of the Tl-Ba-Ca-Cu-O superconductors showed the importance of the thermodynamic factors. There are four types of Tl-based CuO superconductors: $Tl_2Ba_2Ca_2Cu_3O_{10}$ (Tl-2223), $Tl_2Ba_2Ca_1Cu_2O_8$ (Tl-2212), $TlBa_2Ca_2Cu_3O_{8.5}$ (Tl-1223), and $TlBa_2Ca_1Cu_2O_8$ (Tl-1212). High-quality thin films of Tl-1212, Tl-2212, and Tl-

2223 have been reported with transport Jc of 10^6 A/cm^2 at 77°K and zero field (Ren, Wang C. A., & Wang J. H., 1994). Tl-1223 is considered to be the most promising Tl-based superconductive phase, because it has higher zero resistance T_c than Tl-1212 and better flux pinning properties than Tl-2223 and Tl-2212 (Ren, Wang C. A., & Wang J. H., 1994).

Liu, Zheng, Loram, Mirza and Campbell (1992) observed a high critical-current density in the material $(Tl_{0.5}Pb_{0.5})Sr_2Ca_2Cu_3O_9$. This single Tl-O layer material has a J_c of $1.24\text{x}10^5$A/cm^2 when measured at a temperature of 77°K and a field of 1 T. It was indicated that the $(Tl_{0.5}Pb_{0.5})Sr_2Ca_2Cu_3O_9$ compound has a three-dimensional character due to the enhancement of the superconducting coupling along the c axis between the Cu-O planes and thus results in an increase in J_c compared to the two-dimensional Tl-2212 and 2223 compounds.

Ahn, Lee and Beyers (1992) investigated the formation of $Tl_2Ca_2Ba_2Cu_3O_{10\pm\partial}$ as a function of oxygen pressure and temperature. The results explained the difficulties in forming single-phase $Tl_2Ca_2Ba_2Cu_3O_{10\pm\partial}$ and suggested new methods for making bulk and thin-film Tl-Ca-Ba-Cu-O superconductors. It was found out that the temperature needed to form $Tl_2Ca_2Ba_2Cu_3O_{10\pm\partial}$ can be greatly reduced by using low oxygen pressures during synthesis.

Myers, Face, Kountz and Nestlerode (1994) reported the growth of thin films of $(Tl,Pb)Sr_2Ca_{1-x}Y_xCu_2O_7$ with $x = 0-0.2$ by off-axis magnetron sputtering in the presence of thallium oxide vapor. Yttrium doping on the calcium site increased the transition temperatures, 86°K for $x = 0.1$ and 93°K for $x = 0.2$.

It was understood that grain boundaries of high-T_c cuprate superconductors act as a weak link which limit the transport critical current density J_c. Nabatame et al. (1994) investigated weak link properties in the $Tl_1Ba_2Ca_2Cu_3O_x$ [Tl-1223] system and measured the transport properties of artificially grown grain boundaries in a Tl-1223 film. It was found out that the grain boundary J_c for large tilt angles ($T \geq 15°$) at 77°K and 5°K were substantially less than intragrain J_c and limited by a weak link. It was indicated that the low tilt angle grain boundaries do not act as a weak link and retain high J_c (Nabatame et al., 1994).

Sarnell (1994) has investigated the electrical properties of Tl-Ba-Ca-Cu-O high-T_c superconductor bicrystal grain boundary junctions. "The behavior of the transport parameters is very similar to those obtained in $YBaCuO_{7-\partial}$ junctions, . . . [r]elatively good control of the transport parameters as a function of the misorientation angle has been achieved" (Sarnell & Chaudari, 1994, p. 362).

Because of the mixed metal valance state of thallium in a Tl-based superconductor, Chiang et al. (1991) prepared an almost single "1223" phase by a modified Pechini method (US Patent 3 330 697). Tl_2O_3 and other metal nitrates are dissolved in an aqueous solution plus a certain amount of oxalic acid for chelating the metal cations. The desired phase showed zero resistance at 114°K.

Thallium loss is a major problem for the formation of Tl-based single phases because the starting material, Tl_2O_3 is volatile at high temperatures. When the Tl-containing starting materials are heated at low temperatures, the rate of thallium loss can be effectively reduced (Doss, 1989).

Superconducting phases of thallium compound can be formed by diffusion of Tl into Ca-Ba-Cu multilayer films deposited on alumina substrates. However, "for the treated films with the multilayer structure, the morphology was not entirely satisfactory for fine line etching and further processing" (Qiu & Shih, 1988, p. 1122). A sputtering method was tried with a single target. Because of the volatility of the Tl at elevated temperatures, it is relatively difficult to prepare a sintered Tl-Ca-Ba-Cu-O target with appropriate Tl content for the sputtering. Finally, it was prepared by rf magnetron sputtering from a presintered Ca-Ba-Cu-O target (Qiu & Shih, 1988).

The Role of Copper-Oxide

Copper oxides are categorized described as $Cu^{III}-O^{-II}$ or $Cu^{+2}-O^{-1}$. The $Cu^{III}-O^{-II}$ is the correct oxidation state representation, while $Cu^{+2}-O^{-1}$ is the real charge representation. The basic building block for oxides of Cu^{II} and Cu^{III} are CuO_4 units that may be isolated, or share corners or edges to form infinite structures (see Figure 1). The resulting structures tend to be sheets or chains (or both) of linked CuO_4 units. One of the simplest methods of connecting CuO_4 units together is to forms the infinite sheet structure. This sheet is present in all the Cu-O based superconductors (Doss, 1989; Sleight, 1988).

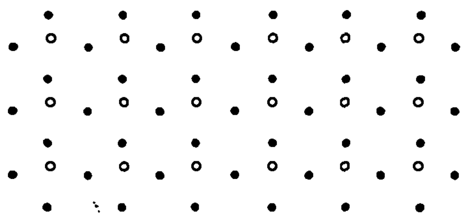

Figure 1. The Cu-O Sheet. From "Chemistry of High-Temperature Superconductors" by A. W. Sleight, 1988, Science, 242, p. 1521.

Sleight (1989) stated, "The recent dramatic increases of T_c have given reason to believe that some totally new mechanism for superconductivity is operating in the oxides of

Cu and Bi" (p. 1525). He also predicted, "On the basis of our guidelines for superconductivity, we can expect to see T_c's continue to rise somewhat for Cu-O systems" (p. 1527).

It has been demonstrated that the critical temperature (T_c) of $TlBa_2Ca_{n-1}Cu_nO_{2n+3}$ rises with increasing the number of Cu-O layers. $TlSr_2Ca_{n-1}Cu_nO_{2n+3}$ with n = 2 and 3 were synthesized and reported that $TlSr_2CaCu_2O_7$ ("1212" phase) (for n = 2) has a critical temperature of 75°K and $TlSr_2Ca_2Cu_3O_9$ ("1223" phase) (for n = 3) 100°K. Lately T_c was raised up to 90°K for 1212 phase and 122°K for the 1223 phase by replacing part of Tl by Pb. The highest zero resistance temperature obtained for $(Tl_{0.5}Pb_{0.5})Sr_2Ca_2Cu_3O_9$ was 115°K. Consequently, $(Tl,Bi)Sr_2CaCu_2O_y$ (Tl/Bi = 1 and y = 7) was prepared with T_c = 95°K by a solid-state reaction method using $Sr_2Tl_2O_7$ as a Tl-containing precursor (Kaneko, Wada, Yamauchi, & Tanaka, 1990, p. 1281).

Kaneko et al. (1990) investigated the superconducting compound $(Tl_{1-x-y}Pb_xBi_y)Sr_2Ca_{2+m}O_z$. "A preliminary investigation was carried out to obtain the optimum composition x and y in $(Tl_{1-x-y}Pb_xBi_y)Sr_2Ca_{2+m}O_z$, at which the T_c was the highest" (p. 1281). They found out that the highest T_c was obtained and the phase was stable at nominal composition with x = 0.2 and y = 0.16 when m = 1.

The Role of the Pb in Superconductors

Recently it has been reported that the addition of small quantities of PbO to an alloy of Bi-Ca-Sr-Cu-O aids in the attainment of zero resistance at 105°K following the onset of the drop in the resistivity at 112°K. Prior to this, it had not been possible to obtain zero resistance above 100°K although the onset of a resistive drop was observed at 110°K (Ramesh et al., 1988).

Ramesh et al. (1988) reported, "The microstructure of samples with and without Pb addition has been characterized by transmission electron microscopy. An amorphous Pb-rich grain boundary phase has been found in Pb-doped samples. It is suggested that a Pb-rich liquid phase assists sintering and homogenization of the composition of the superconducting phase to attain the higher zero T_c" (p. 1759).

For Pb-doped sample, preliminary experiments demonstrated that the c spacing was uniform inside the grains, which indicates that there was a good connectivity of the grains. " . . . the Pb-doped samples have undergone liquid phase sintering. It is suggested that the liquid phase enhances transport and hence homogenization of the composition of the superconducting phase, especially near the grain boundaries" (Ramesh & Thomas, 1988, p. 1759).

"Most of the thallium strontium cuprates could not be synthesized until now. The only compounds which were isolated involve thallium monolayers. $TlSr_2CaCu_2O_7$ is obtained as a pure phase with difficulty, whereas the partial replacement of thallium by lead allows the isostructural oxides $Tl_{.5}Pb_{.5}Sr_2CaCu_2O_7$ and $Tl_{.5}Pb_{.5}Sr_2CaCu_3O_9$ to be synthesized" (Raveau et al., 1989, p. 258).

With the substitution of Pb for Tl and Ba for Sr, the Tl-1223 phase can be prepared in the Tl-Pb-Sr-Ca-Cu-O system to exhibit enhanced flux-pinning behavior, using the procedure of partial melting followed by slow cooling. By partially substituting Pb or Bi for Tl, the value of T_c can be further increased by 5-10°K (Kung, Maley, Wahlbeck, & Peterson, 1993).

Sample Preparation

Generally, there are two methods of sample preparation for High-T_c superconductors: the solid state reaction technique and coprecipitation method. The solid state reaction, starting with either the carbonate or oxide form of the individual component, go through few stages of grinding and mixing. It has the advantage of being relatively rapid. But the disadvantage is that the final sample may contain other undesirable phases. The coprecipitation method, starting with the nitrate forms of the constituents, are precipitated out of solution to provide their corresponding carbonate forms. It has an advantage of reliable sample preparation with the desired phase (Doss, 1989).

Kaneko et al. (1990) used the following steps for preparing $Tl_{0.64}Pb_{0.2}Bi_{0.16}Sr_2Ca_2Cu_3O_z$ samples:

> At step 1, appropriate amounts of PbO, Bi_2O_3, $SrCO_3$, $CaCO_3$, and CuO powders were mixed well to obtain a

> uniform mixture of the nominal composition of $Pb_{0.2}Bi_{0.16}Sr_2Ca_2Cu_3O_z$. The mixture was pressed into pellets and fired at 850°C in an O_2 gas flow for 10 h to obtain a precursor. Then, the precursor was pulverized into powder. Tl_2O_3 was added to the powder to make a mixture of the nominal composition of $Tl_{0.64}Pb_{0.2}Bi_{0.16}Sr_2Ca_2Cu_3O_y$. The powder mixture was pressed into pellets. the pellets were wrapped with a Au foil and fired at 920°C in an O_2 gas flow for 20 h(step 2). The weight loss of a pellet during the firing process in step 2 was measured. The ceramic pellets obtained were crushed again and mixed with CaO and CuO powders to yield the nominal composition of $Tl_{0.64-p}Pb0.2Bi_{0.16}Sr_2Ca_3Cu_4O_z$. Here p stands for the amount of Tl lost in step 2. The powder mixture was pressed into pellets and the pellets were wrapped with a Au foil and sintered at 920°C in an O_2 gas flow for 10 h (step3). Finally, the superconducting ceramics obtained were successively annealed at 600°C for 20 h, at 500°C for 20 h, at 400°C for 20 h in an O_2 gas flow (step 4). (p. 1281)

Because Tl_2O_3 is highly volatile at temperatures above its melting point of 717°C under ambient pressure, some researchers suggested using high pressure techniques for suppressing thallium loss during the reaction of Tl-containing compounds. Using a completely sealed reaction system can also effectively prevent the thallium loss. In the preparation of Tl-based superconductors, reaction atmosphere also plays an important role.

New $TlSr_2(Ca,Cr)Cu_2O_7$ superconducting films on MgO subrates have been prepared and characterized by Tang et al. (1992). They prepared the superconducting films via a two-step process. Precursor SrCaCrCuO films were first deposited by spray pyrolysis at temperature of 350-400°C, and thallium was then incorporated by heating the precursor films between unfired TlSrCaCuO bulk pellets at 880-900°C in an oxygen atmosphere. The films were then cooled down to room temperature in an oxygen or argon atmosphere (Tang, Sheng, Chen, Li, & Peterson, 1992).

Characterization of Superconductors

To determine the crystalline quality (Qiu & Shih, 1988) or to identify the phase present in the samples and determine the lattice parameters (Kaneko et al., 1990), the x-ray diffraction method would be used. By examining the powder X-ray diffraction pattern, the impurity phase in the sample can be identified (Nuffield, 1966 & Ladd & Palmer, 1977).

The electrical resistivity of the samples will be measured by a standard four-probe method (Kaneko et al., 1990). Very small resistances are difficult to measure. However, one can determine if the resistance is below a certain level. Four-terminal measurements of resistivity are very useful for the determination of T_C in superconducting samples (Doss, 1989). A constant current will be applied across the outer probes and the voltage will be measured across the inner probes. The resistivity of the sample is proportional to the ratio of the voltage to the current.

The measurements of magnetic susceptibility are very significant for determining whether a material is actually superconducting. A determination of magnetic susceptibility is also indicative of the percentage of the volume of the sample that is superconducting. Measurement of dc resistivity can not provide such an indication. The methods for determination of susceptibility may also be categorized as dc and ac measurements (Doss, 1989).

CHAPTER III

METHODOLOGY

Test Procedures

To fulfill the research objectives, the whole experiment was divided into the three phases. The experiment was based on the following procedures.

1. Optimizing the sample fabrication process by determining material sintering temperature in the furnace and sintering time in the air inside the Pt crucible. Compound $(Tl_{0.5}Pb_{0.5})Sr_2Ca_2Cu_3O_x$ and $(Tl_{0.5}Pb_{0.5})Ba_{0.7}Sr_{1.25}Ca_2Cu_3O_x$ were used for optimizing process. Based on past experiment and researches, temperature range would be between 850°C and 950°C and sintering time range would be between 6.5 min and 12 min. The best temperature and time were determined for fabricating good superconductive material by experimenting different temperatures and sintering times.

2. Set Tl/Pb = 1, and determine the optimum y in $(Tl_{0.5}Pb_{0.5})Sr_{2-y}Ba_yCa_2Cu_3O_z$ by trying different y, starting from y = 0.1 to 1.0.

C: Using optimum y, which was determined in procedure 1, to determine the effect of Eu doping in Tl, Pb sites by experimenting different doping concentrations for Eu, starting from 1% to 10%.

Sample Preparation Procedures

Sr-Ba-Ca-Cu-O were synthesized first, starting from high purity (99.999%) BaO, CaO, and CuO compounds, and 99% pure

SrO. The samples were prepared as follows: appropriate molar portions of SrO, BaO, CaO and CuO are mixed and ground thoroughly. The mixed compound is then heat treated at 900°C for 24 hours, cooled, ground into a fine powder, and then the same process was repeated to obtain a homogeneous compound.

The second step was to add the appropriate amount of PbO and Tl_2O_3 to form the superconducting compound. The resulting compound was mixed thoroughly using a mortar and pestle. Pellets of approximately 250mg-750mg were prepared by pressing the material in a die using a load exceeding 15,000PSI. The pellet was heat treated at a high temperature of 870°C for 10 minutes and then rapidly air-cooled. The starting composition might have to be adjusted with an excess amount of PbO, and Tl_2O_3 to compensate for the losses during the heat treatments of the samples since PbO and Tl_2O_3 are high volatile materials. Based on past experiment the appropriate compensate amount would be 10% which required 110% of calculated amount of PbO and Tl_2O_3. To dope the samples with Eu, appropriate molar portions of Tl_2O_3, PbO and Eu_2O_3 were mixed to the sintered SrCaBaCuO mixture.

Sample Measurement and Analysis Procedures

After each sample is heat treated, the pellets were characterized by x-ray diffraction (XRD) analysis and magnetic susceptibility measurements using an ac susceptometer. AC susceptibility measurements were performed using a Lakeshore Model 7000 Susceptometer available at Dr.

Paul Shand's lab in the Physics Department, at the University of Northern Iowa. The XRD spectrum is useful for identifying the impurity phases present, if any, in each sample. The ac susceptibility measurements performed at different temperatures gives information on the diamagnetic susceptibility of the superconducting state, and the superconducting volume fraction (Vsc) of the sample. The results for each sample were examined carefully to identify the optimum processing temperature and time and the partial pressure of Tl.

The major part of sample analysis was to study the structural and electrical properties of the $(Tl_{0.5-x}Pb_{0.5-x}Eu_x)Sr_{2-y}Ba_yCa_2Cu_3O_z$ superconducting samples. The objectives were to obtain a single phase superconductor with high Tc (> 115°K), and high volume fraction of superconductivity.

X-Ray Diffraction Analysis

X-Ray Diffraction analysis is a very important tool in the study of superconductivity. The microstructure of a material determines its properties and behavior. Particularly for Tl-based superconductivity, it is even more significant because Tl-based superconductors have a feature of multi-phases, like Tl-1223, Tl-1212 and Tl-1101 phases. Hence, it is very important to understand the microstructure of such Tl-based superconductors, and identify its phases.

High temperature superconductors are typical crystalline materials. In order to understand the properties of superconducting material, it is important to analyze the crystal microstructure. A molecule has structural regularity because covalent bonds maintain a specific number and orientation of neighbors for each atom. A crystalline material has a repeating pattern in three dimensions. Atomic packing may take on one of seven main crystal patterns. Seven major crystal system include: cubic, tetragonal, orthorhombic, monoclinic, triclinic, hexagonal and rhombohedral. Based on past experience, it was found out that Tl-based superconductor have tetragonal crystal pattern which have two equal axial dimension ($a1 = a2$) but an unequal third axial dimension ($a1 = a2 \neq c$).

Tl-based superconductors is a mutli-phase superconducting compound. Each phase has different features in its crystal structure. It is very significant to identify each individual phase and their contribution to the superconducting properties for a material. For a crystal structure, atoms are closely packed in the way that the space can be divided into equal volumes by intersecting plane surfaces. "The simplest and most regular of these involves three mutually perpendicular sets of equally spaced, parallel planes aligned to give a series of cubes" (Vlack, 1964, p. 56).

In order to understand a superconducting material properties and crystal structures it would also be necessary to recognize the crystal directions. One single crystal cell could be used as a basic unit. Vlack (1964) summarized the following features for using crystal direction:

(1) Unit-cell intercepts, and not actual distances, are used.

(2) Crystal axes are used as base directions.

(3) A [222] direction is identical to a [111] direction.

(4) Directions such as [112] may also be identified.

Another important characteristic of crystal structure is crystal planes, which affect the properties and behavior of crystals. A crystal plane with specific direction in the crystal structure covers certain number of atoms on that plane. Identifying the various atomic planes for Tl-superconductor structure is very important for defining the superconductive phase. Every boundary face of a crystal cell is a crystal plane. Other planes would section the crystal unit in a specific crystal direction. Therefore, a crystal plane contains both direction and quantity which would be labeled by (hkl) symbols. (hkl) symbols are called Miller indices. One of the most important features of structural study is interplanar spacings d_{hkl} which identifies the distance between two adjacent parallel planes.

In this research, x-ray analysis was used for determining the superconductor's phases. Each of the

TlPbSrCaCuO superconductor phase has different unit cells, especially the c-axis lattice spacing. When x-rays are directed at a superconducting material they are diffracted by the various crystal planes. The diffraction angle depends on the wavelength of the x-rays used and the interplanar spacing of the material tested. Van Vlack (1964) made following conclusions. Consider the parallel crystal planes of the atoms shown in Figure 2, from which a wave is diffracted.

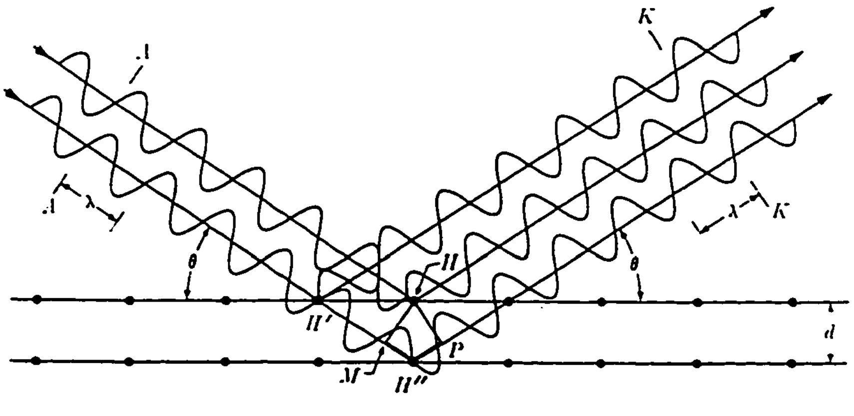

Figure 2. X-Ray Diffraction. From "Elements of Materials Science" by L. H. Van Vlack, 1964.

The waves might be reflected from an atom at H or H', and remain in phase at K. However, x-rays were not only reflected from the surface plane but also from subsurface planes as well. If these reflections were to remain in phase, the distance M'H"P must equal one or more integral wavelengths of the x-rays used. Therefore, from geometry,

$$n\lambda = 2d\sin\theta,$$

where λ is the wavelength, d is the interplanar spacing and θ is the angle of incidence. The value n is the number of waves which occur in the distance MH"P, typically n=1. The reflections are generally weaker when more than one extra wave is present.

For this research, the important factor was to identify the phase of Tl-superconductor. Based on past research and experiments, the lattice constant c for the different phase were determined, ie. c = 15.5A for Tl-1223, c = 12.05A for Tl-1212 and c = 9.01A for Tl-1101. Therefore the interplanar spacings for three different phases are calculated by $d_{hkl} = c/\sqrt{h^2+k^2+l^2}$, where h, k, and l are the indices of the planes. The wavelength of x-ray used for this research was λ = 1.54A (Cu K_α radiation). So the θ will be calculated by $\theta = \sin^{-1}(n\lambda/2d)$ for each specific phase and plane (indentified by Miller indices h, k, and l). The spectra obtained from X-ray diffraction measurements is the intensity vs the Bragg Angle (2θ). From the spectra it is possible for researcher to mark the peak (high density) and match peak point with 2θ. All the main peaks from the XRD spectra were identified. Then it was easier to identify the Tl-phase by looking at XRD spectrum and determine if the sample has only one phase or which phase was dominating in the prepared

Table 5

XRD Peaks for Various Phases

MILLER INDICES			2 THETA	2 THETA	2 THETA	SQRT	1223	1212	1101
h	k	l	c = 15.5 A (1223)	c = 12.05 A (1212)	c = 9.01 A (1101)	h*h+k*k+l*l	d (spacing)	d (spacing)	d (spacing)
0	0	1	5.69	7.33	9.81	1.0000	15.5000	12.0500	9.0100
0	0	2	11.40	14.69	19.68	2.0000	7.7500	6.0250	4.5050
0	0	3	17.14	22.10	29.71	3.0000	5.1667	4.0167	3.0033
0	0	4	22.92	29.62	39.98	4.0000	3.8750	3.0125	2.2525
0	0	5	28.76	37.27	50.59	5.0000	3.1000	2.4100	1.8020
0	0	6	34.68	45.09	61.70	6.0000	2.5833	2.0083	1.5017
0	0	7	40.70	53.14	73.49	7.0000	2.2143	1.7214	1.2871
0	0	8	48.83	61.49	86.26	8.0000	1.9375	1.5063	1.1283
0	0	9	53.12	70.21	100.55	9.0000	1.7222	1.3389	1.0011
0	0	10	59.57	79.44	117.43	10.0000	1.5500	1.2050	.9010
1	0	1	8.06	10.37	13.88	1.4142	10.9602	8.5206	6.3710
1	0	2	12.76	16.43	22.03	2.2361	6.9318	5.3889	4.0294
1	0	3	18.08	23.32	31.36	3.1823	4.9015	3.8105	2.8492
1	0	4	23.64	30.55	41.26	4.1231	3.7593	2.9226	2.1652
1	0	5	29.35	38.03	51.67	5.0990	3.0398	2.3632	1.7670
1	0	6	35.18	45.75	62.64	6.0828	2.5482	1.9810	1.4612
1	0	7	41.13	53.72	74.36	7.0711	2.1920	1.7041	1.2742
1	0	8	47.22	62.02	67.10	8.0623	1.9225	1.4946	1.1176
1	0	9	53.47	70.71	101.41	9.0554	1.7117	1.3307	.9950
1	0	10	59.90	79.91	118.38	10.0499	1.5423	1.1990	.8965
1	1	1	9.87	12.71	17.02	1.7321	8.9489	6.9571	5.2019
1	1	2	13.98	18.01	24.17	2.4495	6.3278	4.9194	3.8783
1	1	3	18.97	24.47	32.93	3.3166	4.6734	3.6332	2.7166
1	1	4	24.33	31.46	42.52	4.2426	3.6534	2.8402	2.1237
1	1	5	29.92	38.78	52.73	5.1962	2.9830	2.3190	1.7340
1	1	6	35.66	46.40	63.58	6.1644	2.5144	1.9548	1.4616
1	1	7	41.56	54.30	75.22	7.1414	2.1704	1.6873	1.2617
1	1	8	47.60	62.55	87.94	8.1240	1.9079	1.4833	1.1091
1	1	9	53.82	71.21	102.26	9.1104	1.7013	1.3227	.9890
1	1	10	60.23	80.39	119.34	10.0995	1.5347	1.1931	.8921

Note. d = c/SQRT(h*h+k*k+l*l); n*W.L. = 2*d*sin(THETA) (n = 2); W.L. = 1.54.

material. The calculation results for different phases are shown in the Table 4.

Magnetic Susceptibility Analysis

Magnetic susceptibility, χ is a measure of how "magnetic" a material is and represents the response of the material to an applied field H: $\chi = M/H$ or $\chi_V = m/HV$, where m is the magnetic moment, V is the sample volume, and M is the volume magnetization defined by $M = m/V$ and H is magnetic field. Magnetic susceptibility measurement is another important activity for any superconductivity study which is a criterion for determining whether a material is actually superconducting or not. It could also determine the percent volume fraction of a sample that is superconducting. Measurements of magnetic susceptibility gives a better and more accurate judgement in determine the superconductivity of a superconducting material than resistivity measurement.

There are many different methods for the determination of susceptibility, which may mainly be categorized as dc and ac susceptibility measurements. A basic ac susceptometer technique which is used in such measurements is illustrated in Figure 3. A primary coil creates a magnetic field using an ac current. Two secondary coils are wound in opposition so that the induced fields are opposing, resulting in zero voltage across the coils when no sample existed. When a sample is positioned into one coil, there would be a susceptibility signal with unbalanced voltage between two coils. A lock-in amplifier is used to detect very low level ac signals.

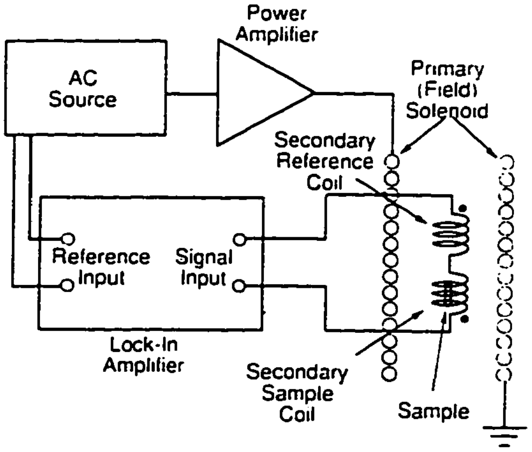

Figure 3. A Basic AC Susceptometer Technique From "Engineer's Guide to Superconductivity-- Fundamentals and Applications" by J. D. Doss, 1989.

The lock-in amplifier would not sense any signal if the sample susceptibility was zero. If the temperature is decreased to the critical level T_C, a superconducting sample would begin to exhibit the Meissner effect and expel magnetic flux. This expulsion of flux would unbalance the coupled coils so that a voltage appears at the lock-in amplifier (Doss, 1989). "The phase will be 90° shifted from the drive voltage, with the direction being dependent on whether the sample was paramagnetic ($\chi \rightarrow$ positive) or, in the case of a superconductor, diamagnetic ($\chi \rightarrow$ negative)" (Doss, 1989, p.308). As sample magnetization *M* increases, the magnitude of the voltage sensed at the lock-in amplifier would then increase. The volume fraction of sample also affects the voltage signal obtained (Doss, 1989).

AC Susceptibility Measurement--LakeShore 7000

A LakeShore 7000 system was used for measuring magnetic susceptibility in this research. The LakeShore 7000 is an AC susceptibility measurement system. In an AC magnetization measurement the applied field has an AC term with the form

$$H_{ac} = h_o\cos(\omega t)$$

The resulting sample magnetization due to this AC term is given by

$$M_{ac} = m_o\cos(\omega t-\theta)$$

where θ is the phase difference between the drive field and the magnetization. The expression

$$M_{ac} = \chi' h_o\cos(\omega t) + \chi'' h_o\sin(\omega t)$$

where $\chi' = m_o\cos\theta/h_o$ is the real or in-phase susceptibility component, and $\chi'' = m_o\sin\theta/h_o$ is the imaginary or out-of-phase susceptibility component. These combine to form the complex susceptibility: $\chi_{ac} = \chi' - i\chi''$.

χ'' is a measure of dissipative losses occurring in the magnetic system under study. In other words, it is a measure of the energy absorbed by the sample from the applied AC field. If there are any time-dependency or relaxation processes occurring in the system, the induced magnetic moment cannot follow the field instantaneously. This leads to non-zero θ and hence a non-zero χ''. χ'' is also non-zero if there are any non-linearities or hysteresis in the DC magnetic response of the material under study.

The development of high T_c superconductors has promoted the use of AC susceptibility technique. High T_c materials are characterized by relatively small first critical fields, H_{c1}, and a small full penetration field, H_p. Therefore, a reliable low-field magnetization measurement technique is necessary for the full magnetic characterization of such materials. AC susceptibility fulfills this important intrinsic requirement. The AC measurement is used to differentiate between inter- and intragranular current coupling, and in determining the overall quality of a superconductor. An analysis of χ'' can provide information about the critical current density J_c of these materials via the invocation of a suitable critical state model, and this sort of analysis has contributed to a better understanding of the mechanisms of superconductivity in these compounds.

In the series 7000 system, an alternating magnetic field is applied to a sample by means of an AC current through a primary coil. Two identical secondary coils connected in series opposition (constructed inside the primary coil) are used to detect variations in flux created by the sample when it is positioned within one of the secondary coils. The resultant signal is measured by a phase-sensitive detector which produces an output signal proportional to the AC susceptibility of the material. The AC current source used to drive the primary coil is a variable amplitude and frequency current source, thus permitting measurements of the AC susceptibility over a broad range of discrete field amplitudes and frequencies. Precision thermometry and heaters, interfaced to a Lake Shore temperature controller and data acquisition and control software permit the automatic measurement of susceptibility from 4.2°K to 325°K.

The two secondary coils are wounded in opposition in order to cancel voltages induced either from unwanted external sources or by the AC field itself. The sensitivity of the measurement electronic system in the Series 7000 exceeds the capability for identically winding two secondary coils. Hence, even in the absence of a sample, there is a small offset voltage present. For measurements requiring very high accuracy, the data acquisition and control software provides for precisely and automatically moving the sample between the centers of the two secondary coils during a measurement sequence. This effectively cancels any effects due to this offset voltage, and also presents a means whereby a DC magnetization measurement can be performed with this apparatus.

The 7000 Series Susceptometer System

The 7000 Series AC Susceptometers (see Figure 4) are built into one single console or enclosure. Measurement of the real susceptibility χ' and imaginary χ'' as a function of temperature, amplitude and frequency of an applied AC field are possible. The system also provides a simple and accurate AC susceptibility measurement by the installation cryogenic

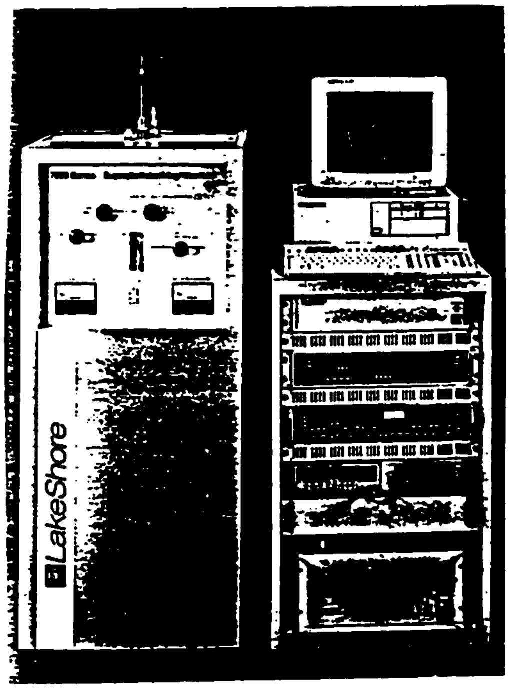

Figure 4. The Lakeshore Susceptometer.

and electronic instrumentation, and data acquisition and control software. Figure 5 shows a diagram of the sample probe used in this study (LakeShore Measurement and Control Technologies, 1992). System operation includes the following steps:

1. Vacuum pump startup.

2. Initial pump out, to remove any accumulated gases in the system.

3. Liquid nitrogen filling. Warm transfer or cold transfer.

4. Sample mounting. A sample is attached to the sample probe. Place the sample inside the sample holder and screw on the sample lid.

5. Sample probe loading and unloading. Contamination or condensation from air must be avoided. In order to avoid the contamination and move a sample into and out of a temperature controlled region, a load lock arrangement is used.

6. Setup Operating parameters--Sample space and vacuum isolation space. The sample space is designed to run with ≈500 to 1000 microns of helium exchange gas present. This assures good thermal contact between the sample, temperature sensors, and control heaters. It is critical that an exchange gas be present during operation. During any automatic operation, the vacuum isolation space should be pumped continuously.

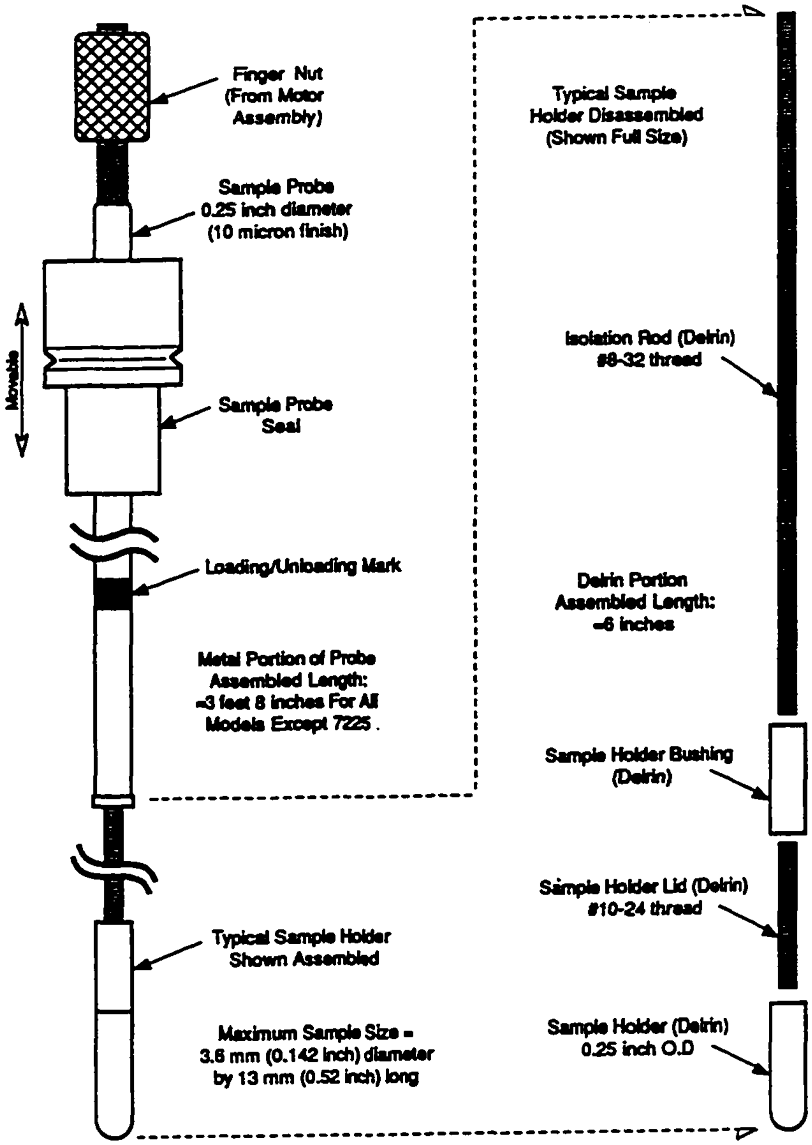

Figure 5. Lakeshore Susceptometer Sample Probe. From "User's Manual--7000 Series AC Susceptometer/DC Magnetometer" by LakeShore Measurement and Control Technologies 1992).

When changing samples and pumping out the load lock, the vacuum space valve should be closed to avoid any contamination.

7. AC susceptibility measurements, ACS7000 software operation for sample measurement includes following steps:

a. Temperature control;

b. Sample positioning-- auto-positioning of samples;

c. Adjustment of parameters.

d. Addenda data;

e. Field control.

f. Data processing ACS (LakeShore Measurement and Control Technologies, 1992).

In AC susceptibility measurements for this research, 133 Hz frequency was selected for the applied AC field. The amplitude of the AC magnetic field was set at 1 or 10 oe. Temperature range was set between 78°K-125°K. Temp Spec was set up as: 78°K-90°K with 1.5°K increment, 91°K-115°K with 1.0°K increment and 117°K-125°K with 2.0°K increment. Because superconducting transitional range is between 91°K-115°K, more data points are required at this temperature range. Liquid Nitrogen which could cool the sample down to ~78°k was used for the cooling system. Auto-sample positioning method was selected for positioning sample. Mass susceptibility was selected for calculation. Waiting times for stabilizing the temperature were set as < 100°K → 200 seconds and > 100°K → 300 seconds. CGS units were selected for magnetic field in oe.

CHAPTER IV

RESULTS AND ANALYSIS OF DATA

Optimizing Sample Fabrication Process

Precursor Preparation Process

For Ba doping in the $(Tl_{0.5}Pb_{0.5})Sr_{2-x}Ba_xCa_2Cu_3O_y$ superconductor, the following procedure was used. The precursor compound of $Sr_{2-x}Ba_xCa_2Cu_3O_y$ was prepared first, starting from 99.9% pure BaO_2, CaO, CuO and SrO compounds.

Precursor calculation was performed as follows:

$(x)BaO_2 + (2-x)SrO + 2CaO + 3Cuo \Rightarrow Sr_{2-x}Ba_xCa_2Cu_3O_y$

$(x)*(137.34+2*16) + (2-x)*(87.62+16) + 2*(40.08+16) + 3*(63.54+16) = \sum M$ gm

$\sum M$ is total precursor atomic weight.

The calculations for the amount of BaO_2, SrO, CaO and CuO needed for 1 gm of precursor $Sr_{2-x}Ba_xCa_2Cu_3O_y$:

$BaO_2 \rightarrow (x)*(137.34+2*16)/\sum M$ gm;

$SrO \rightarrow (2-x)*(87.62+16)/\sum M$ gm;

$CaO \rightarrow 2*(40.08+16)/\sum M$ gm;

$CuO \rightarrow 3*(63.54+16)/\sum M$ gm;

Table 5 shows the results of BaO_2, SrO, Cao, and CuO for precursor preparation.

Table 5

Calculations for Precursor with Ba Doping (0 - 1.0)

(Proportional Amount for 2 gm Precursor)

Ba Concentration	BaO (gm)	SrO (gm)	CaO (gm)	CuO (gm)
0	0	0.7428	0.402	0.8552
0.1	0.03	0.3487	0.1987	0.4226
0.2	0.1186	0.6532	0.3928	0.8356
0.3	0.1758	0.6098	0.3882	0.826
0.4	0.2318	0.5674	0.384	0.8168
0.5	0.2866	0.526	0.3796	0.8076
0.6	0.3402	0.4856	0.3754	0.7988
0.7	0.3924	0.446	0.3714	0.7902
0.75	0.4182	0.4266	0.3694	0.7858
0.8	0.4438	0.4072	0.3674	0.7816
0.9	0.4938	0.3694	0.3634	0.7732
1	0.543	0.3322	0.3596	0.7652

Each sample was prepared according to the following steps:

1. In separate crucibles, took approximately 2 gm of each of these materials, BaO_2, SrO, CaO and CuO; Followed by heat treatment at about 200°C for 10 minutes which would allow the sample to get rid of any moisture content. Samples were, then, cooled down to room temperature and the appropriate amount of sample was measured.

2. SrO powder was measured first; Ground it to a fine powder at least for 15-20 minutes using pestle and mortar.

3. Added appropriate amount of BaO_2 powder to the SrO powder; mixed and ground for at least for 15-20 minutes.

4. Weighed appropriate amounts of CuO powder and added into SrBaO compound; Mixed and ground for 15 minutes.

5. Added appropriate amount of CaO to the above mixture and mixed for about 10-15 minutes.

6. Heat treat the $Sr_{2-x}Ba_xCa_2Cu_3O_y$ compound at 900°C for 24 hours; Cooled down for 20-30 minutes.

7. Ground and mixed the heat treated compound.

8. Re-heat treated at 900°C for another 24 hours; Cooled down and ground the compound to a fine mixture.

The purpose of double heat treatment was to thoroughly homogenize the precursor. After precursors were prepared, they were ready for the preparation of the final samples.

Final Sample Preparation and Sintering Process

Ba doping only: $(Tl_{0.5}Pb_{0.5})Sr_{(2-x)}Ba_xCa_2Cu_3O_y$ was prepared by adding Tl_2O_3 and PbO to the precursors. Because the Tl_2O_3 and PbO are highly volatile materials, no preheat treatment was necessary for Tl_2O_3 and PbO before being mixed. When calculations were performed, 10% differential compensation was taken into account for the Tl_2O_3 and PbO, because of their volatility.

$$(1/4)Tl_2O_3 + (1/2)PbO + Sr_{(2-x)}Ba_xCa_2Cu_3O_y \Rightarrow (Tl_{0.5}Pb_{0.5})Sr_{(2-x)}Ba_xCa_2Cu_3O_y$$

$$(1/4)(2*204.37+3*16) + (1/2)(207.19+16.0) + (\text{precursor}) = \sum Mt$$

$\sum Mt$ is total final sample atomic weight.

To make 1 gm of $(Tl_{0.5}Pb_{0.5})Sr_{(2-x)}Ba_xCa_2Cu_3O_y$, it was needed as follows:

Tl_2O_3 → $(1/4)(2*204.37+3*16)/\sum Mt$;

PbO → $(1/2)(207.19+16.0)/\sum Mt$;

Precursor → $(\text{precursor})/\sum Mt$.

After the adjustment with 10% of Tl_2O_3 and PbO, the calculation results for 0.500gm final samples are shown in Table 6.

Table 6

TlO and PbO Calculation for Final Compound

Ba Concentration	TlO (gm)	PbO (gm)	Precursor
0	0.0802	0.0783	0.356
0.1	0.0795	0.0777	0.3572
0.2	0.0788	0.077	0.3584
0.3	0.0782	0.0764	0.3595
0.4	0.0775	0.0758	0.3606
0.5	0.0769	0.0752	0.3618
0.6	0.0763	0.0746	0.3629
0.7	0.0757	0.074	0.364
0.75	0.0754	0.0737	0.3645
0.8	0.0751	0.0734	0.3651
0.9	0.0745	0.0728	0.3661
1	0.0739	0.0723	0.3671

Following procedures were followed:

1. Heat the precursor for 15-20 minutes at 200°C in order to get rid of moisture from precursor if the precursor had been kept for long time.

2. Measured appropriate amount of precursors and grounded for 15 to 20 minutes to get good mixtures using pestle and mortar.

3. Measured appropriate amount of PbO and added into the precursor. Ground and mixed for about 15 minutes.

4. Measured appropriate amount of Tl_2O_3 and added into the above mixture. Ground and mixed for about 15 minutes.

5. Put the well mixed sample into a specially designed die and plug the plunger into the die. The samples were pressed inside the plug using a hydraulic press, with an applied force of 15,000psi for about 10-15 minutes. Thus the sample pellets were made and they were ready for the final sample sintering process.

Ba doping and Eu doping in Tl, Pb site: $(Tl_{0.5-x}Pb_{0.5-x}Eu_{2x})Sr_{(2-y)}Ba_yCa_2Cu_3O_z$ was prepared by adding Tl_2O_3, PbO and Eu_2O_3 into the precursors. There was no preheat treatment for Tl_2O_3, PbO and Eu_2O_3 before mixing. When calculations were made 10% adjustments were made for Tl_2O_3 and PbO. Eu_2O_3 is not a highly volatile material. After first-phase experiment and sample measurements, it was determined that Ba = 0.75 gave the optimum results. Therefore Ba = 0.75 was used for the calculation of Eu doping. $(Tl_{0.5-x}Pb_{0.5-x}Eu_{2x})Sr_{(2-y)}Ba_yCa_2Cu_3O_z$ becomes $(Tl_{0.5-x}Pb_{0.5-x}Eu_{2x})Sr_{1.25}Ba_{0.75}Ca_2Cu_3O_z$.

$[(0.5-x)/2]Tl_2O_3 + (0.5-x)PbO + xEu_2O_3 + Sr_{1.25}Ba_{.75}Ca_2Cu_3O_z$
$\Rightarrow (Tl_{0.5-x}Pb_{0.5-x}Eu_{2x})Sr_{1.25}Ba_{0.75}Ca_2Cu_3O_z$;

$[(0.5-x)/2](2*204.37+3*16)$ + $(0.5-x)(207.19+16.0)$ +

$x(2*151.96+3*16)$ + (precursor) = $\sum Mt$;

To make 1 gm of $(Tl_{0.5}Pb_{0.5})Sr_{(2-x)}Ba_xCa_2Cu_3O_y$, it was needed as follows:

Tl_2O_3 → $[(0.5-x)/2](2*204.37+3*16)/\sum Mt$;

PbO → $(0.5-x)(207.19+16.0)/\sum Mt$;

Eu_2O_3 → $x(2*151.96+3*16)/\sum Mt$;

Precursor → (precursor)$/\sum Mt$.

Because a very small amount of Eu was doped in Tl and Pb sites, the Eu concentration was varied from 0.00 to 0.1 with increment of 0.02. After the adjustment with 10% of Tl_2O_3 and PbO, the calculation results for 0.2500gm final samples are shown in Table 7.

Table 7

Calculations Results for 0.2500gm Final Samples

Eu Concentration	TlO (gm)	PbO (gm)	EuO (gm)	Precursor
0	0.0375	0.0369	0	0.1823
0.02	0.037	0.0361	0.0011	0.1825
0.04	0.0363	0.0354	0.0021	0.1827
0.06	0.0356	0.0348	0.0032	0.1829
0.08	0.0348	0.034	0.0042	0.1831
0.1	0.0341	0.0334	0.0053	0.1833

The following procedures were followed:

1. Heat the precursor for 15-20 minutes at 200°C in order to get rid of moisture from precursor if the precursor had been kept for long time.

2. Measured appropriate amount of precursors and ground for 15 to 20 minutes to get a good mixture.

3. Measured appropriate amount of PbO and added into the precursor. Ground and mixed for about 15 minutes.

4. Measured appropriate amount of Tl_2O_3 and added into the above mixture. Ground and mixed for about 15 minutes.

5. Measured appropriate amount of Eu_2O_3 and added into the above mixture. Ground and mixed for about 15 minutes.

6. Put the well mixed sample into the die and plug the plunger into the die. The sample was applied a pressure of 15000psi for about 10-15 minutes to make the pallet. Thus the sample pallets were made and were ready for final sample sintering process.

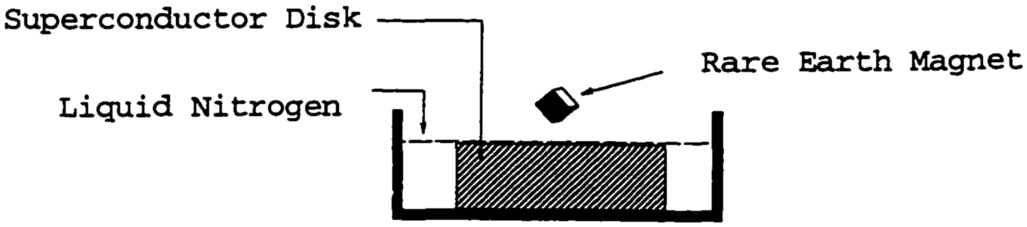

Figure 6. Superconductivity Demo. From "Instruction Manual for Superconductor Demonstrations" by Colorado Superconductor Inc, 1992.

The High Temperature Superconductor Demonstration Kits made by Colorado Superconductor, Inc., was used to judge if the sample was superconducting. High Temperature Superconductor Demonstration Kits consist of a simple magnetic detector based on Meissner Effect (see Figure 6). The Kit is a simple and complete package. Superconducting researchers can investigate the basic phenomenon of superconductivity with little effort and time consumption. Liquid nitrogen would be necessary to cool the sample.

A small amount of liquid nitrogen was poured into the dish or styrofoam cup until the liquid is about a quarter of an inch deep. The liquid boiled furiously for a few seconds. Waited until it stopped boiling, a teflon tweezer was used to place the superconducting sample piece in the liquid nitrogen. The nitrogen boiled around the disk for a while and stopped. The teflon tweezer was used again to pick up the provided magnet (smaller one would be used due to the sample size limitation), and attempted to balance it on the top of the superconductor disk. If the sample is superconducting, the magnet would simply float away a few millimeters from the sample surface, in another words the magnet would be repelled by the superconductor which would indicate that the sample is superconducting. If the sample is not superconducting, the magnet would actually touch against the surface of the sample. This is a typical demonstration of the Meissner Effect.

As known, superconductors are strongly diamagnetic materials. At temperatures below its Critical temperature, T_C, a superconducting sample would prevent any magnetic field from entering it. Microscopic magnetic dipoles induced in the superconducting sample oppose the applied field. This induced field then repels the source of the applied field, and would consequently repel the magnet providing that field. This is why when a magnet was placed close to the superconducting sample at a temperature below T_C, it would be repelled and kept a little distance from the superconducting sample. If the researcher wishes to place magnet on the top of sample, the force of repulsion must exceed the magnet's weight to keep magnet floating. This phenomena will occur only if the strength of the applied magnetic field does not exceed the value of the critical magnetic field, H_C for that superconductor material. Therefore the researcher could easily and quickly identify if a sample is superconducting. This is a very efficient way for primary or trial studies of a material compared to expensive AC susceptometer, even though it is not an accurate measure.

The predicted T_C of superconductivity for this research was found to be above 115°K. Therefore, superconducting samples with T_C above 90°K should be expected and detected by this simple device. This would be the judgement or criteria of superconductivity for optimizing the final superconducting sample process.

The sintering furnace and fume extractor conditions, sinter temperature, and sintering time are critical factors that affect superconducting sample fabricating process. In this research, furnace and fume hood were made to be fixed variables. Only the sintering temperature and sintering time were considered as independent variables.

Based on past experiments and other similar researches, the sintering temperature was narrowed down to the range from 850°C to 900°C and sintering time from 6 minutes to 13 minutes. Samples of $(Tl_{0.5}Pb_{0.5})Sr_2Ca_2Cu_3O_x$ and $(Tl_{0.5}Pb_{0.5})Ba_{0.7}Sr_{1.25}Ca_2Cu_3O_x$ were experimented for these conditions. Samples with each combination of temperature and time were tested for Meissner effect using High Temperature Superconductor Demonstration Kit as described above. From those test and results, it was concluded that best superconducting condition in this particular experiment environment is the sintering temperature 880°C and sintering time 10 minutes. This was also verified with XRD and ac susceptibility measurements.

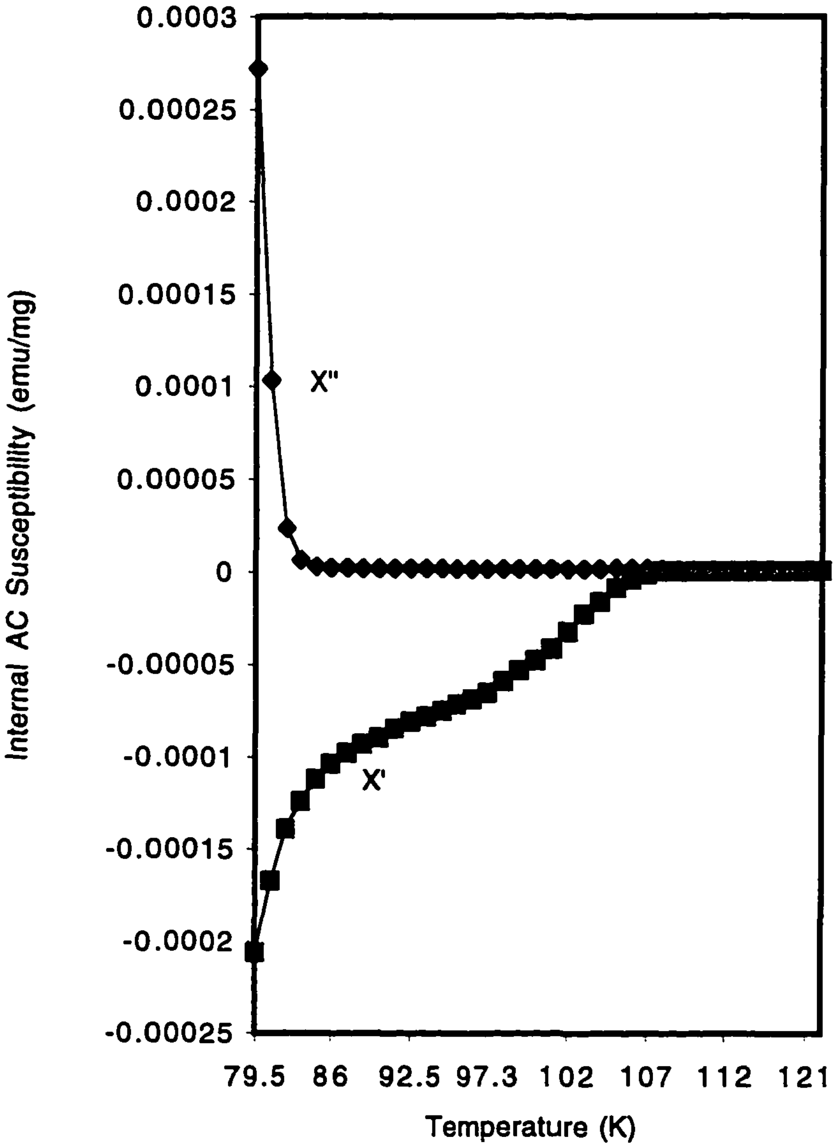

Figure 7. (TlPbSrCaCu) Doping Ba = 0

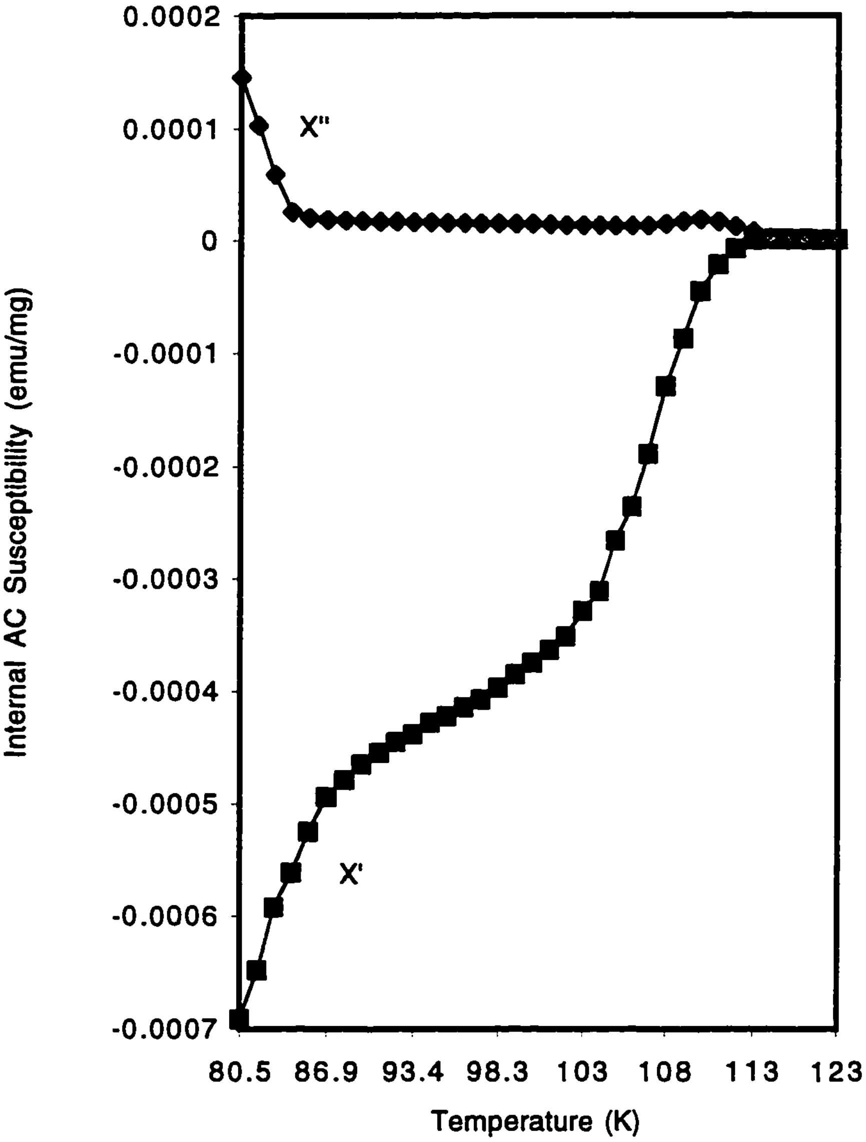

Figure 8. (TlPbSrCaCu) Doping Ba = 0.1

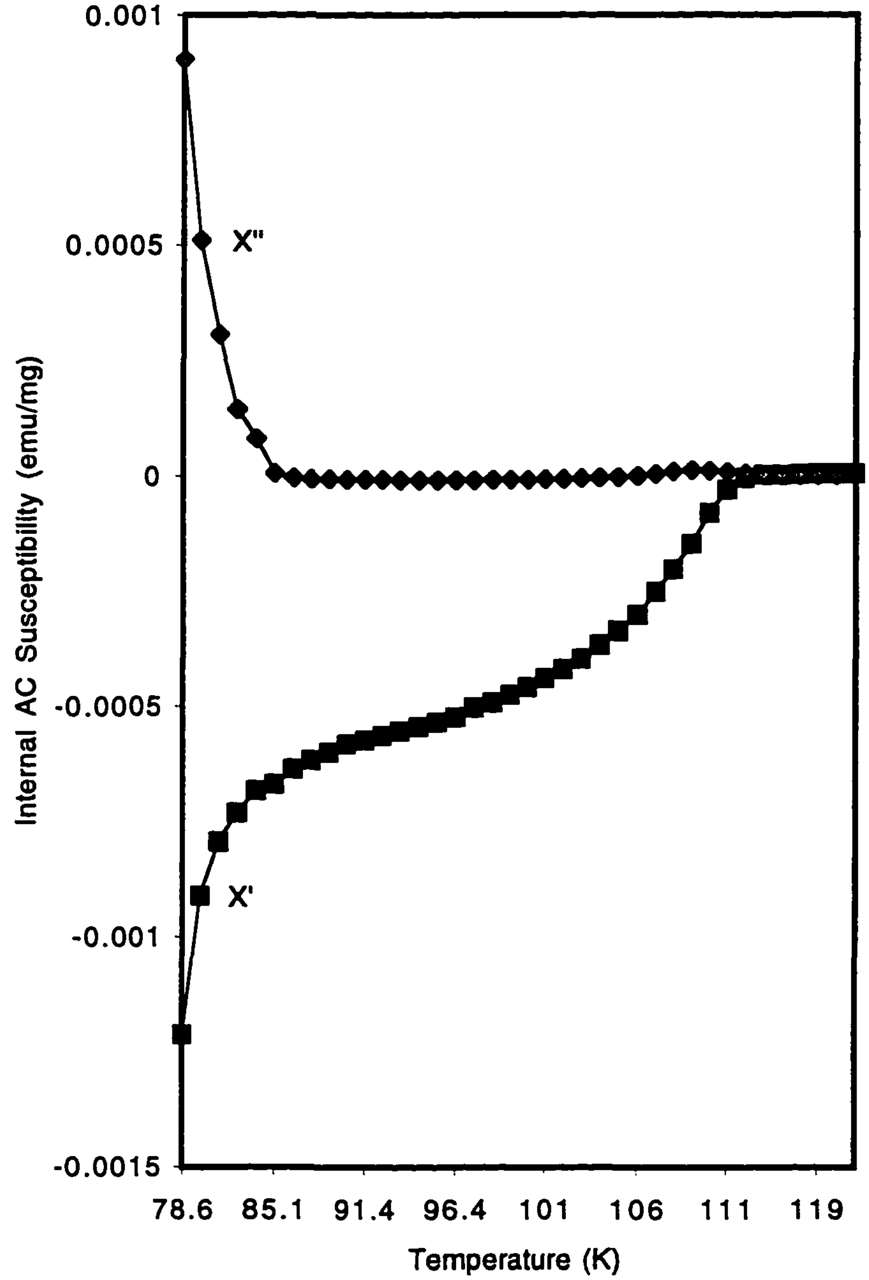

Figure 9. (TlPbSrCaCu) Doping Ba = 0.2

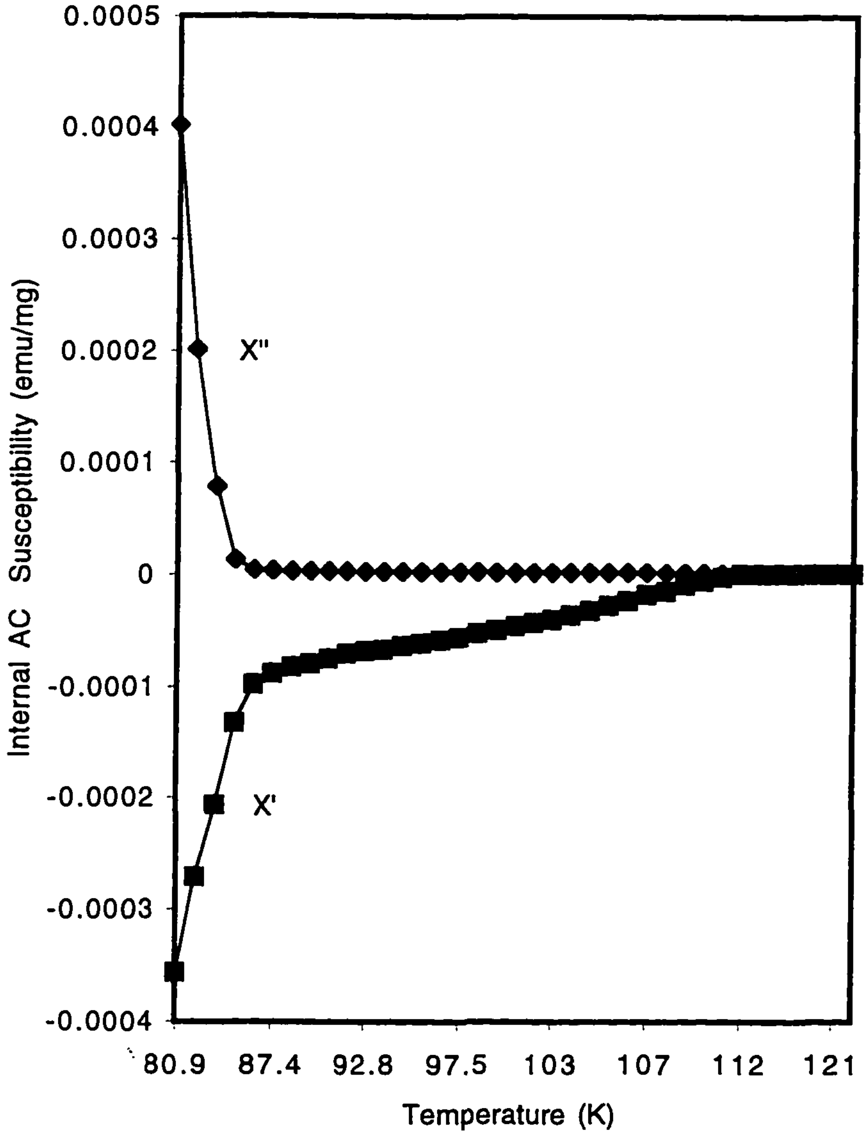

Figure 10. (TlPbSrCaCu) Doping Ba = 0.3

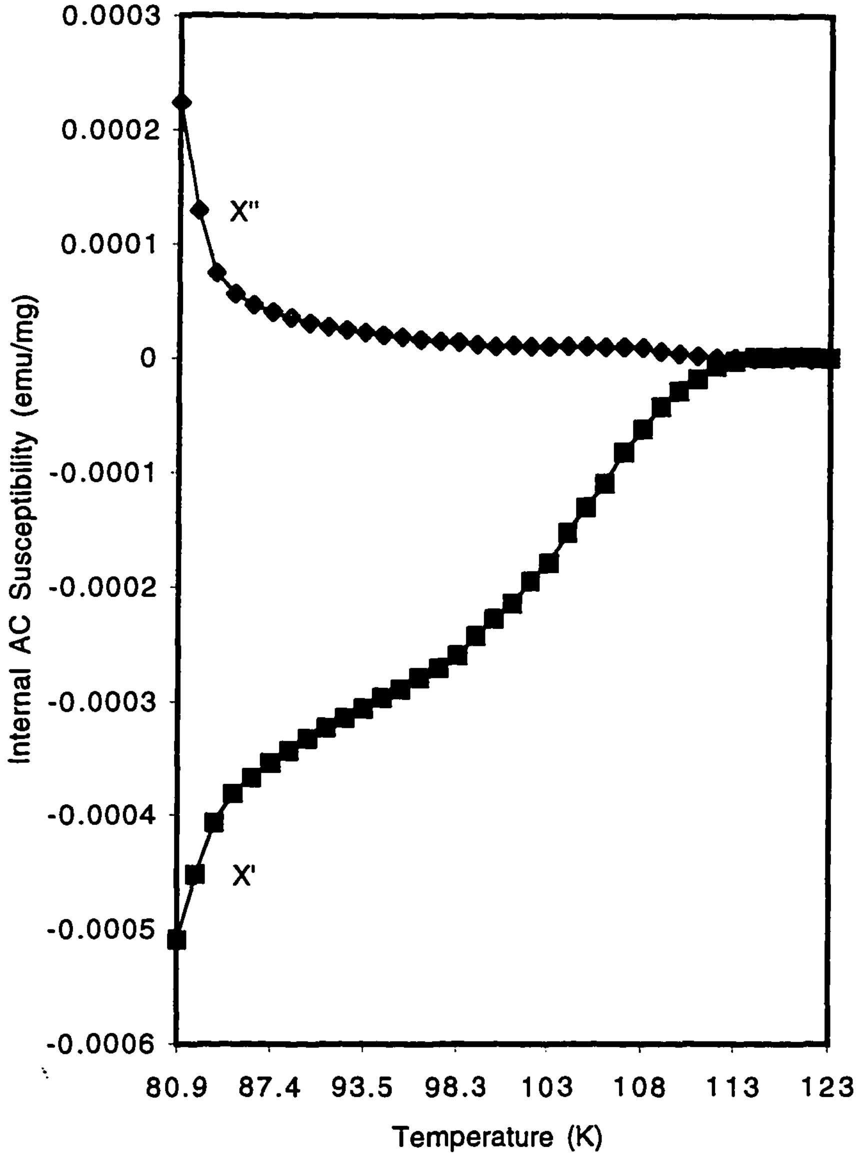

Figure 11. (TlPbSrCaCu) Doping Ba = 0.4

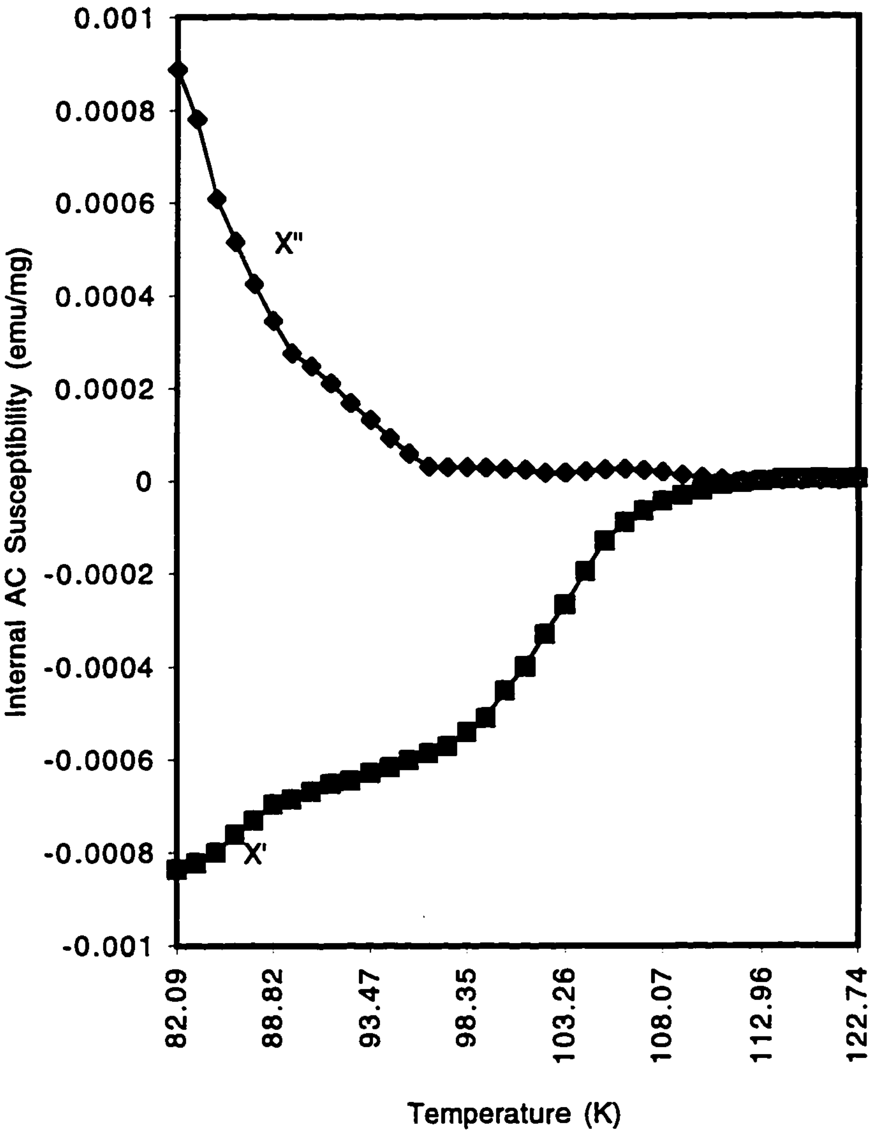

Figure 12. (TlPbSrCaCu) Doping Ba = 0.5

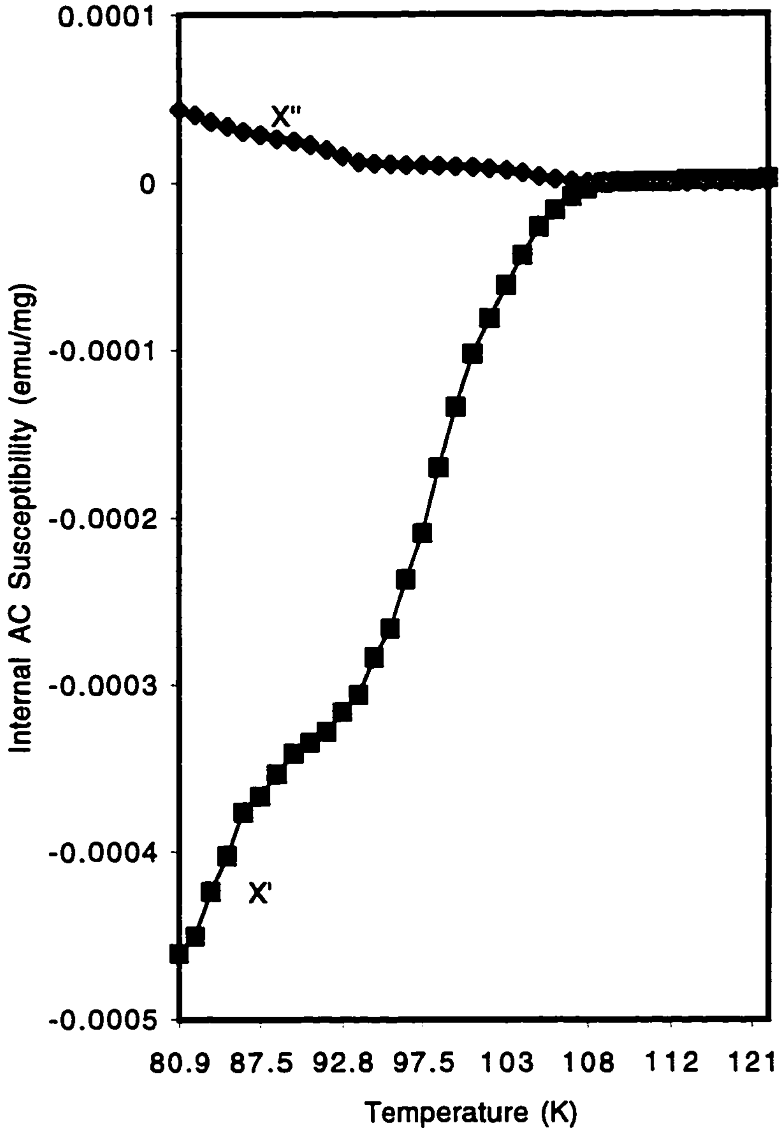

Figure 13. (TlPbSrCaCu) Doping Ba = 0.6

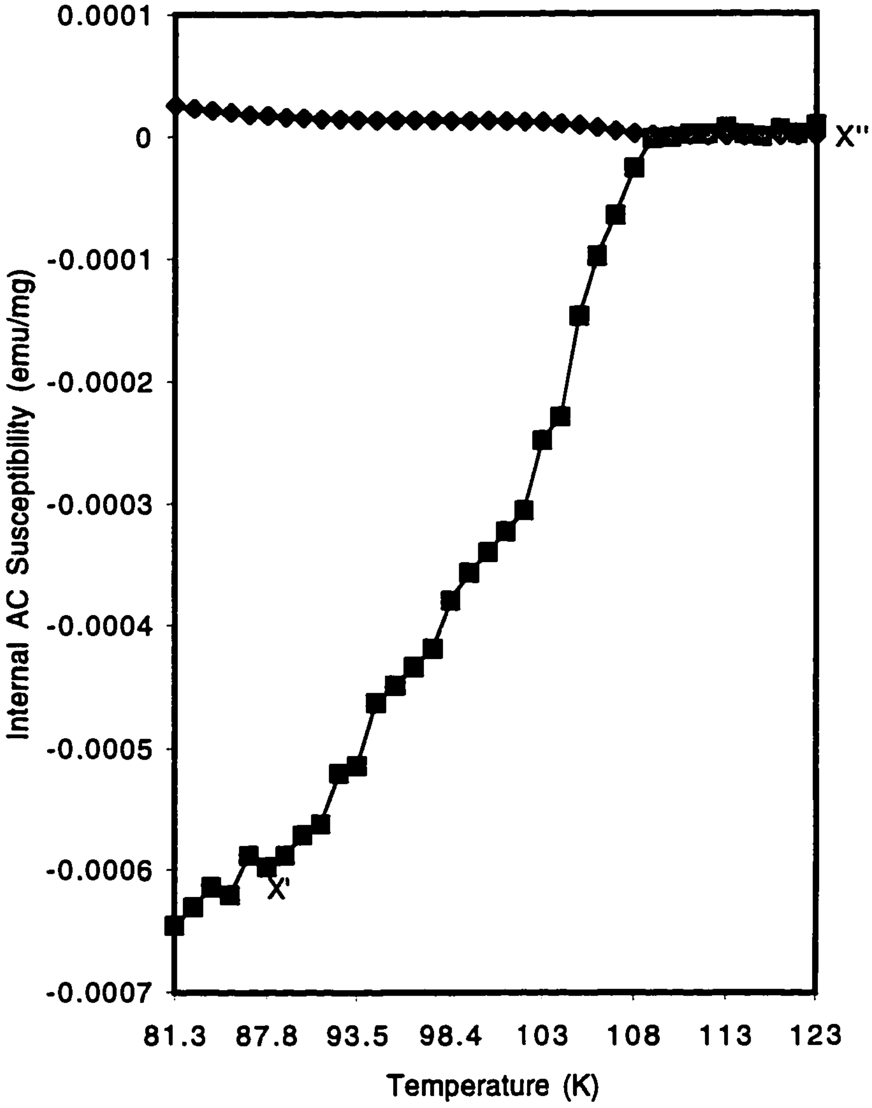

Figure 14. (TlPbSrCaCu) Doping Ba = 0.7

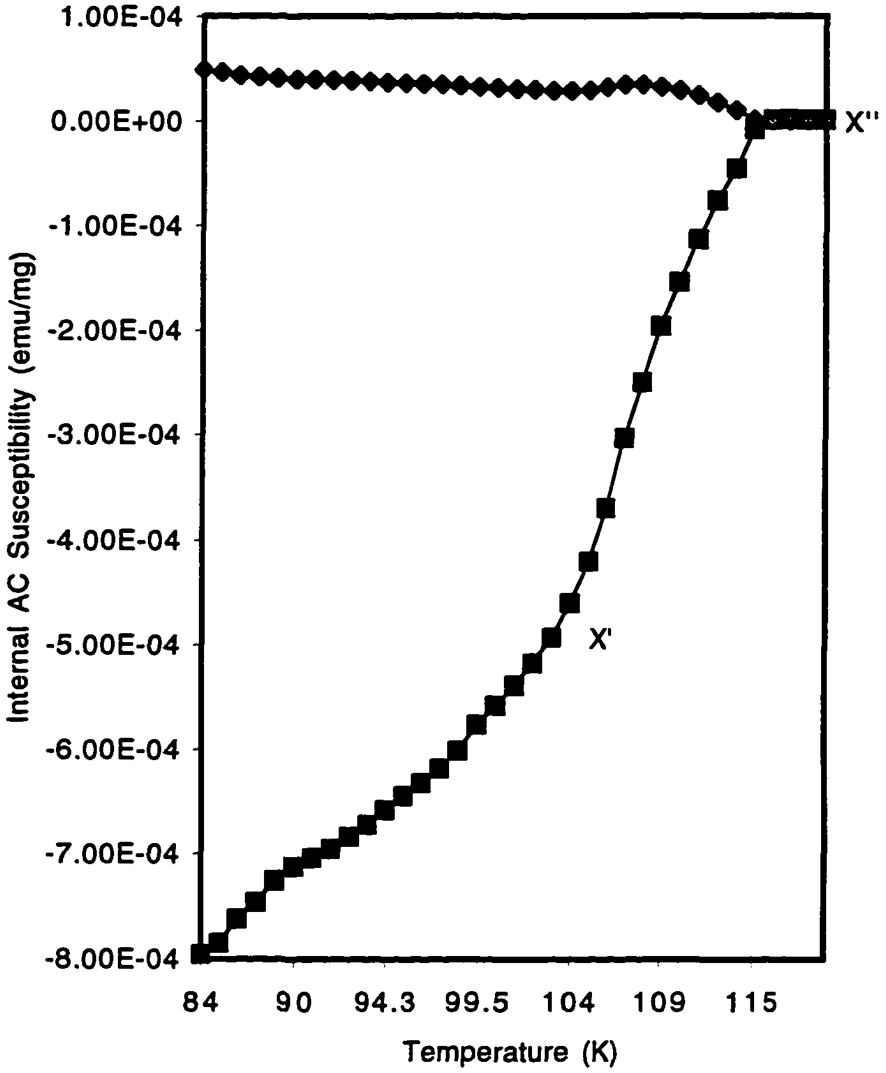

Figure 15. (TlPbSrCaCu) Doping Ba = 0.75

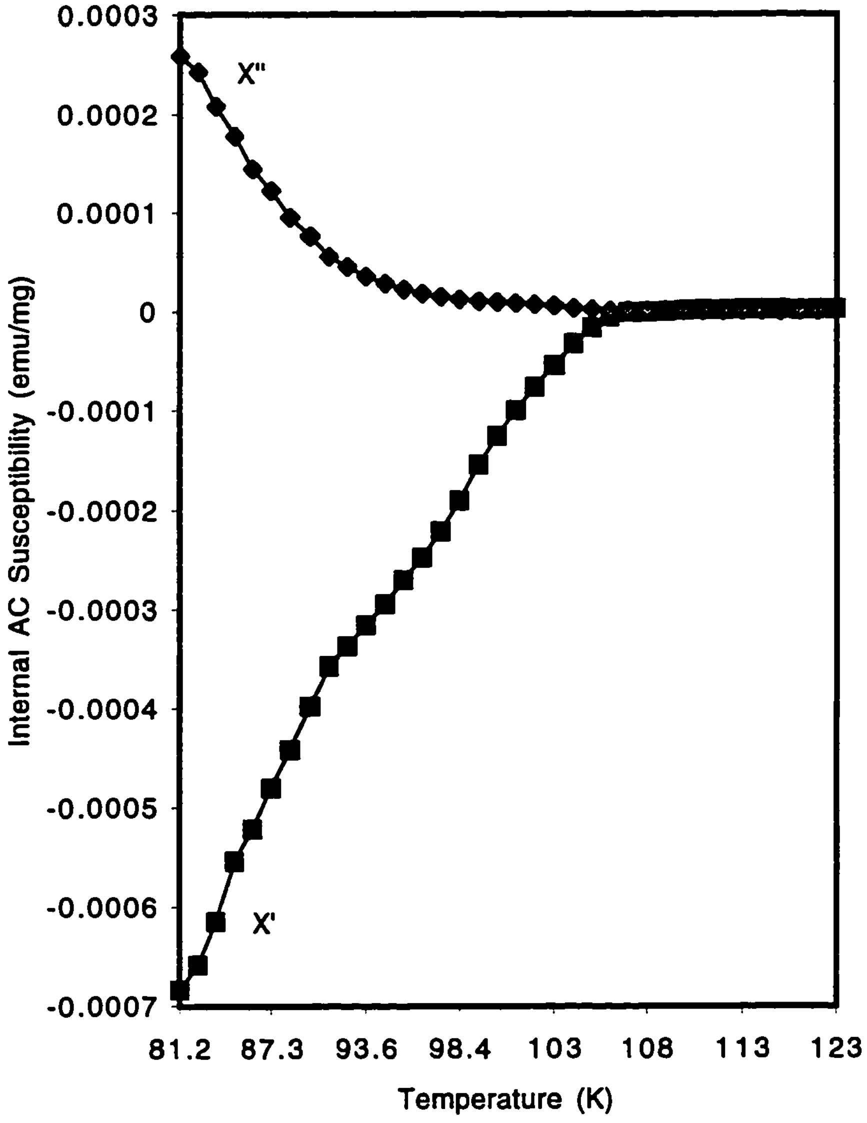

Figure 16. (TlPbSrCaCu) Doping Ba = 0.8

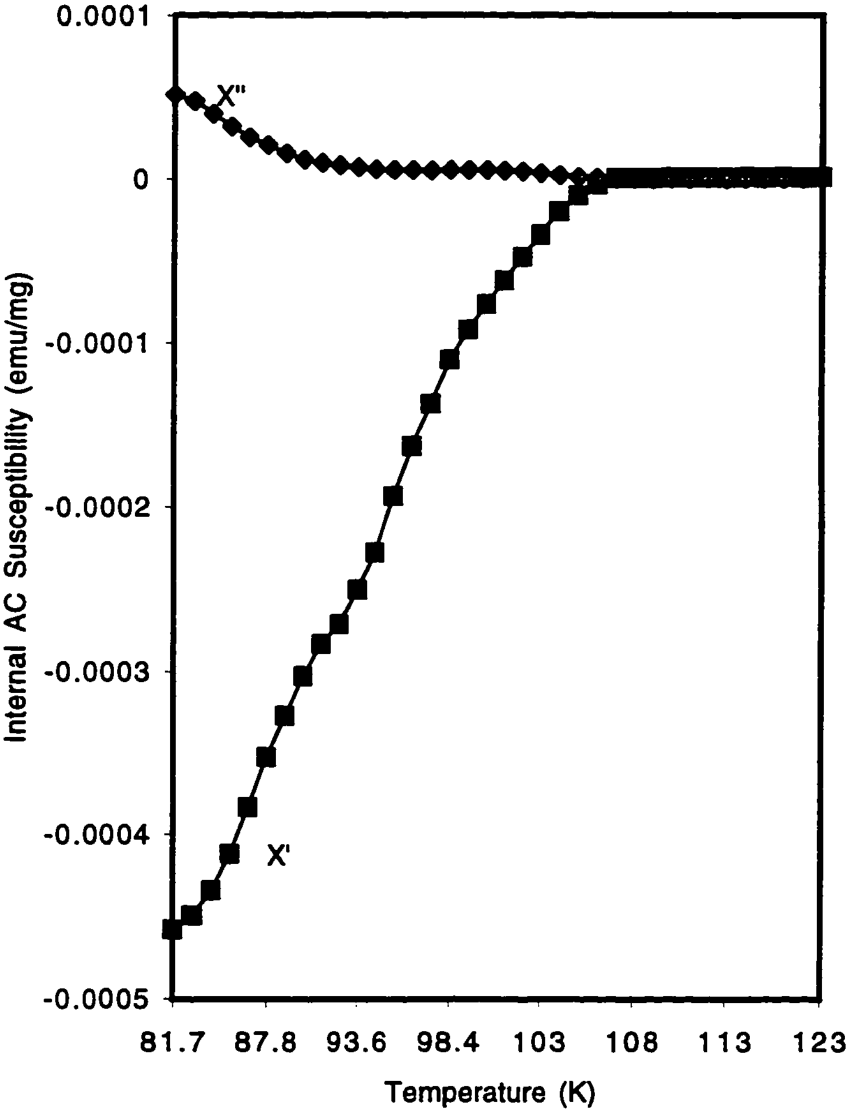

Figure 17. (TlPbSrCaCu) Doping Ba = 0.9

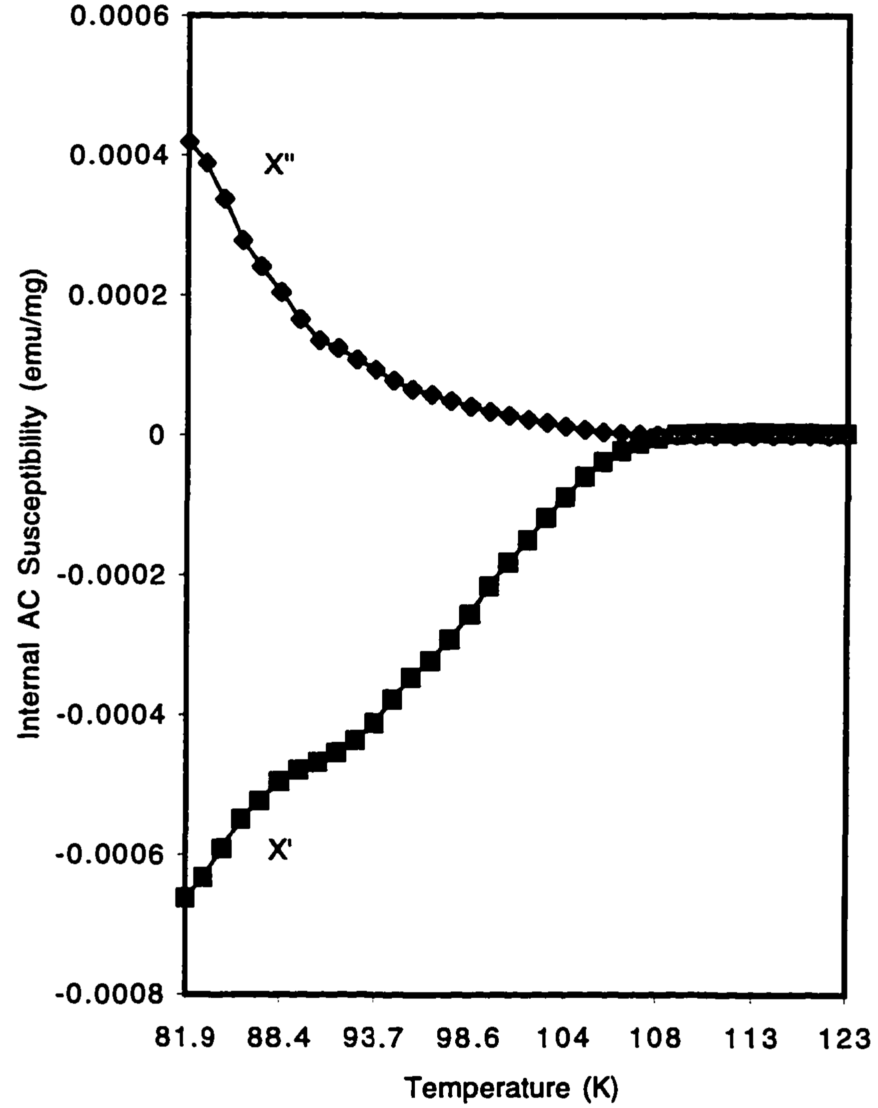

Figure 18. (TlPbSrCaCu) Doping Ba = 1.0

Optimizing The Ba Doping Process

Based on optimized processing for sample fabrication, it was concluded that the best set-up for sample sintering in this particular environment and conditions is 880°C temperature and 10 minutes sintering time. Following this sample fabrication procedure, samples with Ba = 0.0 to 1.0 were prepared for $(Tl_{0.5}Pb_{0.5})Sr_{2-x}Ba_xCa_2Cu_3O_y$. The samples were sintered in air by placing the samples inside a platinum crucible. Platinum is the only known material that doesn't react violently with Tl. The Platinum crucible was covered by a lid, made of Platinum.

AC susceptibility measurement for those samples were conducted and the results were listed in Appendix A. Magnetization vs. Temperature graphics are shown in Figure 7 through 18. Table 8 summarize the T_c, phase purity and transitional width which represents the volume fraction.

X-ray diffraction analysis was conducted to identify the phase (see Figures 19 to 30) in each of the samples. X-ray with wave length λ = 1.54 A was used to scan each sample and was directed to the samples at angles θ = 0.00 to 30.00, i.e. 2θ = 0.00 to 60.00. All peaks on Intensity vs. Angle 2θ charts were marked with the corresponding crystal plane and phase number based on the d-spacing and x-ray angle calculation.

Table 8

Summary Chart of T_c, Volume Fraction and Phase Purity

Ba Concentration	Superconducting Tc (K)	Phase	Transition
0	108.005	1212	wider
0.1	112.855	1212	wider
0.2	113.018	1212	wider
0.3	112.215	1212&1223	wider
0.4	114.955	1212&1223	narrow
0.5	112.955	1212&1223	narrow
0.6	109.45	1223	narrow
0.7	111.08	1223	narrow
0.75	116.67	1223	Sharper
0.8	110.205	1223	narrow
0.9	107.45	1223	narrow
1	110.345	1223	wider

From those charts, it was shown that at Ba=0.00, the (1212) phase is the dominant phase with small amounts of 1223 phase present. However, as Ba concentration increases to Ba=0.75, there is a trend that the 1223 phase gradually dominates in the material. After Ba concentration becomes greater than 0.75, the 1212 phase increases in the material.

Based on the above analysis, it was concluded that the Ba=0.75 provides the best superconducting results and the optimum properties, i.e. $(Tl_{0.5}Pb_{0.5})Sr_{1.25}Ba_{0.75}Ca_2Cu_3O_y$ is the best superconducting material compared to other Ba doped superconductors. It has the highest T_c, Volume fraction and purity of Tl-1223 phase. This compound will be used for studies on Eu doping.

The Tl-based copper oxides are thermally unstable phases, and hence, are very difficult to prepare as pure phases. Moreover, the thallium compounds are severely nonstoichiometric and contain a considerable concentration of structural defects, which significantly affects its physical properties. The copper and thallium chemistry are particularly interesting in terms of the Cu and Tl valences and the correlation between their valences and superconductivity of Tl-based cuprate materials (Greenblatt, 1989).

Tl_2O_3 is highly volatile at temperatures above its melting point of ~710°C. In preparing these high T_c superconducting oxides, prevention of thallium loss is the most important factor for maintaining the stoichiometry of the products to ensure the formation of the desired phase.

Most of the well characterized thallium based compounds are generally prepared with modifications of the reaction temperature, time, and the starting chemical composition. In open reaction systems, a phase transformation phenomenon was observed as a result of Tl loss. Under controlled reactions, single phase samples can be obtained (Greenblatt, 1989).

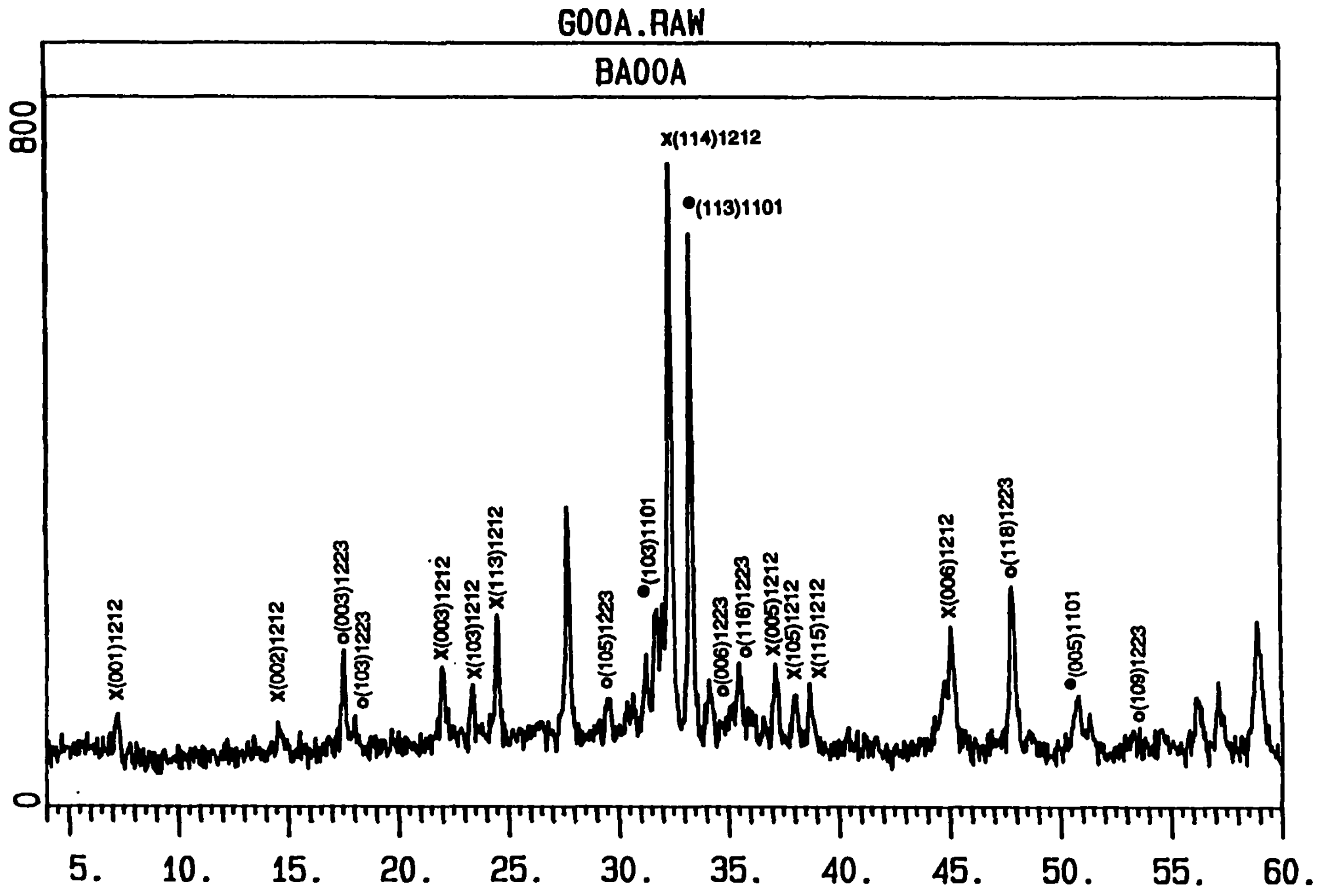

Figure 19. X-Ray Diffraction for Ba = 0.0

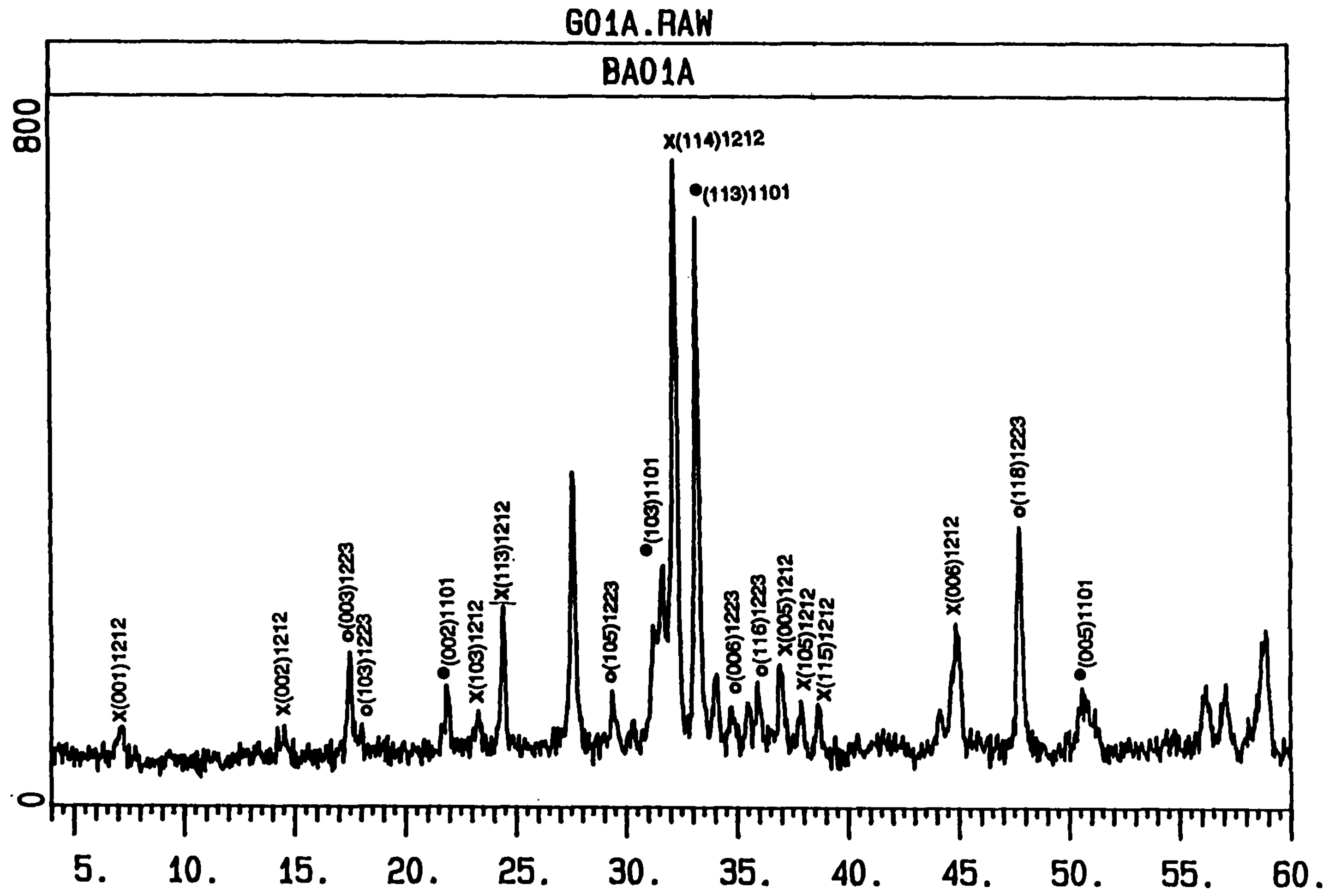

Figure 20. X-Ray Diffraction for Ba = 0.1

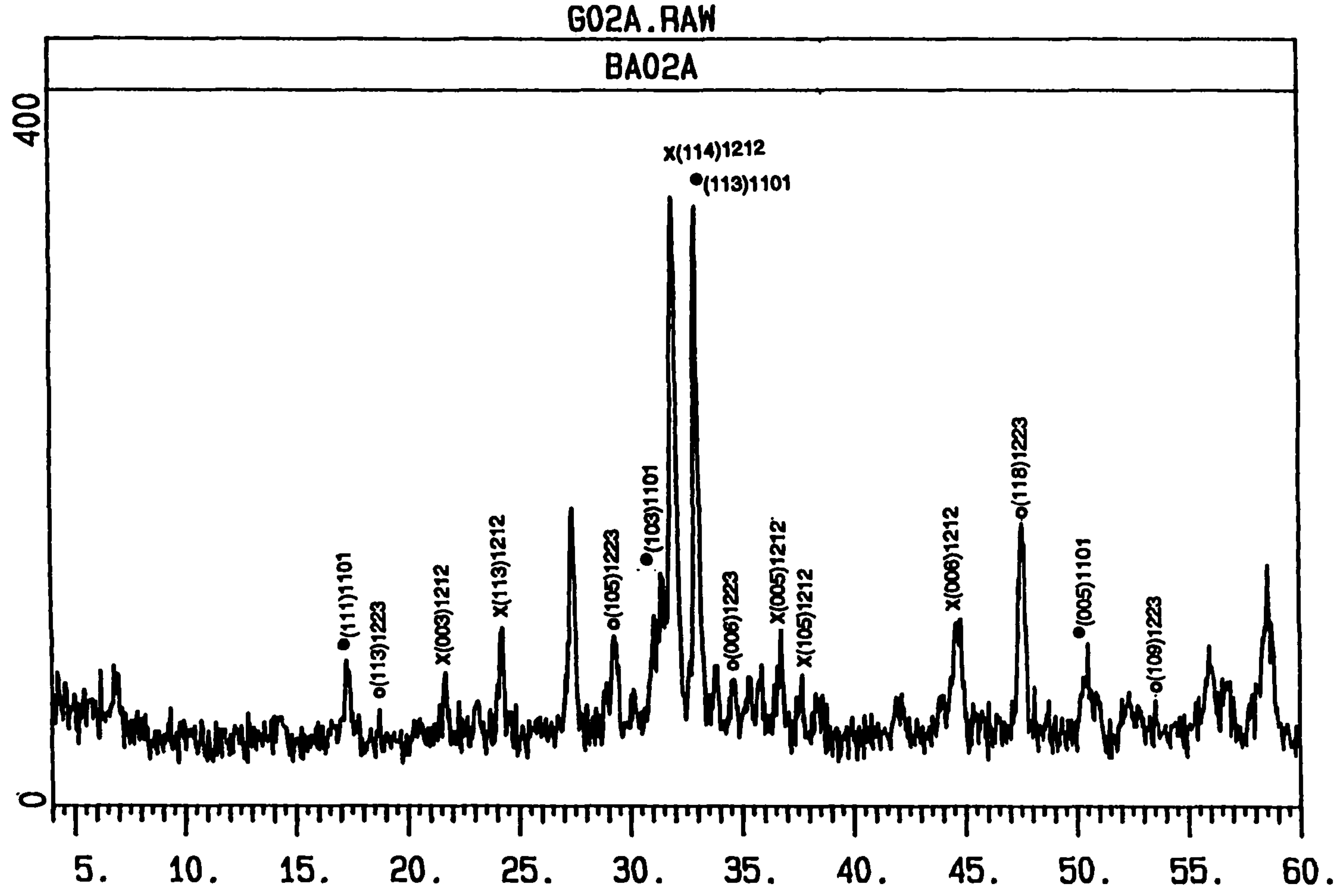

Figure 21. X-Ray Diffraction for Ba = 0.2

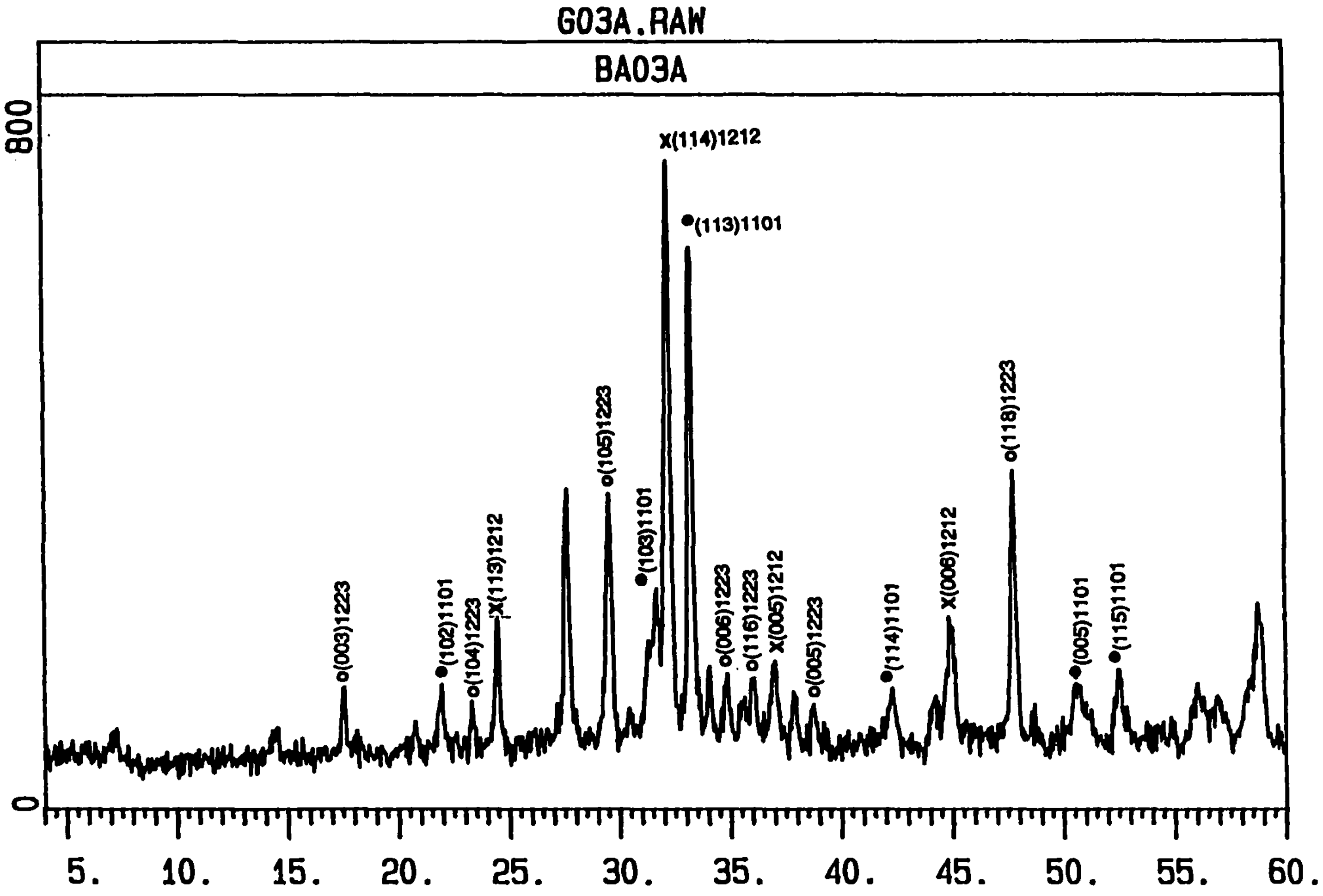

Figure 22. X-Ray Diffraction for Ba = 0.3

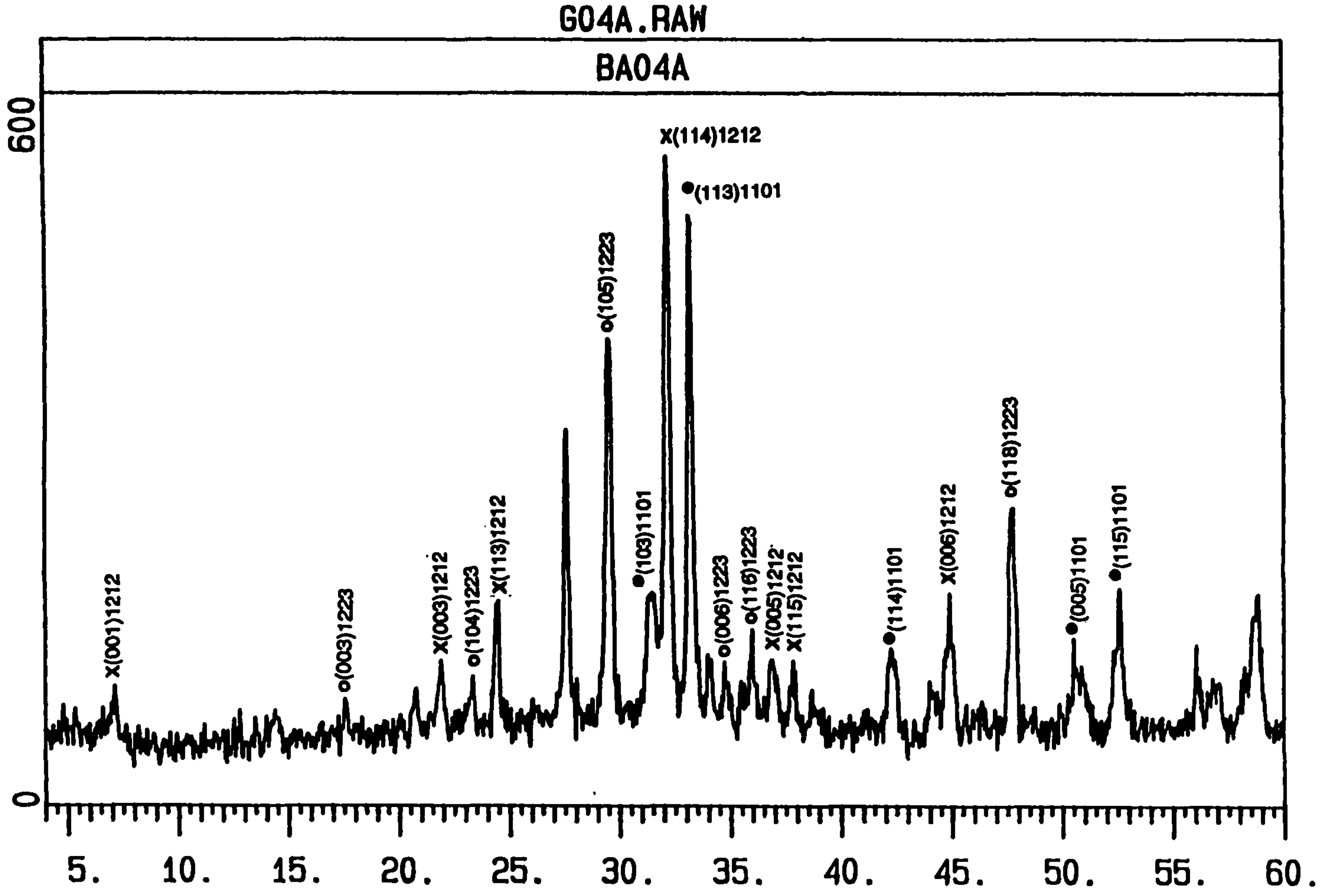

Figure 23. X-Ray Diffraction for Ba = 0.4

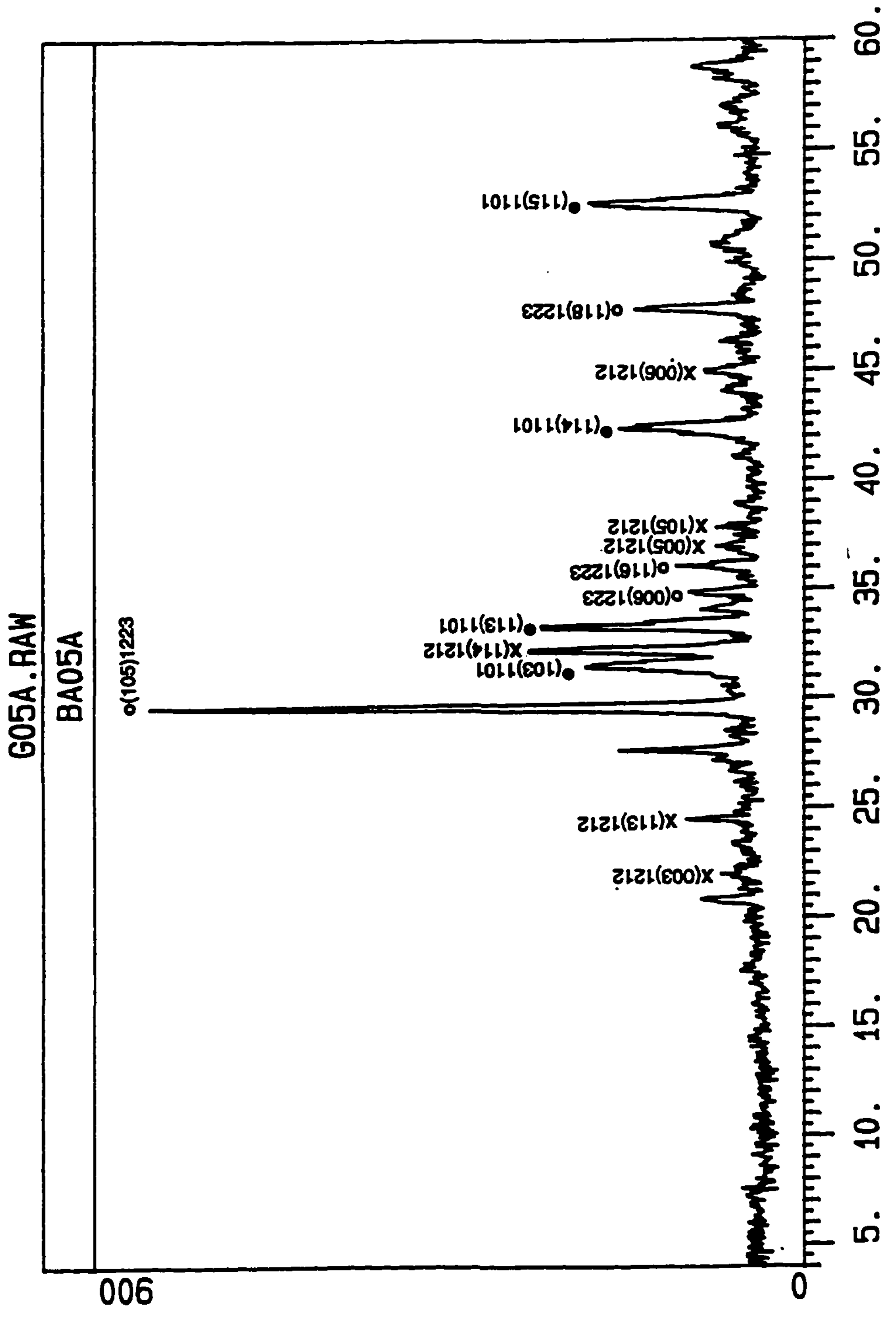

Figure 24. X-Ray Diffraction for Ba = 0.5

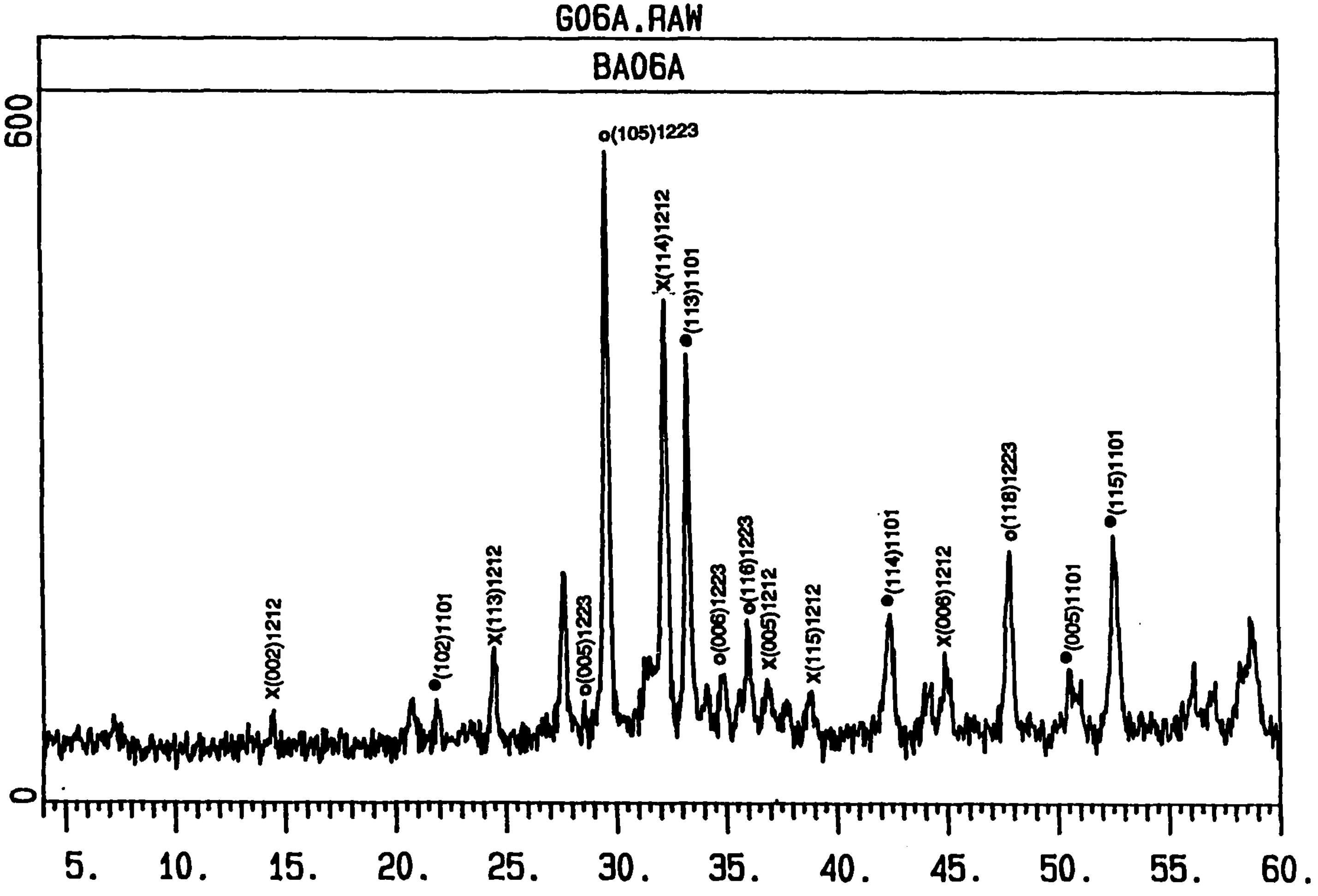

Figure 25. X-Ray Diffraction for Ba = 0.6

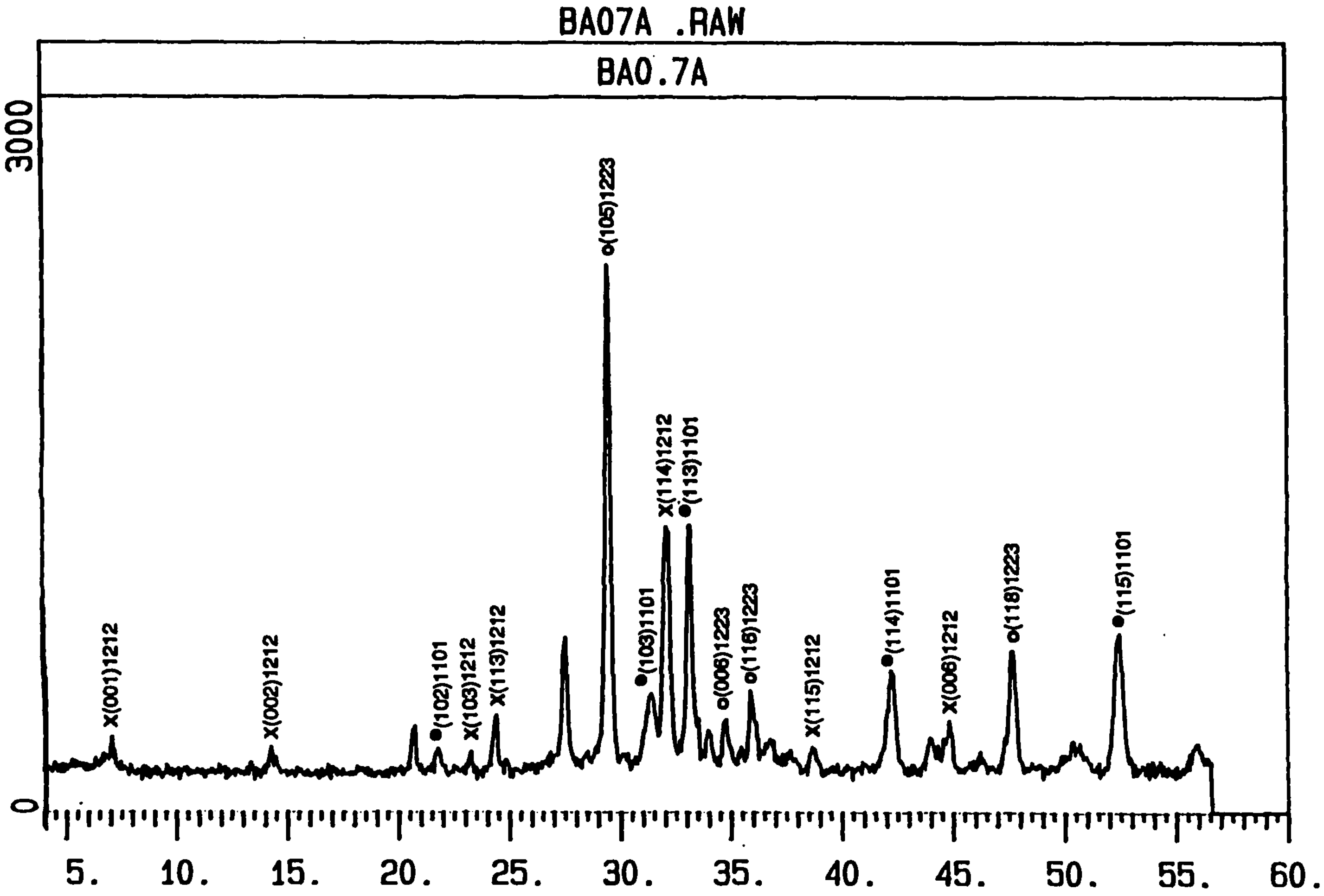

Figure 26. X-Ray Diffraction for Ba = 0.7

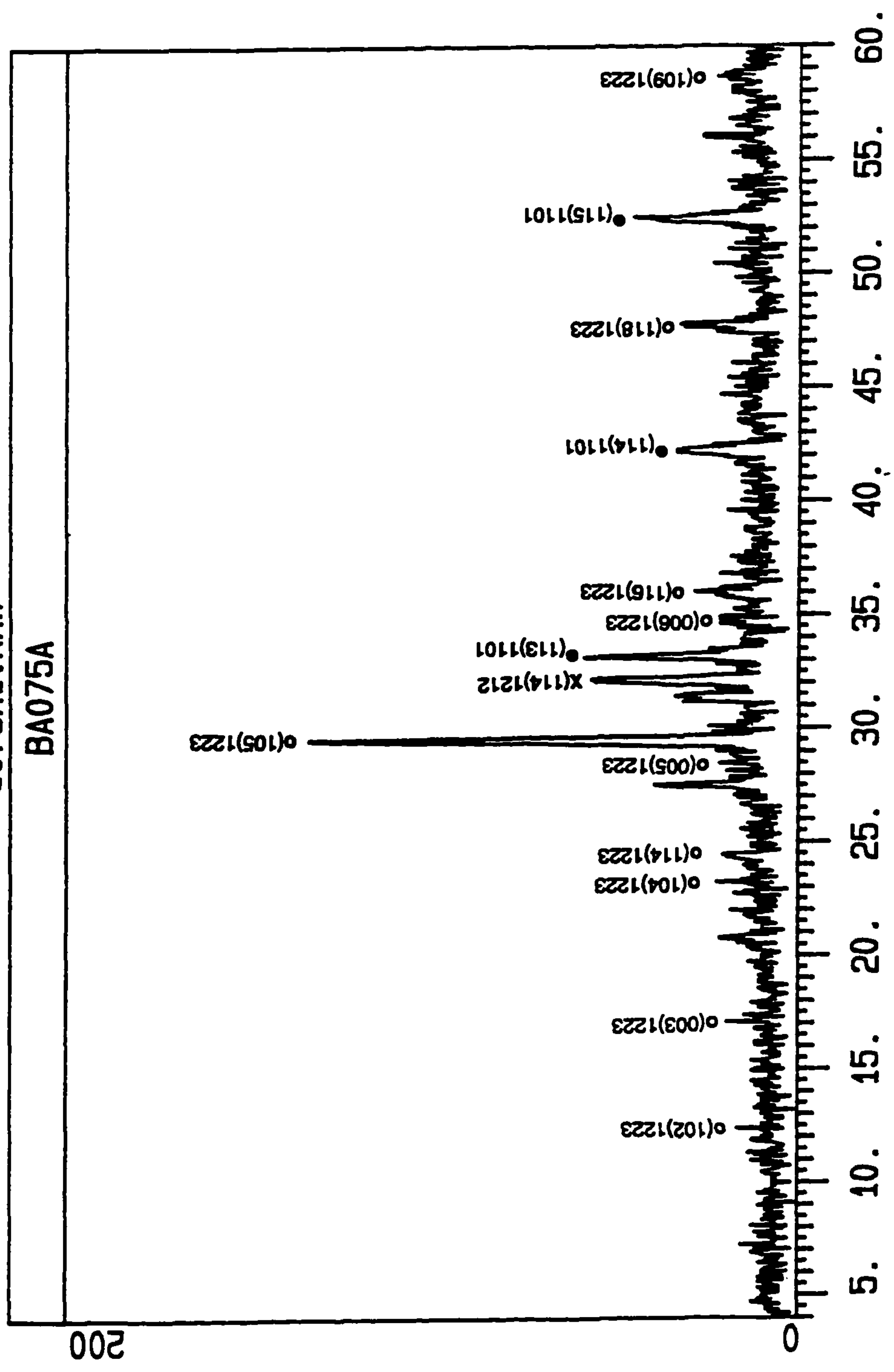

Figure 27. X-Ray Diffraction for Ba = 0.75

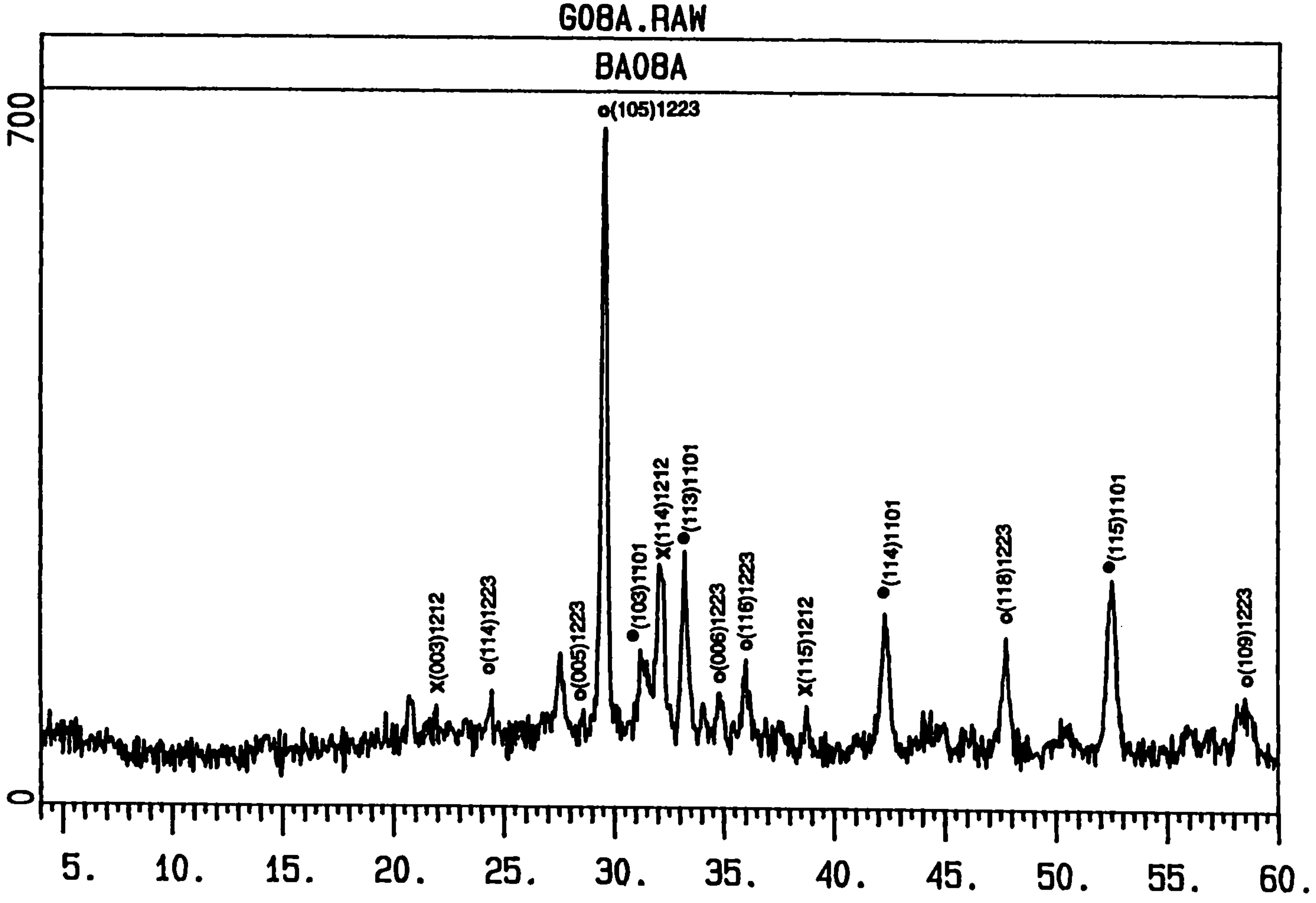

Figure 28. X-Ray Diffraction for Ba = 0.8

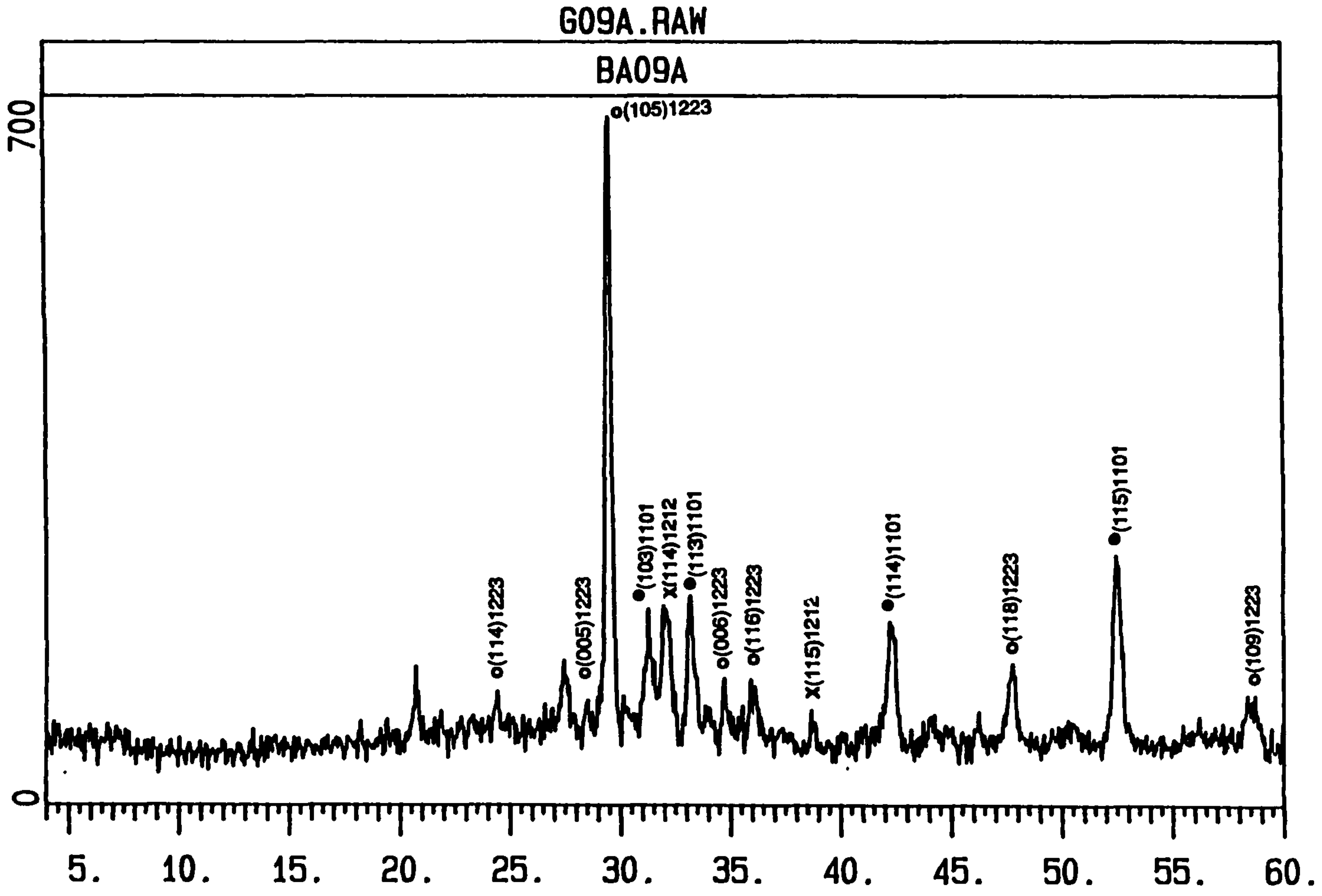

Figure 29. X-Ray Diffraction for Ba = 0.9

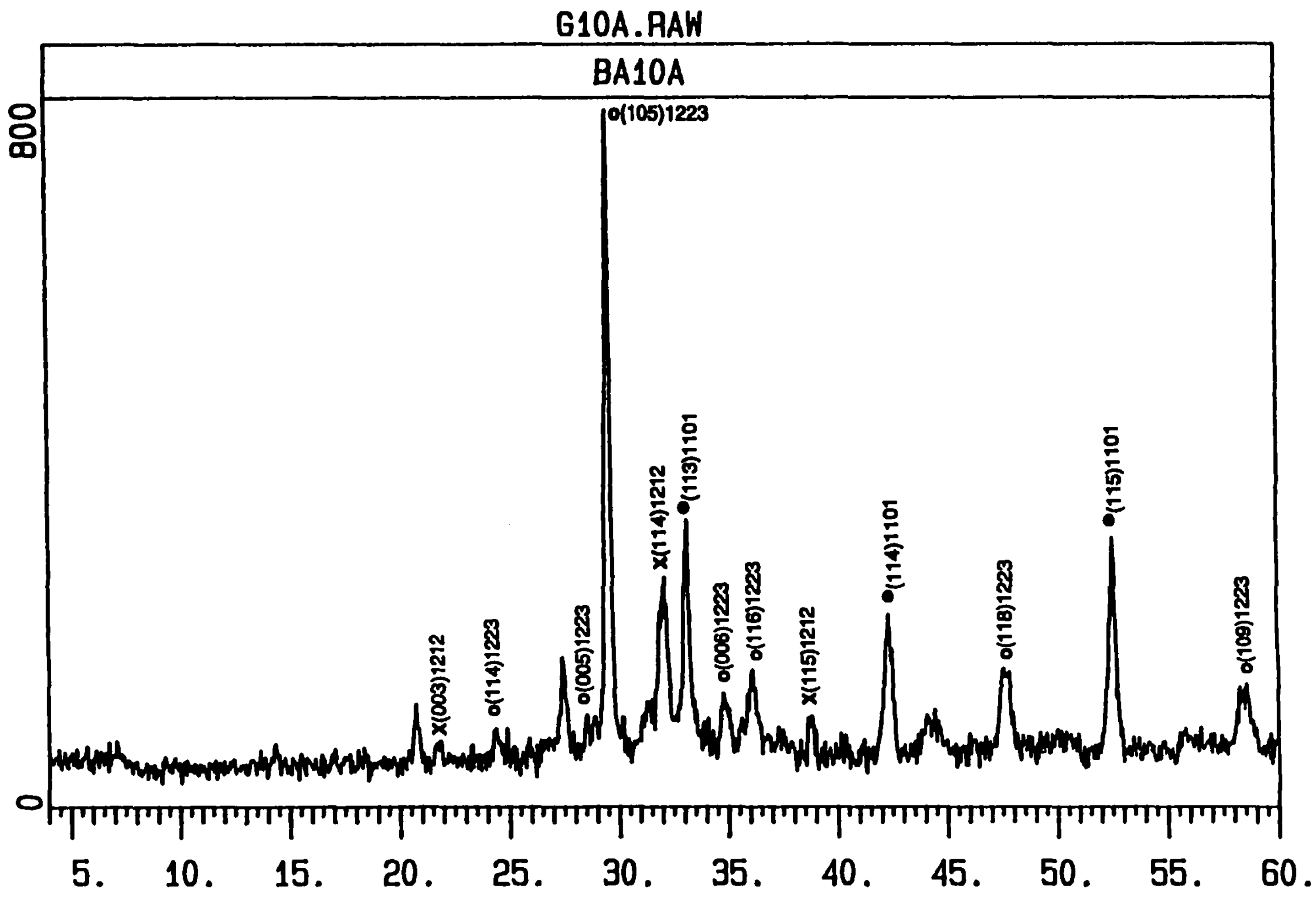

Figure 30. X-Ray Diffraction for Ba = 1.0

Optimizing Eu Doping Process

Eu was doped in the Tl and Pb site for Ba optimized compounds $(Tl_{0.5}Pb_{0.5})Sr_{1.25}Ba_{0.75}Ca_2Cu_3O_y$. Samples with Eu = 0, 0.02, 0.04, 0.06, 0.08, 0.10 were made as $(Tl_{0.5-x}Pb_{0.5-x}Eu_{2x})Sr_{(2-y)}Ba_yCa_2Cu_3O_z$ to optimize the Eu doping process.

X-ray diffraction analysis was performed to identify the phases present in the material.

AC susceptibility measurements were conducted for Eu doped samples. Data of temperature, χ', χ'' were recorded by AC susceptometer as shown in Appendix B. Temperature vs. Susceptibility graphs (with χ'') are shown from Figure 31 to Figure 36. Eu doping created a paramagnetic material because the susceptibility χ' shows positive value (above the 0.00 level on the graphs) above the critical temperature. Such a behavior is unusual in superconductors.

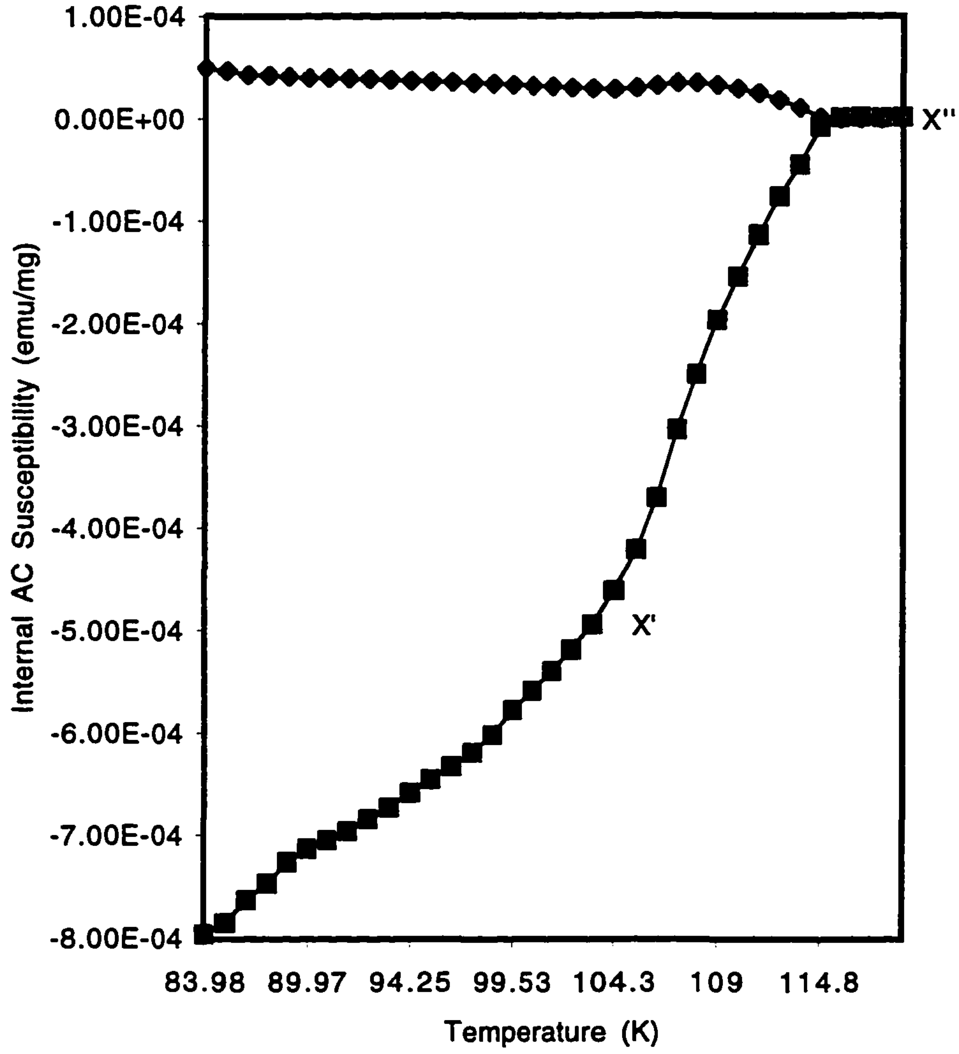

Figure 31. (TlPbBaSrCaCu) Doping Eu = 0.00

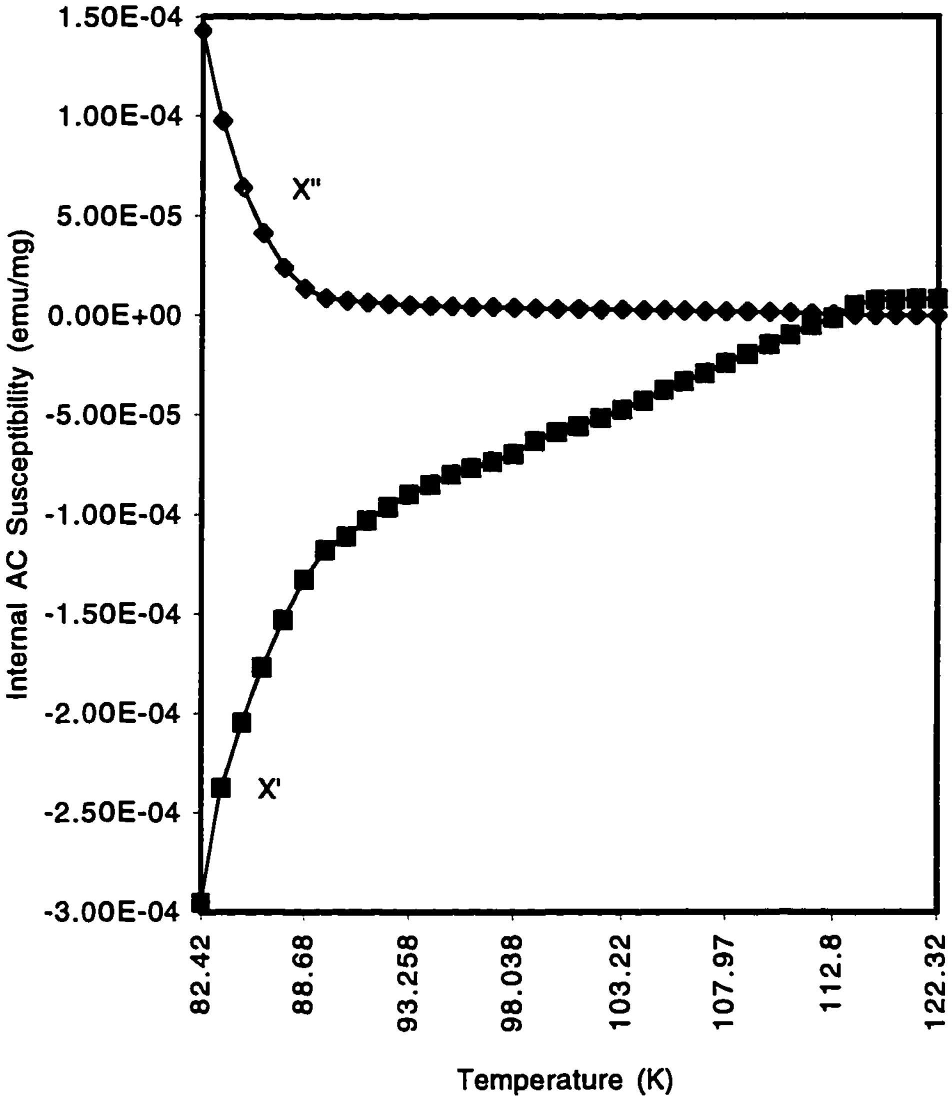

Figure 32. (TlPbBaSrCaCu) Doping Eu = 0.02

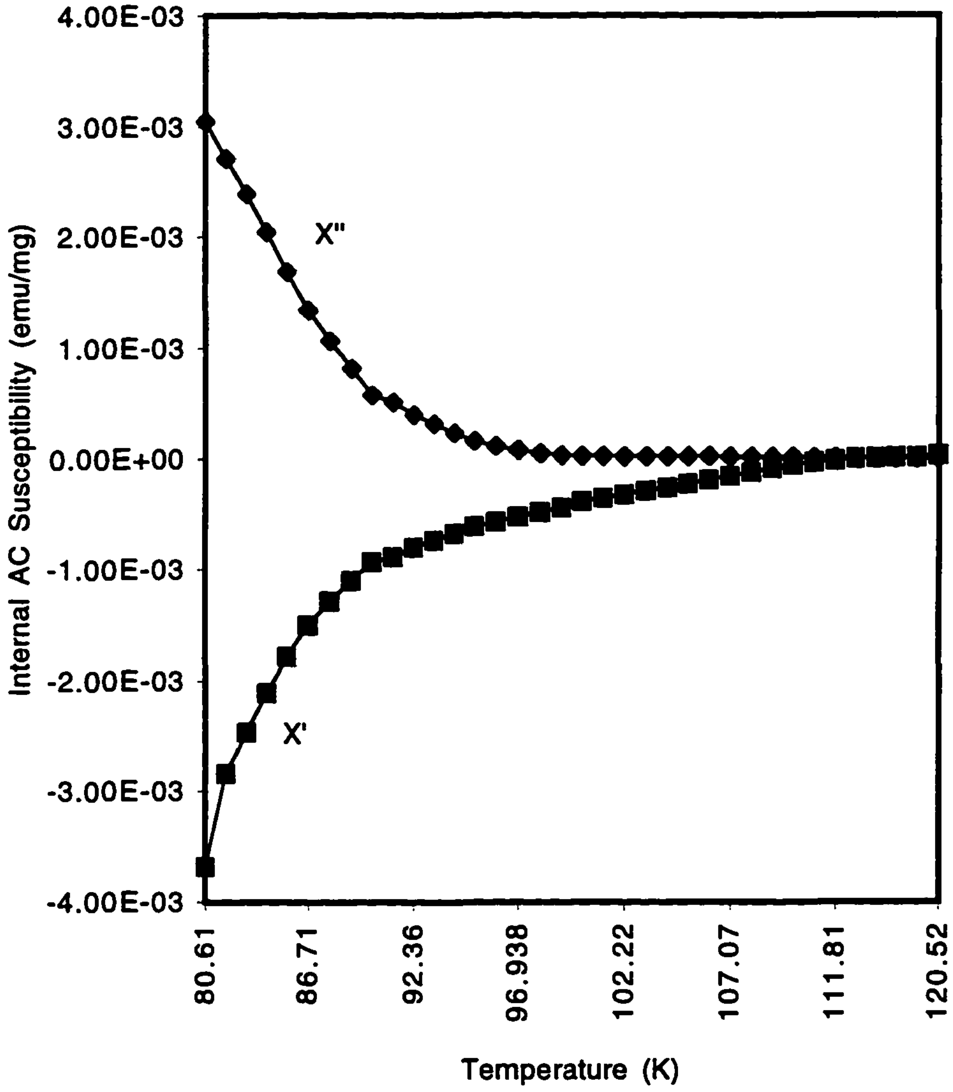

Figure 33. (TlPbBaSrCaCu) Doping Eu = 0.04

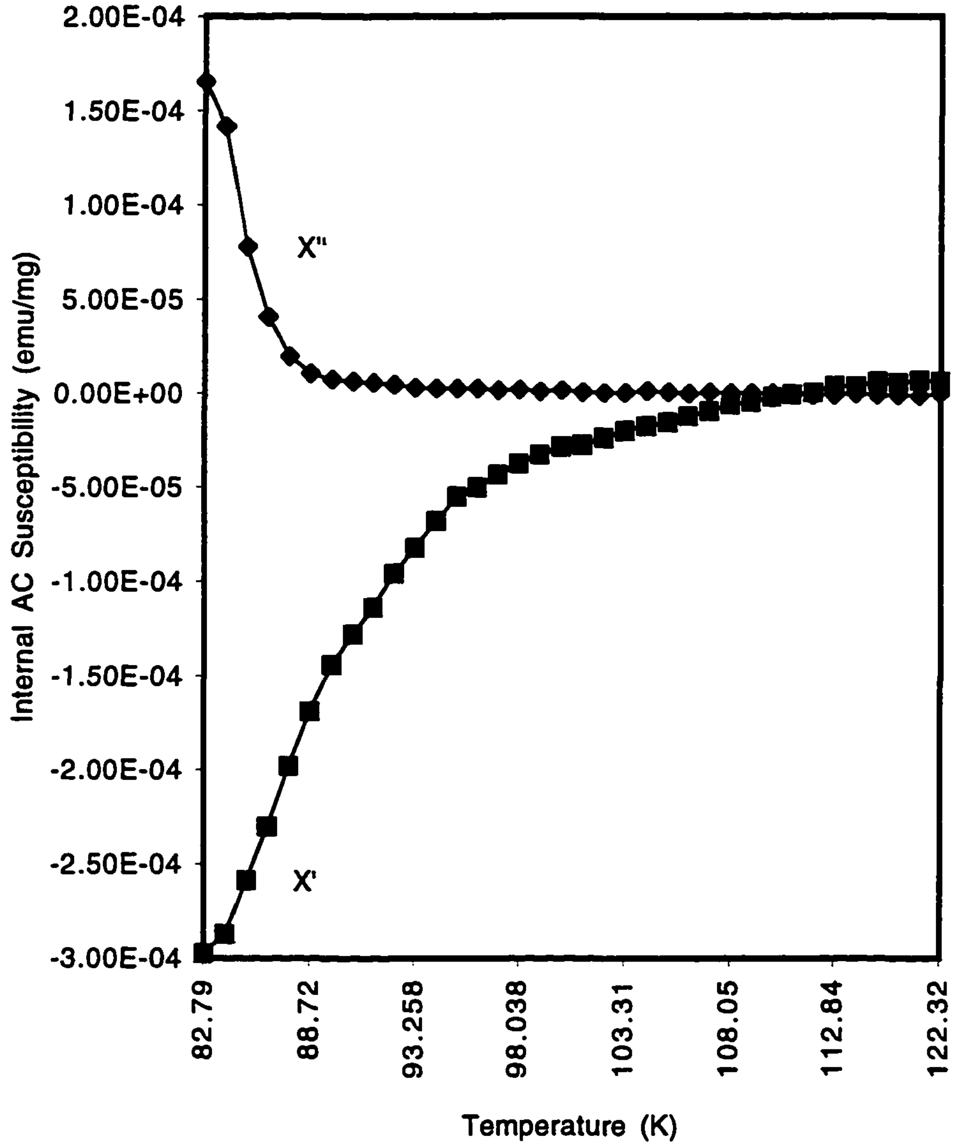

Figure 34. (TlPbBaSrCaCu) Doping Eu = 0.06

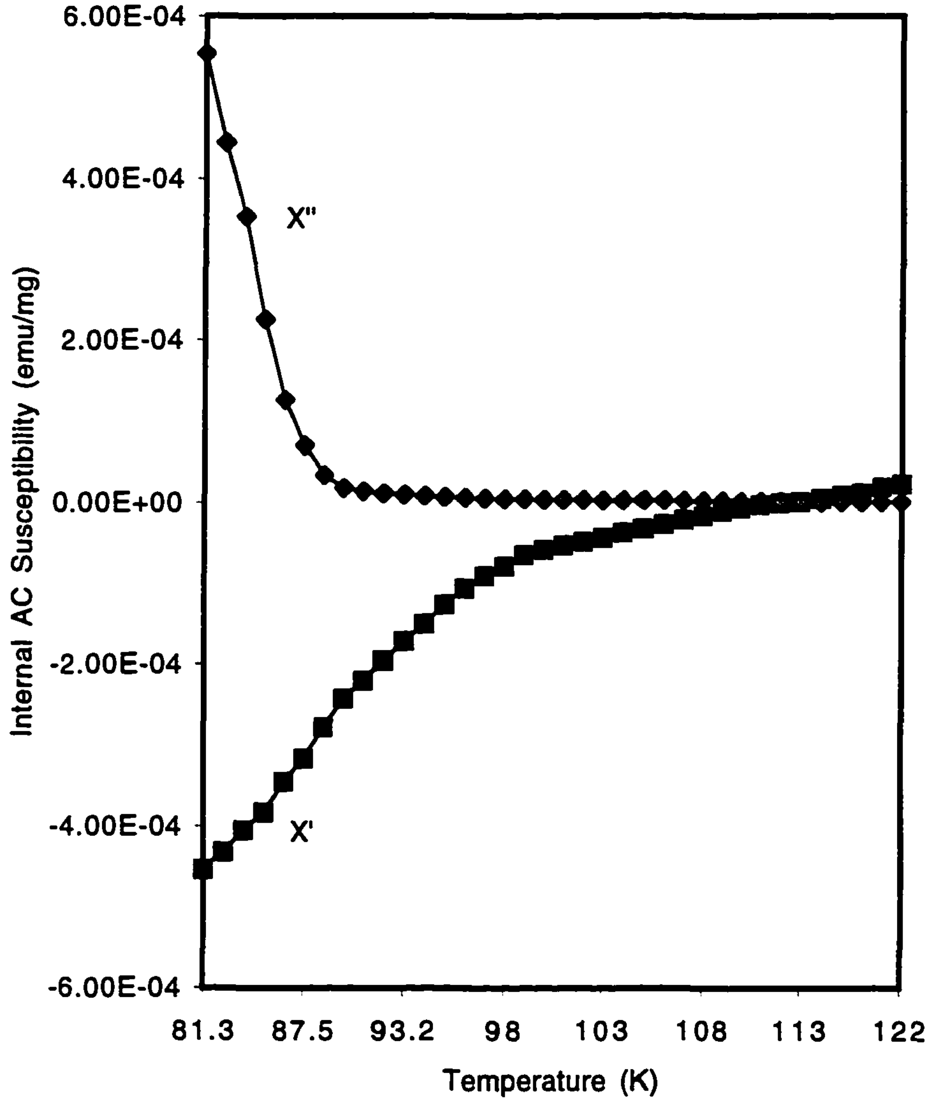

Figure 35. (TlPbBaSrCaCu) Doping Eu = 0.08

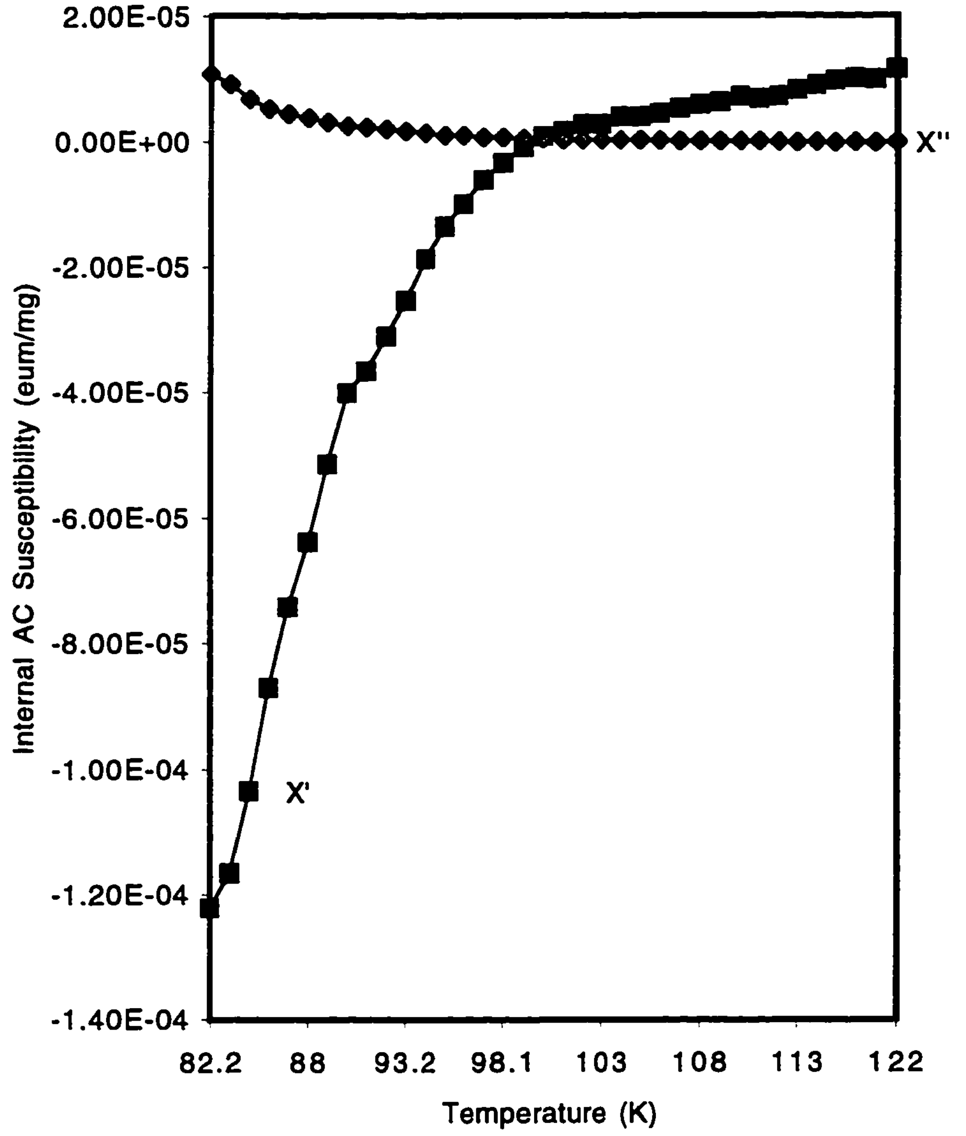

Figure 36. (TlPbBaSrCaCu) Doping Eu = 0.10

The summary of T_C, Phase purity and other superconductor properties is listed in Table 9. Based on the above analysis, it is concluded that Eu doping doesn't improve the supercondutivity for a material. It increased the tendency to become a paramagnetic material ($\chi' \rightarrow$positive). T_C was reduced by Eu doping. Therefore, basically the best superconducting material is still $(Tl_{0.5}Pb_{0.5})Sr_{1.25}Ba_{0.75}Ca_2Cu_3O_y$ with non Eu doping Eu=0.

Table 9

Summary of T_C, Phase Purity and Superconductor Properties for Eu Doping

Eu Concentration	Superconducting Tc	Phase	Transition
0	116.67	1223	sharper
0.02	114.715	1212	wider
0.04	114.283	1212	wider
0.06	111.88	1212	wider
0.08	112.888	1212	wider
0.1	100.42	1212	wider

However, the paramagnetic properties of the superconductor above Tc will be very useful to determine the site locations of atoms, and the spacing between atoms in the material using the Mossbaur spectroscopy technique.

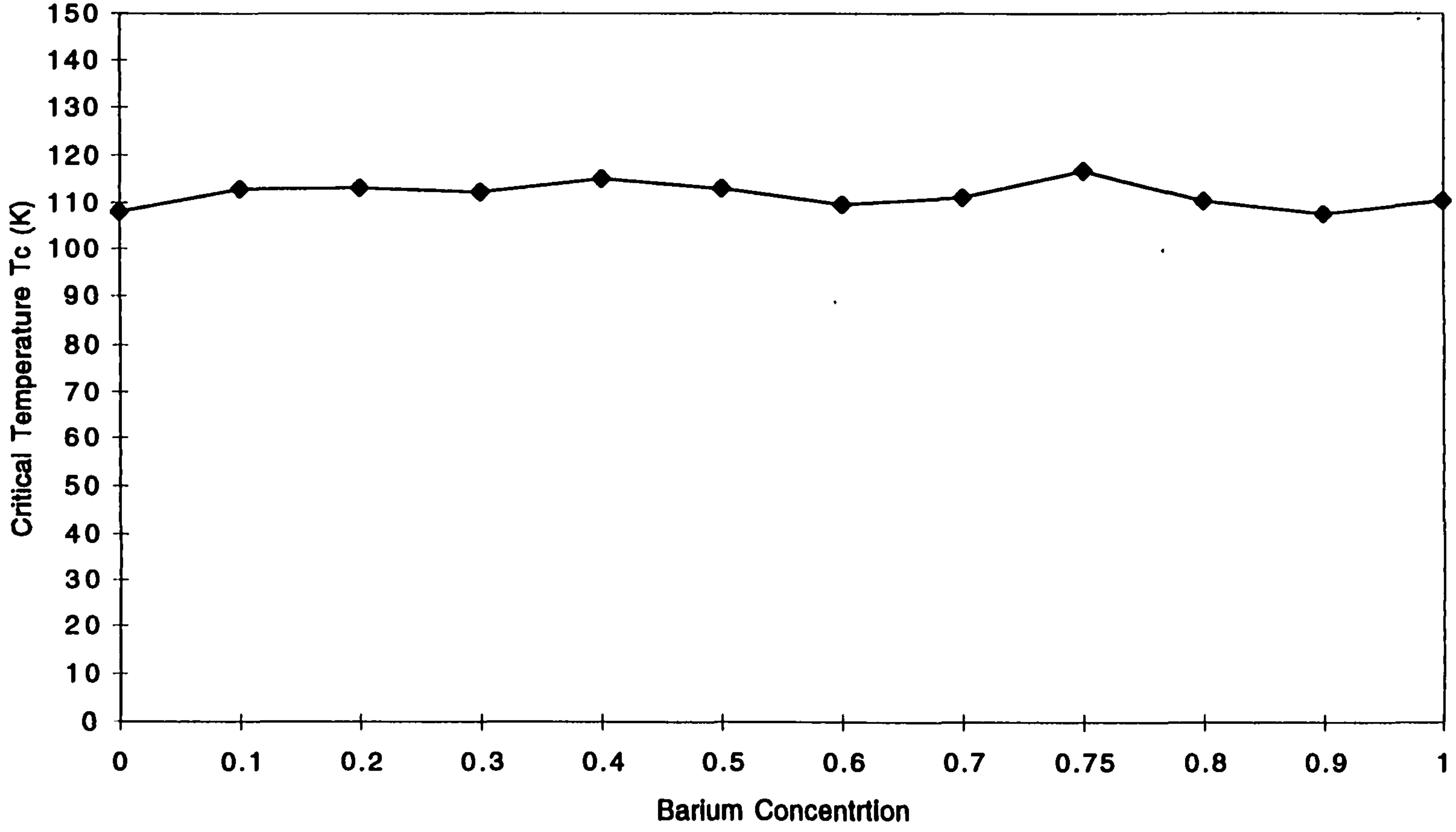

Figure 37. Critical Temperature Tc (K) Vs Barium Concentration

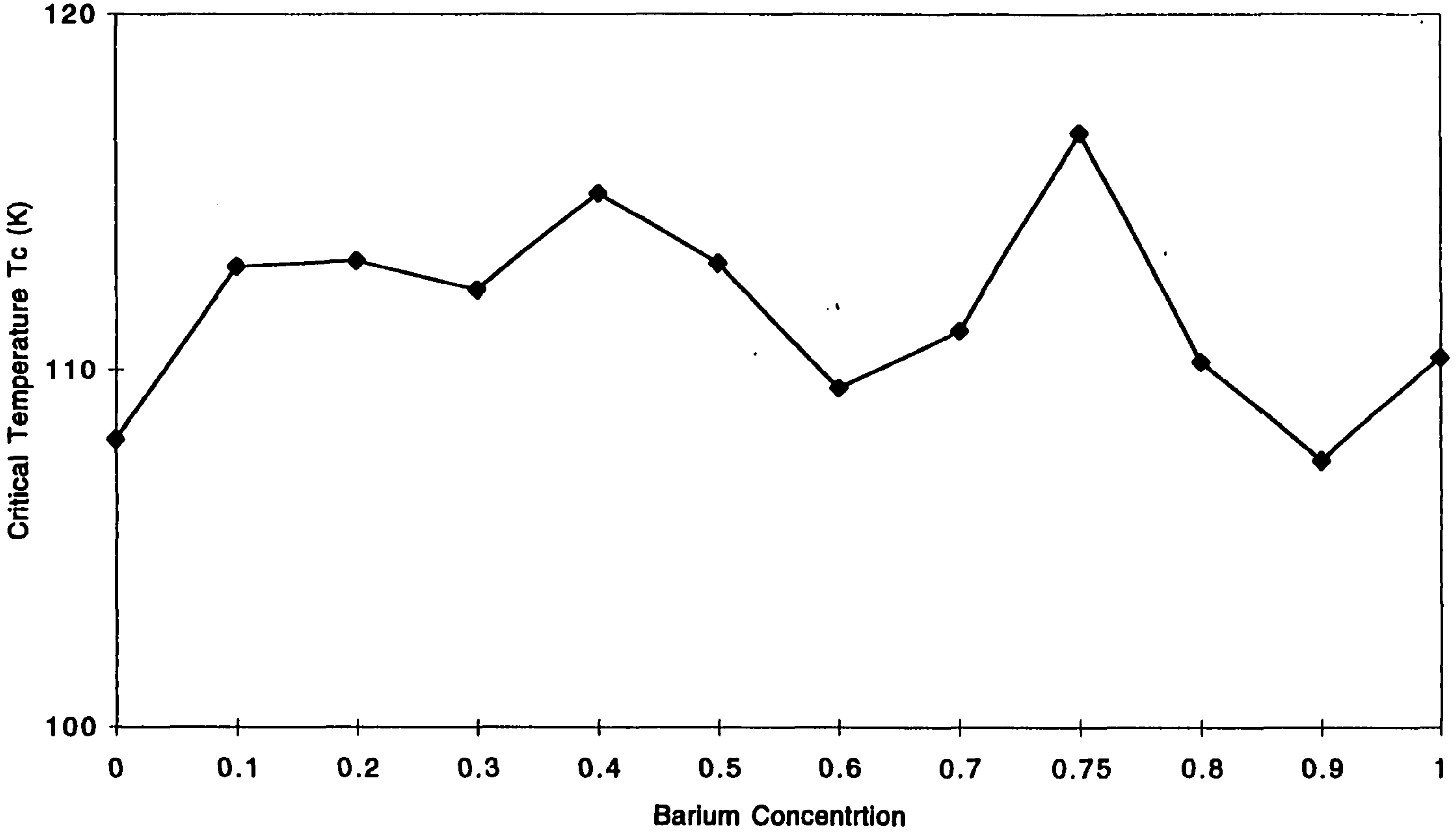

Figure 38. Critical Temperature Tc (K) Vs Barium Concentration

Results and Discussions

Following results were summarized from collected data and the findings to answer the research questions.

Research Question #1

The **$(Tl_{0.5}Pb_{0.5})Sr_{2-y}Ba_yCa_2Cu_3O_z$** (1223) superconductors with y varying from y = 0 to y = 1.0 were processed after optimizing the sintering temperature. Samples were sintered in air inside a Pt crucible at 880°C for 10 min. Sintered samples were characterized using the susceptibility and x-ray diffraction measurements.

Research Question #2

From the AC susceptibility measurements, the zero resistance critical temperature (**T_{co}**) is the temperature at which the real part of the complex susceptibility (χ') becomes negative as the samples are cooled down. Figure 37 and Figure 38 shows the critical temperature vs Barium doping for a set of samples processed under the same conditions. The critical temperature of the samples is between 100°K and 120°K. Sample with Ba doping of 0.75 showed the highest T_c of 118°K indicating the dominance of 1223 phase in this superconducting compound. The magnitude of real part of AC susceptibility (χ') at 80°K and 100°K give us an indication of volume fraction of superconductivity. Larger the magnitude, higher is the volume fraction of superconductivity. Figure 39 and Figure 40 show the magnitude of real part of AC susceptibility at 80°K and 100°K vs the Barium concentration. The Barium concentration of 0.75 again shows higher magnitude of χ'. The magnitude ranges from 2×10^{-4} to 10^{-3} emu/gm.

Research Question #3

The powder x-ray diffraction measurements performed on the superconducting samples is very useful in determining the superconducting phases present in the samples since each of the phases has different unit cell dimensions. The strongest peak for the 1223 phase in powder x-ray diffraction measurements is the (105) peak close to $2\theta = 30°$. Figure 41 shows the intensity of the 1223 peak normalized to the maximum intensity peak vs the Barium concentration. This figure shows that at lower concentration of Ba below 0.5, the intensity of (105) peak is slowly increasing and then above 0.5, the intensity of the 1223 peak improves, indicating the growth of significant 1223 phase in the superconductor. The strongest peak for the 1212 phase is close to $2\theta = 32°$. At $y < 0.5$, the 1212 peak's intensity is larger than the (105) peak of 1223 showing that the sample is predominantly 1212 phase for Barium concentration below 0.5.

Research Question #4

Other studies of importance in this research is the doping of Europium in Tl and Pb sites. Europium being a rare-earth metal offers the possibility of magnetic studies using Mossbaur spectroscopy. The magnetic signatures of the

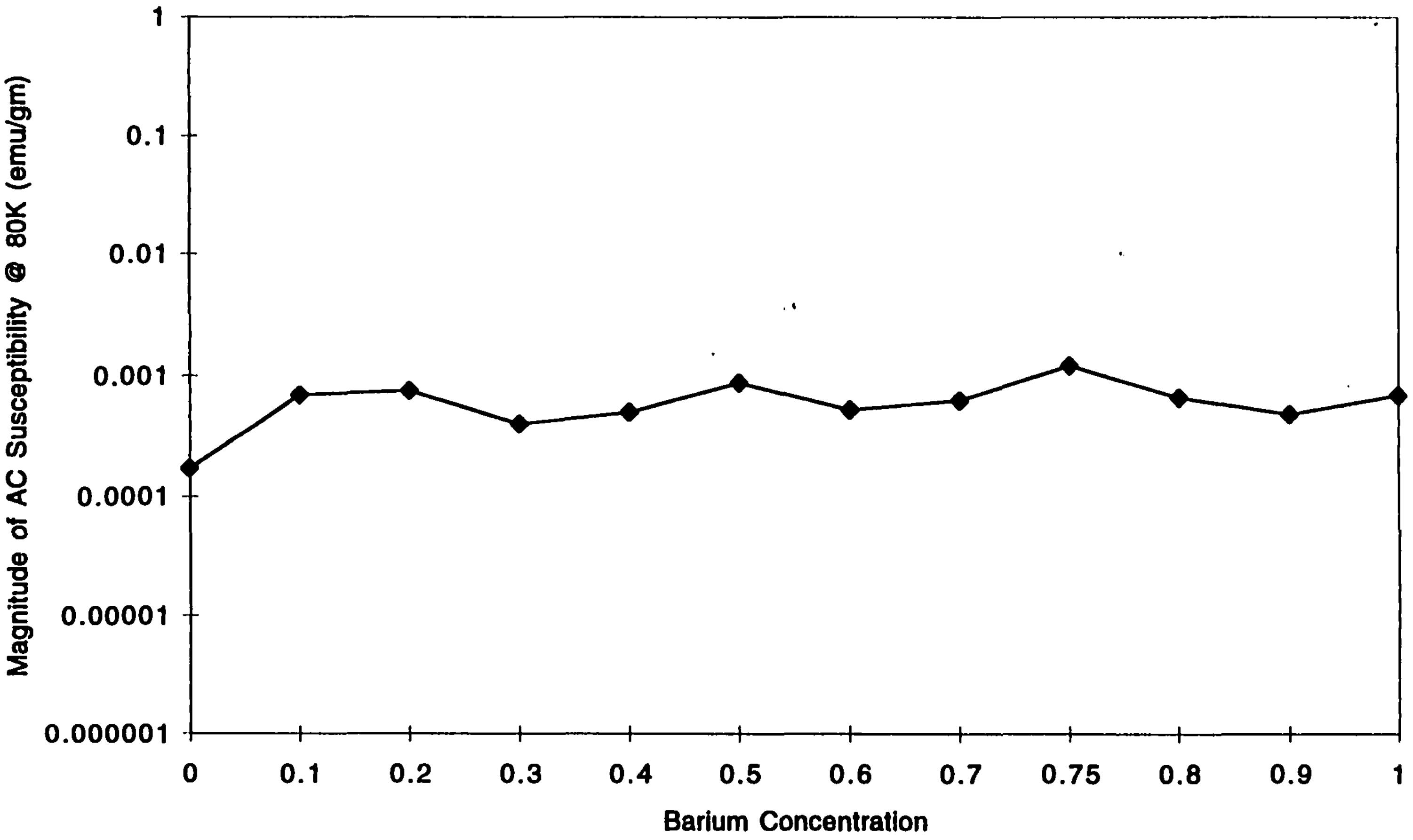

Figure 39. Magnitude of AC Susceptibility Vs Barium Concentration

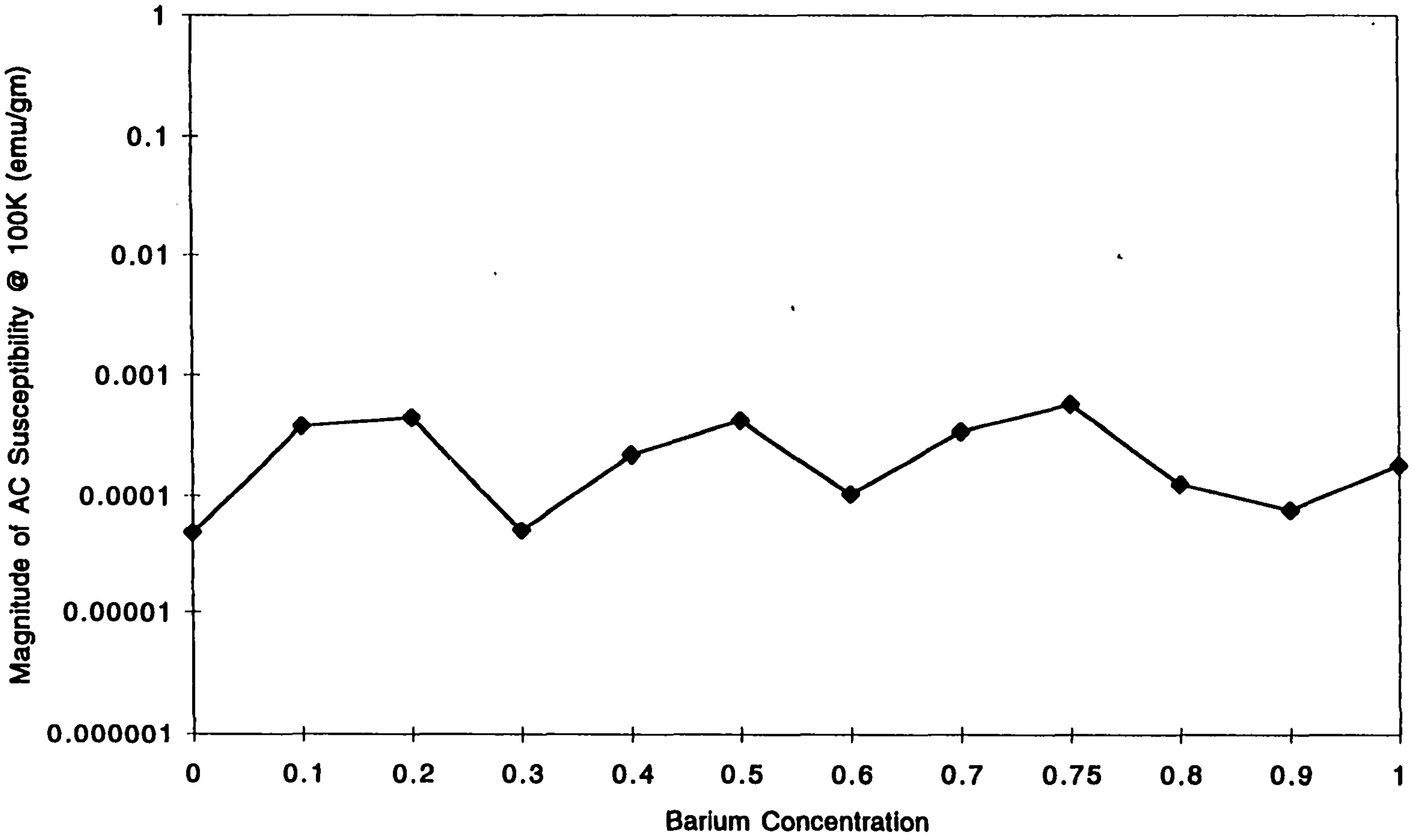

Figure 40. Magnitude of AC Susceptibility Vs Barium Concentration

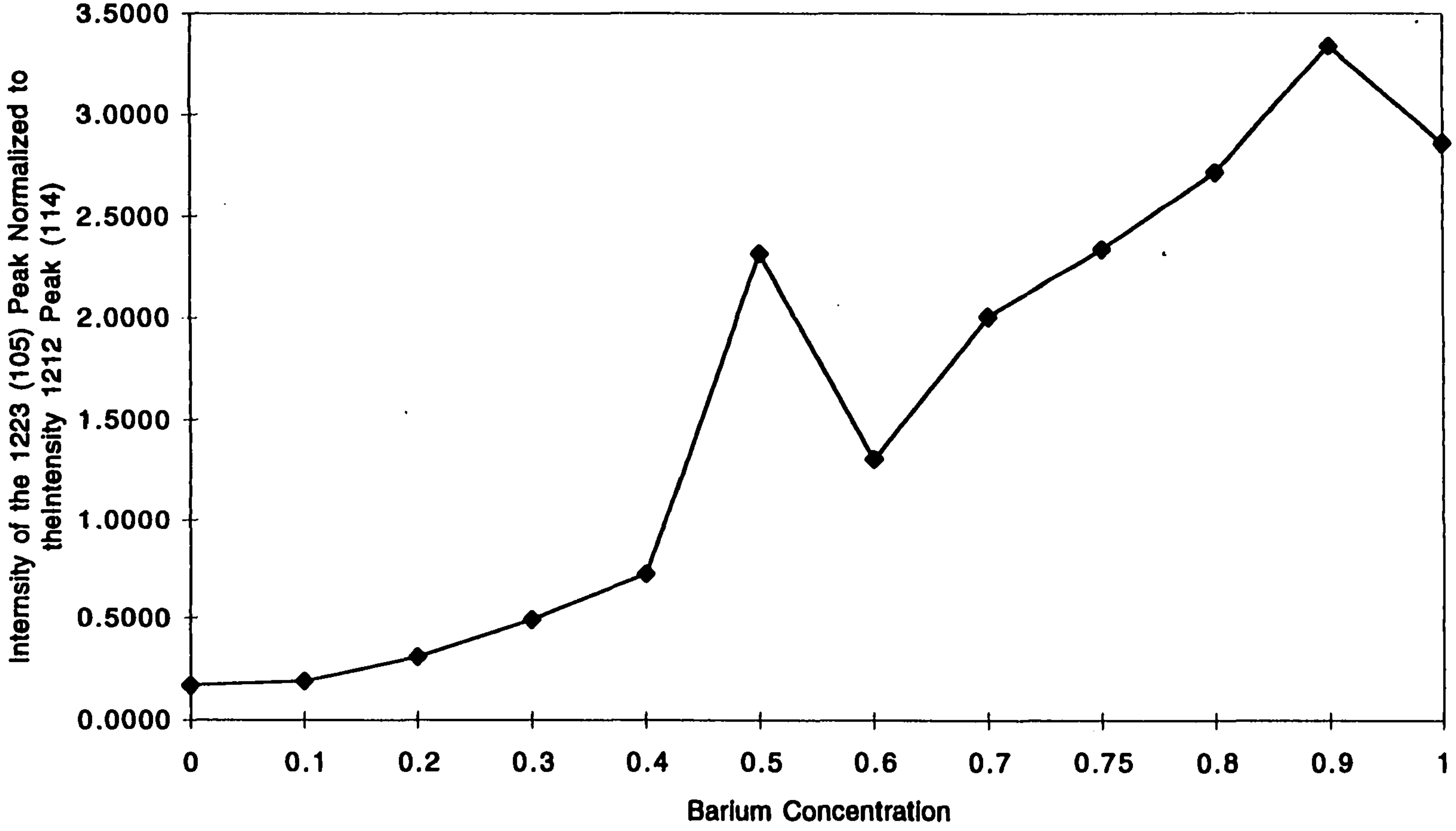

Figure 41. Internsity of 1223(105) Peak Vs Barium Concentration

Eu atoms in the place of Tl and Pb atoms would be different because of alterations in the bonds with oxygen atoms. This gives us a unique opportunity to study the structure of TlPbSrBaCaCuO superconductor precisely. The study in this research was limited to the structural and superconducting properties of the Eu doped TlPbSrBaCaCuO superconductor. The current studies was limited to Eu concentration between 0 and 0.1 in the $\mathbf{(Tl_{0.5-x}Pb_{0.5-x}Eu_{2x})Sr_{1.25}Ba_{0.75}Ca_2Cu_3O_z}$ superconductor. The Figure 42 and Figure 43 show the critical temperature obtained from AC susceptibility measurements vs Eu doping fraction. The Eu concentration in the superconductor increases, the T_C decreases gradually. With the maximum doping fraction of $2x = 0.1$, the T_C dropped close to 100°K from 118°K for $2x = 0.0$.

The magnitude of real part of susceptibility was plotted vs the Eu doping concentration in Figure 44. The magnitude is higher than $1x10^{-3}$ (emu/gm) for Eu = 0, and decreases to $3x10^{-4}$ (emu/gm). A notable observation in the Eu doping superconducting samples is the paramagnetic nature of the samples above the critical temperature. The samples showed the $\chi' > 0$ above T_c, and hence paramagnetism above the T_c. This is why the Eu doped superconductors could reveal exact atomic locations when studied using Mossbour Spectroscopy.

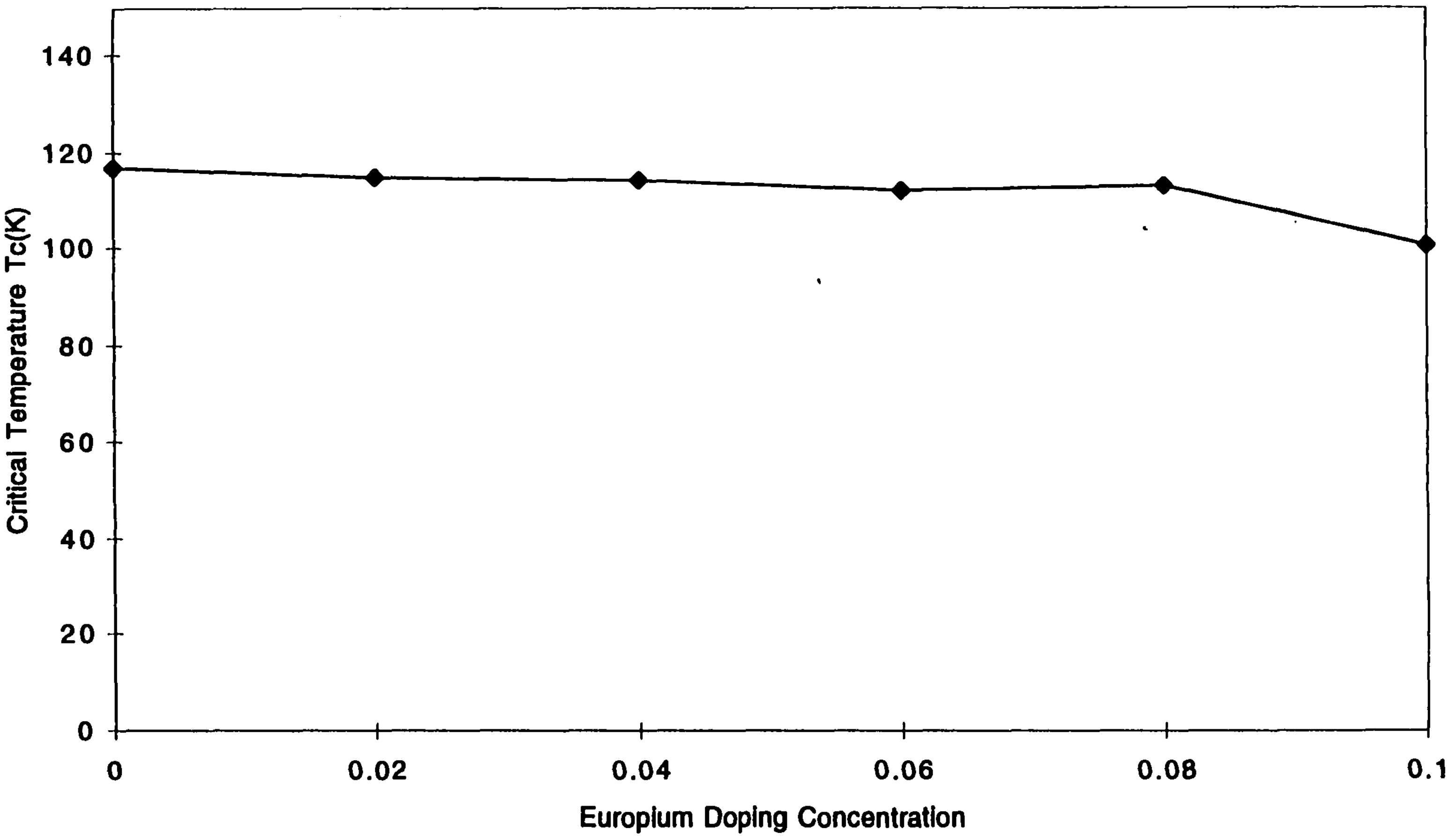

Figure 42. Critical Tc (K) Vs Europium Doping Concentration

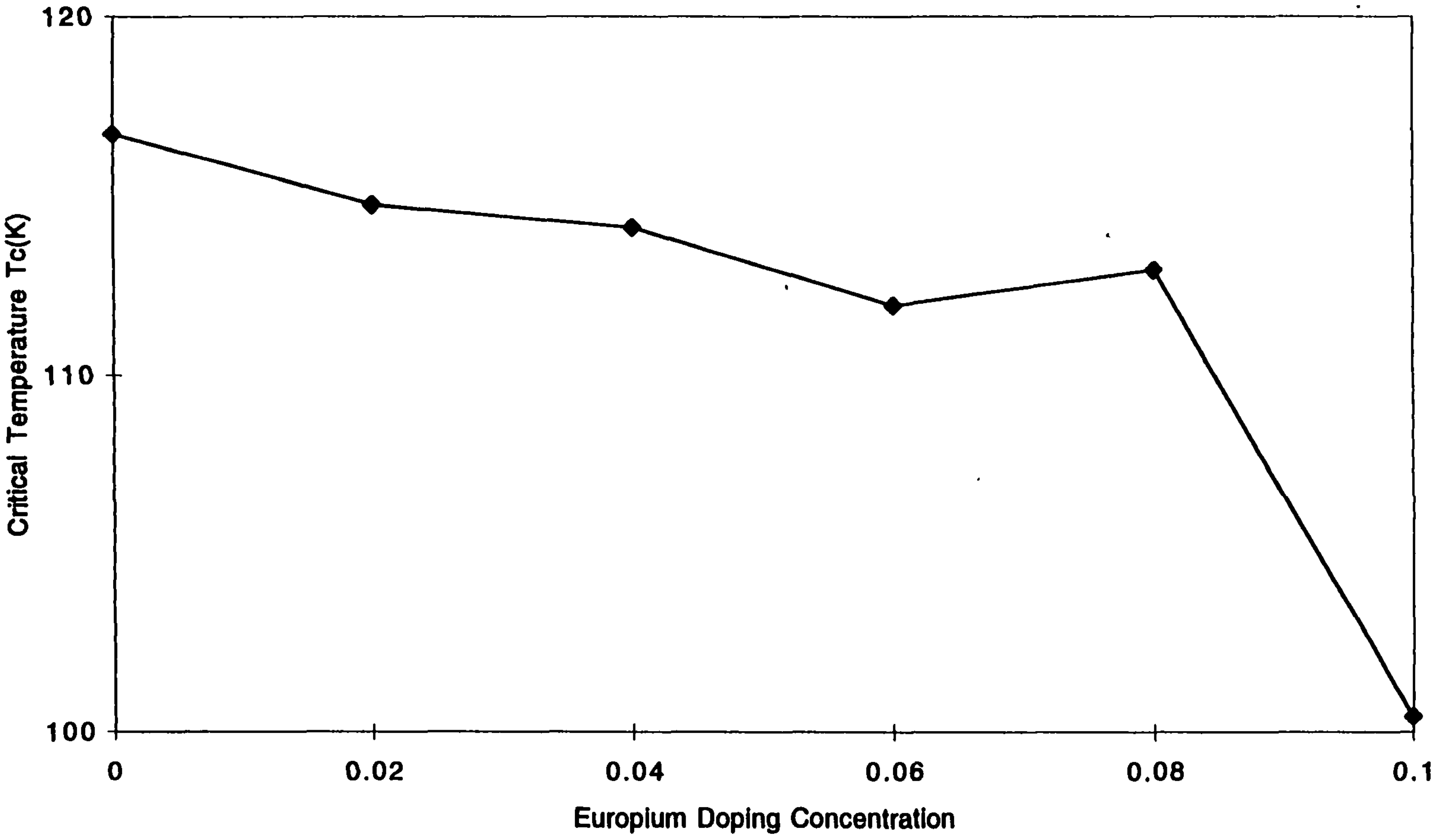

Figure 43. Critical Tc (K) Vs Europium Doping Concentration

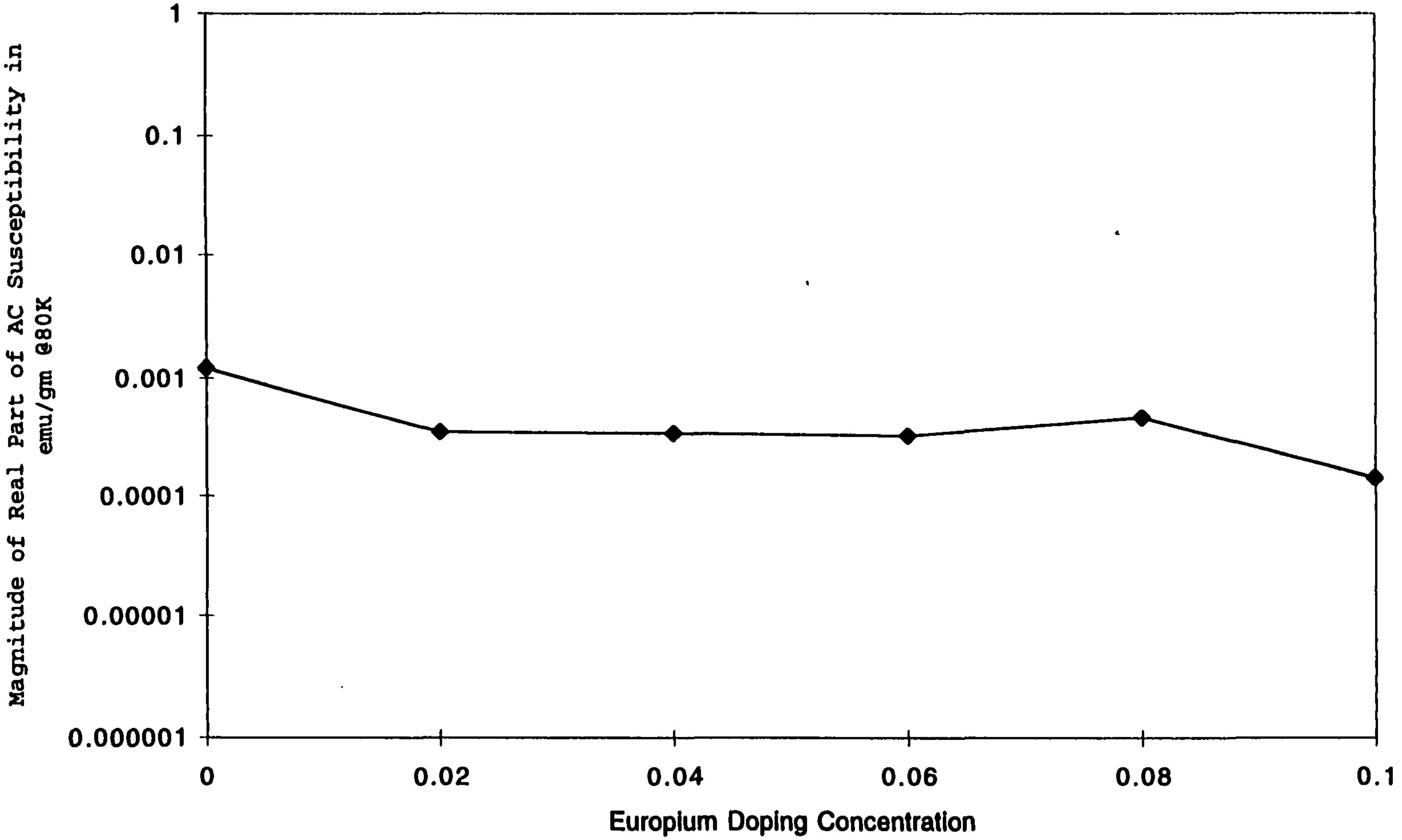

Figure 44. Magnitude of AC Susceptibility Vs Eu Concentration

CHAPTER V

CONCLUSIONS

Summary

Statement of the Problem

The problem of this study was to determine the optimum Barium concentration (y) in the **$(Tl_{0.5-x}Pb_{0.5-x}Eu_{2x})Sr_{2-y}Ba_yCa_2Cu_3O_z$** high temperature superconducting compound to obtain superior superconducting properties and phase purity. In addition, substitution of Eu (Europium) for Tl and Pb was investigated to obtain the optimum Europium concentration (x).

Significance of the Study

As mentioned earlier, the Tl-Pb-Sr-Ca-Cu-O compound superconductors have proven to be superior for electronic applications among the high temperature superconducting materials available (Sheng & Hermann, 1992), because of its lower intrinsic defect structure and strong flux pinning resulting in higher critical current density (Tkaczyk et al., 1992). This research focused on the **$(Tl_{0.5-x}Pb_{0.5-x}Eu_{2x})Sr_{2-y}Ba_yCa_2Cu_3O_z$** compound to determine the optimum y and then study the effects of doping Eu in Tl, Pb sites. Studies on optimum y would give rise to high quality superconducting materials as a target material for sputter deposition of thin films for electronics applications. Studies on Eu doping is a new investigation and might lead to better understanding of structural properties of superconductors.

Methodology

This research followed three procedures. A). Optimizing the sample fabrication process; B). Set Tl/Pb = 1, and determine the optimum y in $(Tl_{0.5}Pb_{0.5})Sr_{2-y}Ba_yCa_2Cu_3O_z$ by trying different y concentration (0.0-1.0); C). Using optimum y to determine the effect of Eu doping in Tl, Pb sites by varying Eu from 1% to 10%.

The precursor (SrBaCaCuO) compounds were made by using high purity BaO_2, CaO, CuO and SrO compounds. Appropriate molar portions of SrO, BaO_2, CaO and CuO were mixed and ground thoroughly. The mixed compounds were heat treated at 900°C for 24 hours and ground again. The same process was repeated again to obtain a homogeneous compound.

Final compounds were made by introducing appropriate amounts of PbO and Tl_2O_3 into precursors. A special die and a hydraulic press were used to apply 15000psi to the powder sample and make pellets. Pellets were sintered in air in a Platinum crucible to make the final product.

After final samples were made, they were characterized by x-ray diffraction (XRD) analysis and ac magnetic susceptibility measurements. Lakeshore Model 7000 AC Susceptometer was used for susceptibility measurement.

Findings

1. Sintering temperature range (850°C-900°C) and sintering time (7-13 minutes) were experimented to optimizing sample fabrication process. Temperature range and sintering time range were decided based on the past research. It was found that optimum sample process condition is 880°C and 10 minutes.

2. Ba doping into $(Tl_{0.5}Pb_{0.5})Sr_{2-x}Ba_xCa_2Cu_3O_z$ compound was experimented with Ba = 0.0 to 1.0. It was found that compound with Ba = 0 created superconducting material in which 1212 phase dominates. Increasing Ba doping, the 1223 phase increased in the material. When Ba = 0.75, 1223 phase dominated the material. As Ba concentration increased beyond 0.75, 1223 phase started reducing, increasing the 1212 phase. Based on XRD charts and AC magnetic susceptibility graphs, it was found that sample with Ba = 0.75 doping gave the best results.

3. Eu doping into $(Tl_{0.5-x}Pb_{0.5-x}Eu_{2x})Sr_{1.25}Ba_{0.75}Ca_2Cu_3O_z$ compound was experimented with Eu = 0% to 10%. Based on XRD and AC magnetic susceptibility graphs, it was found that Eu doping didn't improve the superconducting properties and behaviors. Eu doping increased the tendency to turn a material into paramagnetic material, above the critical temperature.

Conclusions

The following conclusions were made as result of this research:

1. Optimum condition for (TlPbBaSrCaCu) superconducting fabrication process is sintering Tl based material at 880°C for 10 minutes (Research Question #1).

2. Ba doping in the Tl based superconducting materials **$(Tl_{0.5}Pb_{0.5})Sr_{2-x}Ba_xCa_2Cu_3O_z$** provides the best superconducting properties and behavior when Ba = 0.75. The T_c for $(Tl_{0.5}Pb_{0.5})Sr_{1.25}Ba_{0.75}Ca_2Cu_3O_y$ is 118°K. 1223 phase dominates in its microstructure. Figure 37 and Figure 38 shows the relationship between critical temperature (T_c) and Barium concentration in which the highest peak is at Barium concentration Ba = 0.75. In Figure 39 of magnitude of ac susceptibility vs Barium concentration at 80°K, the wave peak appears at Ba = 0.1, Ba = 0.5 and Ba = 0.75 with the highest peak at Ba = 0.75. The trend of intensity of 1223 (105) XRD peak normalized to the maximum intensity peak with the increasing Barium concentration (showed in Figure 41). Except a little jump at Ba = 0.5, XRD intensity increases as Barium concentration increases and turns into the direction of decreasing at Ba = 0.9. The reason that Ba = 0.5 sample shows the good superconducting propertise is properbably this sample was made with over mixed Ba concentration and lead to Ba > 0.5 sample (Research Question #2 & #3).

3. Eu doping created the paramagnetic material with magnetic susceptibility $\chi' \rightarrow$ positive, above T_C. As Eu concentration increases in a compound, the tendency of being paramagnetic is higher. There is a trend of decreasing of critical temperature as Europium doping increases (showed in Figure 42 and 43). As the Europium concentration increases, the magnitude of suscepitibility decreases (Research Question #4).

Recommendations

The following recommendations are provided for future studies or research:

1. The effects caused by the sintering furnace and fume extractor or other concerned environmental conditions should be taken into consideration in the preparation of Tl-based superconductors.

2. Try Eu doping beyond the scope of this research Eu > 0.10.

3. Try different rare earth element doping in Tl-based compound.

4. Apply Mossbaur spectroscopy technique to study microstructure for Eu doping Tl-based superconductor. The paramagnetic properties of the superconductor above Tc will be very useful to determine the site locations of atoms, and the spacing between atoms in the material using the Mossbaur spectroscopy technique.

5. Prepare thin films of **$(Tl_{0.5-x}Pb_{0.5-x}Eu_{2x})Sr_{1.25}Ba_{0.75}Ca_2Cu_3O_z$** superconductor and study its microwave properties and current density.

6. Apply the compound to industrial fields and manufacture some applicable products and find its potential usage in industry.

7. Try different Tl/Pb ratio in (TlPbBaSrCaCu) compounds.

8. Apply high pressure for sample formation. Because the thallium oxide decomposition: $Tl_2O_3(s) \rightarrow Tl_2O(s) + O_2(g)$ and vaporization: $Tl_2O_{(s)} \rightarrow Tl_2O_{(g)}$ can be effectively suppressed by applying a high pressure regardless of the oxygen partial pressure. In addition, high pressure can raise the melting point of Tl_2O_3 so that Tl-based samples can be heated for longer duration at higher temperatures, which lead to the formation of single phase samples with sharper superconducting transition and higher critical current density (Pan, 1993).

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APPENDIX A

AC susceptibility measurement results for Ba Doping

Table 10

(TlPbSrCaCuO) with Ba Doping Ba = 0.00

T	V'(ÊV)	V"(ÊV)	X'	X"
79.46	-15.6472	-20.6357	-2.06E-04	2.72E-04
80.41	-12.6602	-7.8351	-1.67E-04	1.03E-04
81.76	-10.5757	-1.7728	-1.39E-04	2.34E-05
83.03	-9.4282	-0.4697	-1.24E-04	6.19E-06
84.51	-8.5178	-0.1979	-1.12E-04	2.61E-06
85.95	-7.8737	-0.1428	-1.04E-04	1.88E-06
87.31	-7.4388	-0.1333	-9.81E-05	1.76E-06
88.86	-7.0638	-0.1278	-9.32E-05	1.69E-06
90.09	-6.7939	-0.1213	-8.96E-05	1.60E-06
91.57	-6.419	-0.1149	-8.47E-05	1.52E-06
92.47	-6.1489	-0.1144	-8.11E-05	1.51E-06
93.41	-5.939	-0.1077	-7.83E-05	1.42E-06
94.40	-5.699	-0.1067	-7.52E-05	1.41E-06
95.40	-5.444	-0.1039	-7.18E-05	1.37E-06
96.21	-5.2341	-9.69E-02	-6.90E-05	1.28E-06
97.25	-4.949	-9.97E-02	-6.53E-05	1.32E-06
98.22	-4.724	-9.84E-02	-5.93E-05	1.30E-06
99.47	-4.3491	-8.94E-02	-5.35E-05	1.18E-06
100.42	-4.019	-9.09E-02	-4.80E-05	1.20E-06
101.37	-3.764	-8.81E-02	-4.16E-05	1.16E-06
102.28	-3.464	-8.64E-02	-3.27E-05	1.14E-06
103.20	-3.1039	-8.78E-02	-2.29E-05	1.16E-06
104.27	-2.6238	-9.31E-02	-1.66E-05	1.23E-06
105.19	-2.1884	-0.1095	-8.89E-06	1.44E-06
106.08	-1.7381	-0.122	-4.29E-06	1.61E-06
107.12	-1.1532	-0.1149	-1.52E-06	1.52E-06
108.01	0.8083	-0.1019	1.07E-05	1.34E-06
109.01	0.4188	-7.11E-02	1.89E-07	9.37E-07
109.93	0.1492	-4.60E-02	2.17E-07	6.06E-07
110.88	0.075422	-2.39E-02	2.55E-07	3.15E-07
111.87	0.2103	-1.61E-02	2.77E-07	2.13E-07
112.84	0.2553	-1.32E-02	3.37E-07	1.75E-07
114.82	0.3903	-1.57E-02	5.15E-07	2.07E-07
116.76	0.4052	-1.06E-02	5.34E-07	1.40E-07
118.57	0.4052	-8.23E-03	5.34E-07	1.09E-07
120.55	0.4053	-1.33E-02	5.35E-07	1.76E-07

Note. DC:0Oe,F:133,AC:10,Mass:9.92E-02,Units: CGS

Table 11

(TlPbSrCaCuO) with Ba Doping Ba = 0.10

T	V'(ÊV)	V"(ÊV)	X'	X"
80.53	-49.3012	-23.8561	-1.02E-04	1.45E-04
81.35	-42.378	-13.0931	-6.91E-04	1.03E-04
82.71	-33.352	-3.3263	-6.48E-04	5.91E-05
84.00	-28.4815	-1.3075	-5.93E-04	2.52E-05
85.56	-25.2921	-0.9628	-5.61E-04	2.00E-05
86.93	-23.8232	-0.8838	-4.95E-04	1.84E-05
88.22	-23.0736	-0.8534	-4.79E-04	1.77E-05
89.71	-22.384	-0.8263	-4.65E-04	1.72E-05
91.16	-21.8891	-0.8129	-4.55E-04	1.69E-05
92.51	-21.4244	-0.7942	-4.45E-04	1.65E-05
93.37	-21.1096	-0.7812	-4.38E-04	1.62E-05
94.50	-20.6148	-0.7647	-4.28E-04	1.59E-05
95.36	-20.3299	-0.7583	-4.22E-04	1.57E-05
96.24	-19.9551	-0.7436	-4.14E-04	1.54E-05
97.23	-19.6103	-7.30E-01	-4.07E-04	1.51E-05
98.26	-19.1154	-7.17E-01	-3.97E-04	1.49E-05
99.51	-18.5457	-6.95E-01	-3.85E-04	1.44E-05
100.46	-18.0508	-6.85E-01	-3.75E-04	1.42E-05
101.37	-17.511	-6.68E-01	-3.64E-04	1.39E-05
102.28	-16.9412	-6.53E-01	-3.52E-04	1.35E-05
103.24	-16.3113	-6.40E-01	-3.29E-04	1.33E-05
104.31	-15.4714	-6.29E-01	-3.11E-04	1.31E-05
105.21	-14.7364	-6.21E-01	-2.66E-04	1.29E-05
106.13	-13.7615	-0.6076	-2.36E-04	1.26E-05
107.14	-12.501	-0.6226	-1.90E-04	1.29E-05
108.04	-11.0448	-0.6837	-1.29E-04	1.42E-05
109.03	-9.0321	-0.8172	-8.76E-05	1.70E-05
109.93	-6.9907	-8.81E-01	-4.52E-05	1.83E-05
110.92	-4.9818	-7.97E-01	-2.14E-06	1.66E-05
111.88	-3.37E+00	-5.95E-01	-6.99E-06	1.24E-05
112.86	2.1088	-3.77E-01	1.04E-06	7.82E-06
114.89	0.5087	-7.89E-02	1.36E-06	1.64E-06
116.78	7.53E-02	-1.77E-02	1.56E-06	3.68E-07
118.62	7.54E-02	-2.00E-02	1.56E-06	4.15E-07
120.61	3.02E-02	-1.36E-02	6.28E-07	2.82E-07
122.53	4.53E-02	-1.66E-02	9.41E-07	3.45E-07

Note. DC:0Oe,F:133,AC:10,Mass:6.30E-02,Units:CGS

Table 12

(TlPbSrCaCuO) with Ba Doping Ba = 0.20

T	V'(ÊV)	V"(ÊV)	X'	X"
78.46	-461.056	-484.72	-1.21E-03	9.03E-04
79.31	-296.297	-378.359	-9.10E-04	5.10E-04
80.66	-144.612	-189.963	-7.95E-04	3.06E-04
82.23	-82.1979	-43.8117	-7.32E-04	1.42E-04
83.46	-69.6105	-9.4609	-6.90E-04	8.02E-05
85.06	-61.8462	-0.4724	-6.70E-04	5.12E-06
86.39	-58.7704	0.3714	-6.36E-04	-4.02E-06
87.72	-56.9292	0.604	-6.17E-04	-6.54E-06
89.27	-55.3701	0.6668	-6.00E-04	-7.22E-06
90.83	-53.7668	0.7798	-5.82E-04	-8.44E-06
91.63	-52.9574	0.8239	-5.74E-04	-8.92E-06
92.67	-51.9975	0.8362	-5.63E-04	-9.06E-06
93.53	-51.1729	0.8675	-5.54E-04	-9.40E-06
94.52	-50.273	0.8788	-5.44E-04	-9.52E-06
95.55	-49.298	8.85E-01	-5.34E-04	-9.58E-06
96.43	-48.278	8.98E-01	-5.23E-04	-9.73E-06
97.43	-46.3873	8.74E-01	-5.02E-04	-9.47E-06
98.53	-45.4266	8.41E-01	-4.92E-04	-9.11E-06
99.63	-43.7908	8.07E-01	-4.74E-04	-8.74E-06
100.56	-42.2905	8.04E-01	-4.58E-04	-8.70E-06
101.47	-40.5487	7.16E-01	-4.39E-04	-7.76E-06
102.42	-38.6727	6.75E-01	-4.19E-04	-7.31E-06
103.34	-36.5855	5.69E-01	-3.96E-04	-6.16E-06
104.41	-33.9133	0.4647	-3.67E-04	-5.03E-06
105.33	-31.1192	0.2569	-3.37E-04	-2.78E-06
106.22	-27.8748	3.10E-02	-3.02E-04	-3.36E-07
107.27	-23.3371	-0.3708	-2.53E-04	4.02E-06
108.14	-18.7535	-8.22E-01	-2.03E-04	8.90E-06
109.16	-13.6172	-1.14E+00	-1.47E-04	1.23E-05
110.06	-9.61E+00	-1.05E+00	-8.04E-05	1.14E-05
111.04	-6.2121	-7.92E-01	-2.97E-05	8.57E-06
112.03	-3.8172	-4.81E-01	-4.13E-06	5.21E-06
113.02	2.14E+00	-2.31E-01	2.32E-05	2.51E-06
114.88	2.39E-01	-4.17E-02	2.59E-06	4.51E-07
116.76	4.21E-01	-4.72E-02	4.56E-06	5.11E-07
118.65	4.66E-01	-4.84E-02	5.05E-06	5.24E-07

Note. DC:0Oe,F:133,AC:10,Mass:1.21E-01,Units:CGS

Table 13

(TlPbSrCaCuO) with Ba Doping Ba = 0.30

T	V'(ÊV)	V"(ÊV)	X'	X"
80.88	-14.2385	-17.9737	-3.57E-04	4.02E-04
81.78	-10.9779	-4.5122	-2.71E-04	2.01E-04
83.09	-7.8809	-0.5878	-2.07E-04	7.82E-05
84.67	-5.8917	-0.2382	-1.32E-04	1.02E-05
86.09	-5.2024	-0.193	-9.81E-05	4.13E-06
87.37	-4.7978	-0.1696	-8.81E-05	3.34E-06
88.95	-4.5429	-0.1582	-8.27E-05	2.94E-06
90.32	-4.303	-0.1506	-7.96E-05	2.74E-06
90.89	-4.213	-0.1505	-7.50E-05	2.61E-06
91.75	-4.0781	-0.1454	-7.07E-05	2.52E-06
92.80	-3.9431	-0.1431	-6.83E-05	2.48E-06
93.62	-3.8531	-0.1405	-6.68E-05	2.43E-06
94.63	-3.6733	-0.1309	-6.37E-05	2.27E-06
95.62	-3.5233	-0.1274	-6.11E-05	2.21E-06
96.48	-3.3733	-1.25E-01	-5.85E-05	2.16E-06
97.45	-3.2384	-1.20E-01	-5.61E-05	2.08E-06
98.50	-3.0134	-1.16E-01	-5.22E-05	2.01E-06
99.73	-2.8485	-1.12E-01	-4.94E-05	1.94E-06
100.68	-2.6535	-1.08E-01	-4.60E-05	1.87E-06
101.61	-2.4736	-1.02E-01	-4.29E-05	1.76E-06
102.56	-2.3087	-9.72E-02	-4.00E-05	1.68E-06
103.52	-2.0836	-9.62E-02	-3.61E-05	1.67E-06
104.59	-1.8437	-8.96E-02	-3.19E-05	1.55E-06
105.49	-1.5887	-8.64E-02	-2.75E-05	1.50E-06
106.42	-1.3637	-8.88E-02	-2.36E-05	1.54E-06
107.43	-1.0337	-8.14E-02	-1.79E-05	1.41E-06
108.34	-0.8538	-7.72E-02	-1.48E-05	1.34E-06
109.34	-0.539	-6.40E-02	-9.34E-06	1.11E-06
110.24	-0.3442	-4.92E-02	-5.96E-06	8.53E-07
111.24	-1.20E-01	-2.94E-02	-2.07E-06	5.09E-07
112.22	1.52E-02	-1.40E-02	2.64E-07	2.42E-07
113.21	0.195	4.04E-04	3.38E-07	-6.99E-09
115.20	2.70E-01	3.96E-03	4.68E-07	-6.87E-08
117.07	3.60E-01	4.03E-03	6.24E-07	-6.99E-08
119.00	3.60E-01	7.03E-03	6.24E-07	-1.22E-07
120.97	3.60E-01	6.88E-03	6.24E-07	-1.19E-07

Note. DC:0Oe,F:133,AC:10,Mass:7.55E-02,Units: CGS

Table 14

(TlPbSrCaCuO) with Ba Doping Ba = 0.40

T	V'(ÊV)	V"(ÊV)	X'	X"
80.86	-18.61	-8.1863	-5.09E-04	2.24E-04
81.82	-16.5252	-4.7171	-4.52E-04	1.29E-04
83.18	-14.88	-2.7108	-4.07E-04	7.42E-05
84.53	-13.9317	-2.0315	-3.81E-04	5.56E-05
86.01	-13.4125	-1.6903	-3.67E-04	4.63E-05
87.42	-12.9366	-1.4562	-3.54E-04	3.99E-05
88.86	-12.5347	-1.275	-3.43E-04	3.49E-05
90.22	-12.1624	-1.1151	-3.33E-04	3.05E-05
91.52	-11.8046	-0.9828	-3.23E-04	2.69E-05
92.57	-11.5062	-0.8901	-3.15E-04	2.44E-05
93.45	-11.1927	-0.8044	-3.06E-04	2.20E-05
94.50	-10.8489	-0.7294	-2.97E-04	2.00E-05
95.47	-10.5802	-0.6527	-2.90E-04	1.79E-05
96.30	-10.2216	-0.5695	-2.80E-04	1.56E-05
97.21	-9.9073	-5.26E-01	-2.71E-04	1.44E-05
98.26	-9.5029	-4.89E-01	-2.60E-04	1.34E-05
99.53	-8.889	-4.20E-01	-2.43E-04	1.15E-05
100.47	-8.3352	-3.49E-01	-2.28E-04	9.54E-06
101.38	-7.8394	-3.87E-01	-2.15E-04	1.06E-05
102.33	-7.1348	-3.61E-01	-1.95E-04	9.88E-06
103.26	-6.5499	-3.52E-01	-1.79E-04	9.63E-06
104.32	-5.5896	-3.58E-01	-1.53E-04	9.80E-06
105.26	-4.7794	-3.62E-01	-1.31E-04	9.91E-06
106.15	-4.0149	-3.28E-01	-1.10E-04	8.97E-06
107.17	-3.0096	-3.33E-01	-8.24E-05	9.12E-06
108.07	-2.2899	-3.14E-01	-6.27E-05	8.59E-06
109.08	-1.5568	-1.99E-01	-4.26E-05	5.45E-06
109.99	-1.0784	-9.88E-02	-2.95E-05	2.71E-06
110.95	-0.6893	-4.60E-02	-1.89E-05	1.26E-06
111.93	-2.85E-01	5.02E-03	-7.80E-06	-1.38E-07
112.89	-1.20E-01	1.79E-02	-3.29E-06	-4.90E-07
114.96	4.45E-02	3.13E-02	1.22E-07	-8.56E-07
116.84	8.96E-02	2.16E-02	2.45E-07	-5.90E-07
118.68	2.96E-02	2.40E-02	8.10E-07	-6.57E-07
120.73	-7.77E-04	4.45E-02	-2.13E-08	-1.22E-06
122.62	-1.51E-02	3.24E-03	-4.12E-07	-8.86E-08

Note. DC:0Oe,F:133,AC:10,Mass:4.78E-02,Units:CGS

Table 15

(TlPbSrCaCuO) with Ba Doping Ba = 0.50

T	V'(ÊV)	V"(ÊV)	X'	X"
82.09	-149.886	-86.059	-8.35E-04	8.88E-04
82.89	-142.153	-76.6976	-8.21E-04	7.81E-04
84.51	-130.349	-61.6643	-7.99E-04	6.08E-04
85.87	-124.204	-53.5508	-7.61E-04	5.15E-04
87.33	-118.181	-45.6795	-7.31E-04	4.25E-04
88.82	-112.806	-38.7321	-6.96E-04	3.45E-04
90.22	-108.062	-32.6194	-6.85E-04	2.75E-04
90.75	-106.35	-30.1686	-6.68E-04	2.47E-04
91.61	-103.676	-26.9205	-6.52E-04	2.09E-04
92.57	-100.92	-23.2373	-6.44E-04	1.67E-04
93.47	-98.3805	-20.028	-6.29E-04	1.30E-04
94.50	-95.5736	-16.6749	-6.16E-04	9.15E-05
95.47	-92.6857	-13.6604	-6.01E-04	5.89E-05
96.33	-90.1484	-11.1907	-5.86E-04	3.05E-05
97.29	-87.1737	-8.85E+00	-5.70E-04	2.93E-05
98.35	-83.5689	-6.52E+00	-5.41E-04	2.90E-05
99.45	-79.2842	-4.46E+00	-5.11E-04	2.79E-05
100.46	-74.9575	-3.09E+00	-4.51E-04	2.57E-05
101.38	-69.6495	-2.07E+00	-4.00E-04	2.38E-05
102.31	-63.8825	-1.56E+00	-3.31E-04	1.79E-05
103.26	-56.6074	-1.50E+00	-2.67E-04	1.73E-05
104.31	-47.1964	-1.76E+00	-1.95E-04	2.03E-05
105.26	-37.9045	-2.08E+00	-1.29E-04	2.39E-05
106.13	-29.261	-2.20E+00	-8.96E-05	2.53E-05
107.15	-20.9832	-2.01E+00	-6.41E-05	2.31E-05
108.07	-14.9741	-1.62E+00	-4.38E-05	1.86E-05
109.11	-9.7478	-1.07E+00	-3.09E-05	1.23E-05
110.01	-5.8403	-6.10E-01	-2.00E-05	7.00E-06
110.97	-2.7852	-2.97E-01	-8.01E-06	3.41E-06
112.00	-7.94E-01	-8.24E-02	-4.12E-06	9.46E-07
112.96	9.14E-02	-7.89E-02	1.05E-06	9.06E-07
115.10	4.36E-01	-4.49E-02	5.01E-06	5.16E-07
116.96	4.66E-01	-2.64E-02	5.35E-06	3.03E-07
118.84	5.25E-01	-1.93E-02	6.04E-06	2.22E-07
120.88	4.66E-01	-3.64E-02	5.35E-06	4.18E-07
122.74	5.41E-01	-6.81E-02	6.22E-06	7.82E-07

Note. DC:0Oe,F:133,AC:10,Mass:1.14E-01,Units:CGS

Table 16

(TlPbSrCaCuO) with Ba Doping Ba = 0.60

T	V'(ÊV)	V"(ÊV)	X'	X"
80.90	-11.6072	-1.1192	-4.61E-04	4.38E-05
81.82	-11.2641	-1.0082	-4.50E-04	4.05E-05
83.26	-10.6965	-0.8693	-4.23E-04	3.63E-05
84.51	-10.3378	-0.787	-4.02E-04	3.38E-05
86.15	-9.8145	-0.6859	-3.77E-04	3.07E-05
87.46	-9.5006	-0.6219	-3.67E-04	2.88E-05
89.05	-9.0519	-0.5451	-3.53E-04	2.65E-05
90.41	-8.6478	-0.491	-3.41E-04	2.48E-05
90.98	-8.438	-0.4768	-3.35E-04	2.24E-05
91.77	-8.2134	-0.4514	-3.28E-04	1.96E-05
92.78	-7.8238	-0.4216	-3.16E-04	1.57E-05
93.72	-7.4791	-0.4016	-3.06E-04	1.21E-05
94.73	-7.0745	-0.3745	-2.84E-04	1.13E-05
95.79	-6.5047	-0.3611	-2.67E-04	1.09E-05
96.52	-6.19	-3.43E-01	-2.37E-04	1.04E-05
97.55	-5.6049	-3.39E-01	-2.09E-04	1.03E-05
98.59	-4.9751	-3.22E-01	-1.70E-04	9.74E-06
99.85	-4.1201	-3.16E-01	-1.34E-04	9.55E-06
100.81	-3.4002	-3.05E-01	-1.03E-04	9.21E-06
101.73	-2.6955	-2.83E-01	-8.14E-05	8.55E-06
102.68	-2.0361	-2.40E-01	-6.15E-05	7.25E-06
103.64	-1.437	-1.84E-01	-4.34E-05	5.55E-06
104.69	-0.8832	-1.13E-01	-2.67E-05	3.43E-06
105.60	-0.554	-6.37E-02	-1.67E-05	1.92E-06
106.51	-0.2845	-2.85E-02	-8.60E-06	8.60E-07
107.55	-0.1348	-1.39E-02	-4.07E-06	4.19E-07
108.44	-1.49E-02	-4.26E-03	-4.51E-07	1.29E-07
109.49	4.50E-02	-2.15E-04	1.36E-07	6.48E-09
110.38	4.50E-02	1.29E-03	1.36E-07	-3.88E-08
111.36	1.50E-02	2.62E-04	4.53E-07	-7.91E-09
112.33	1.50E-02	-7.38E-04	4.54E-07	2.23E-08
113.32	4.50E-02	2.85E-04	1.36E-06	-8.63E-09
115.34	4.51E-02	-2.71E-03	1.36E-06	8.20E-08
117.21	4.50E-02	-2.14E-04	1.36E-06	6.48E-09
119.11	4.50E-02	7.85E-04	1.36E-06	-2.37E-08
121.14	3.00E-02	-1.48E-03	9.07E-07	4.46E-08

Note. DC:0Oe,F:133,AC:10,Mass:4.33E-02,Units:CGS

Table 17

(TlPbSrCaCuO) with Ba Doping Ba = 0.70

T	V'(ÊV)	V"(ÊV)	X'	X"
81.27	-8.4363	-0.5703	-6.45E-04	2.54E-05
82.23	-7.8672	-0.5164	-6.30E-04	2.30E-05
83.61	-7.5079	-0.4746	-6.14E-04	2.11E-05
85.22	-7.6885	-0.4407	-6.20E-04	1.96E-05
86.63	-6.9241	-0.3969	-5.88E-04	1.77E-05
87.80	-7.1344	-0.3821	-5.98E-04	1.70E-05
89.37	-6.7749	-0.3528	-5.88E-04	1.57E-05
90.75	-6.5502	-0.3339	-5.72E-04	1.49E-05
91.73	-5.9054	-0.3146	-5.63E-04	1.40E-05
92.67	-6.0854	-0.3168	-5.21E-04	1.41E-05
93.53	-5.9356	-0.3061	-5.14E-04	1.36E-05
94.54	-5.2308	-0.2858	-4.63E-04	1.27E-05
95.55	-4.9157	-0.2868	-4.59E-04	1.28E-05
96.39	-5.0358	-0.2844	-4.34E-04	1.27E-05
97.34	-4.7057	-2.85E-01	-4.19E-04	1.27E-05
98.39	-4.0458	-2.78E-01	-3.80E-04	1.24E-05
99.65	-3.5207	-2.75E-01	-3.57E-04	1.22E-05
100.61	-3.1457	-2.74E-01	-3.40E-04	1.22E-05
101.54	-2.7707	-2.68E-01	-3.23E-04	1.19E-05
102.45	-2.3808	-2.61E-01	-3.06E-04	1.16E-05
103.40	-1.991	-2.49E-01	-2.46E-04	1.11E-05
104.45	-1.5563	-2.24E-01	-2.29E-04	9.95E-06
105.37	-1.5118	-1.94E-01	-1.47E-04	8.63E-06
106.28	-0.7476	-1.44E-01	-9.83E-05	6.39E-06
107.30	-0.3434	-9.50E-02	-6.53E-05	4.23E-06
108.19	-5.92E-02	-4.65E-02	-2.63E-06	2.07E-06
109.19	-6.04E-02	-2.00E-02	-2.09E-06	8.88E-07
110.09	-2.40E-01	2.19E-03	-1.07E-06	-9.74E-08
111.08	2.99E-02	-4.02E-03	1.33E-06	1.79E-07
112.06	3.45E-01	6.52E-03	1.53E-06	-2.90E-07
113.05	4.20E-01	7.33E-03	6.87E-06	-3.26E-07
115.07	3.30E-01	4.26E-03	1.47E-06	-1.90E-07
116.95	-1.50E-02	7.38E-04	-6.68E-07	-3.28E-08
118.84	3.45E-01	5.52E-03	5.54E-06	-2.46E-07
120.76	3.75E-01	3.05E-03	1.67E-06	-1.36E-07
122.71	3.60E-01	4.28E-03	8.60E-06	-1.91E-07

Note. DC:0Oe,F:133,AC:10,Mass:2.94E-02,Units: CGS

Table 18

(TlPbSrCaCuO) with Ba Doping Ba = 0.75

T	V'(ÊV)	V"(ÊV)	X'	X"
83.98	-120.159	-7.4232	-7.96E-04	4.92E-05
84.63	-118.515	-7.0658	-7.85E-04	4.68E-05
85.97	-115.073	-6.5513	-7.62E-04	4.34E-05
87.17	-112.661	-6.3717	-7.46E-04	4.22E-05
88.78	-109.559	-6.175	-7.26E-04	4.09E-05
89.97	-107.596	-6.0647	-7.13E-04	4.02E-05
90.60	-106.367	-5.9918	-7.05E-04	3.97E-05
91.39	-105.003	-5.9104	-6.96E-04	3.92E-05
92.32	-103.234	-5.8221	-6.84E-04	3.86E-05
93.20	-101.511	-5.7055	-6.72E-04	3.78E-05
94.25	-99.3826	-5.5883	-6.58E-04	3.70E-05
95.25	-97.359	-5.4885	-6.45E-04	3.64E-05
96.08	-95.4705	-5.3895	-6.32E-04	3.57E-05
97.03	-93.3571	-5.2771	-6.18E-04	3.50E-05
98.06	-90.7641	-5.14E+00	-6.01E-04	3.41E-05
99.53	-87.0169	-4.95E+00	-5.76E-04	3.28E-05
100.46	-84.1989	-4.81E+00	-5.58E-04	3.19E-05
101.37	-81.3509	-4.67E+00	-5.39E-04	3.09E-05
102.28	-78.2028	-4.53E+00	-5.18E-04	3.00E-05
103.22	-74.4846	-4.40E+00	-4.93E-04	2.91E-05
104.27	-69.5052	-4.32E+00	-4.60E-04	2.86E-05
105.21	-63.4866	-4.48E+00	-4.21E-04	2.96E-05
106.06	-55.7685	-4.87E+00	-3.69E-04	3.23E-05
107.10	-45.8	-5.27E+00	-3.03E-04	3.49E-05
107.99	-37.7135	-5.28E+00	-2.50E-04	3.50E-05
108.99	-2.96E+01	-4.95E+00	-1.96E-04	3.28E-05
109.88	-2.32E+01	-4.45E+00	-1.54E-04	2.95E-05
110.86	-1.71E+01	-3.72E+00	-1.13E-04	2.47E-05
111.82	-1.15E+01	-2.74E+00	-7.61E-05	1.81E-05
112.80	-6.84E+00	-1.60E+00	-4.53E-05	1.06E-05
114.77	-1.20E+00	-1.67E-01	-7.93E-06	1.11E-06
116.67	2.25E-01	-2.07E-03	1.49E-06	1.37E-08
118.53	2.55E-01	-4.92E-05	1.69E-06	3.26E-10
120.49	2.10E-01	-1.33E-03	1.39E-06	8.84E-09
122.38	2.40E-01	1.69E-03	1.59E-06	-1.12E-08

Note. DC:0Oe,F:133,AC:10,Mass:1.98E-01,Units:CGS

Table 19

(TlPbSrCaCuO) with Ba Doping Ba = 0.8

T	V'(ÊV)	V"(ÊV)	X'	X"
81.19	-56.4888	-25.4862	-6.84E-04	2.59E-04
81.78	-55.0017	-23.8745	-6.59E-04	2.43E-04
83.18	-51.6561	-20.4411	-6.15E-04	2.08E-04
84.53	-48.6934	-17.4185	-5.55E-04	1.77E-04
86.11	-45.524	-14.2042	-5.22E-04	1.44E-04
87.31	-43.4014	-12.045	-4.81E-04	1.22E-04
88.90	-40.5231	-9.3424	-4.42E-04	9.49E-05
90.18	-38.2151	-7.491	-3.98E-04	7.61E-05
91.67	-35.2196	-5.485	-3.58E-04	5.57E-05
92.67	-33.2723	-4.4569	-3.38E-04	4.53E-05
93.60	-31.1736	-3.5016	-3.17E-04	3.56E-05
94.54	-29.0413	-2.7598	-2.95E-04	2.80E-05
95.57	-26.6509	-2.1885	-2.71E-04	2.22E-05
96.41	-24.408	-1.7612	-2.48E-04	1.79E-05
97.38	-21.803	-1.45E+00	-2.21E-04	1.47E-05
98.43	-18.7319	-1.20E+00	-1.90E-04	1.22E-05
99.67	-15.1644	-1.03E+00	-1.54E-04	1.04E-05
100.67	-12.3903	-9.51E-01	-1.26E-04	9.66E-06
101.58	-9.842	-8.31E-01	-1.00E-04	8.44E-06
102.50	-7.5041	-6.88E-01	-7.62E-05	6.98E-06
103.47	-5.3913	-5.46E-01	-5.48E-05	5.54E-06
104.52	-3.1901	-3.09E-01	-3.24E-05	3.14E-06
105.46	-1.6324	-1.66E-01	-1.66E-05	1.68E-06
106.35	-0.4943	-4.71E-02	-5.02E-06	4.79E-07
107.38	-0.166	-5.41E-02	-8.19E-07	5.50E-07
108.29	-4.21E-01	-6.97E-02	-6.28E-07	7.08E-07
109.31	-5.41E-01	-3.06E-02	-4.49E-07	3.11E-07
110.21	5.26E-01	-2.23E-02	1.34E-06	2.27E-07
111.18	6.01E-01	-6.20E-02	2.11E-06	6.30E-07
112.19	5.56E-01	-3.33E-02	1.64E-06	3.38E-07
113.16	6.16E-01	-3.73E-02	2.25E-06	3.78E-07
115.19	6.31E-01	-4.70E-02	2.41E-06	4.77E-07
117.03	6.30E-01	-4.00E-03	2.40E-06	4.07E-08
119.02	5.86E-01	-5.13E-02	1.95E-06	5.21E-07
120.93	6.46E-01	-5.62E-02	2.56E-06	5.71E-07
122.88	6.31E-01	-2.90E-02	2.41E-06	2.95E-07

Note. DC:00e,F:133,AC:10,Mass:1.29E-01,Units:CGS

Table 20

(TlPbSrCaCuO) with Ba Doping Ba = 0.9

T	V'(ÊV)	V"(ÊV)	X'	X"
81.72	-25.3072	-4.3913	-4.58E-04	5.17E-05
82.36	-24.8184	-4.0332	-4.49E-04	4.75E-05
83.63	-23.7799	-3.3673	-4.34E-04	3.96E-05
85.14	-22.6063	-2.7034	-4.12E-04	3.18E-05
86.53	-21.5507	-2.1519	-3.83E-04	2.53E-05
87.78	-20.6123	-1.7655	-3.53E-04	2.08E-05
89.35	-19.2999	-1.3225	-3.27E-04	1.56E-05
90.83	-18.0906	-0.9824	-3.03E-04	1.16E-05
91.61	-17.3131	-0.8338	-2.84E-04	9.81E-06
92.65	-16.2806	-0.6797	-2.72E-04	8.00E-06
93.58	-15.3223	-0.574	-2.50E-04	6.75E-06
94.58	-14.2734	-0.5012	-2.28E-04	5.90E-06
95.64	-13.029	-0.4555	-1.93E-04	5.36E-06
96.37	-12.1442	-0.4395	-1.63E-04	5.17E-06
97.38	-10.8092	-4.30E-01	-1.37E-04	5.06E-06
98.44	-9.3536	-4.50E-01	-1.10E-04	5.30E-06
99.67	-7.778	-4.71E-01	-9.15E-05	5.54E-06
100.65	-6.4729	-4.63E-01	-7.62E-05	5.44E-06
101.59	-5.2282	-4.36E-01	-6.15E-05	5.13E-06
102.54	-4.014	-3.78E-01	-4.72E-05	4.45E-06
103.47	-2.8602	-2.98E-01	-3.37E-05	3.51E-06
104.57	-1.6621	-1.79E-01	-1.96E-05	2.11E-06
105.49	-0.8536	-8.79E-02	-1.00E-05	1.03E-06
106.42	-0.2693	-3.97E-02	-3.17E-06	4.67E-07
107.45	0.1354	-2.11E-02	5.19E-07	2.49E-07
108.37	3.90E-01	-7.69E-03	5.49E-07	9.05E-08
109.41	5.10E-01	-4.10E-03	8.00E-07	4.82E-08
110.30	5.55E-01	-1.03E-02	8.53E-07	1.21E-07
111.31	5.55E-01	-8.12E-04	8.53E-07	9.56E-09
112.32	6.15E-01	-7.26E-03	1.24E-06	8.55E-08
113.32	6.15E-01	-4.26E-03	1.24E-06	5.02E-08
115.28	6.30E-01	-9.50E-03	1.42E-06	1.12E-07
117.27	6.00E-01	-7.53E-03	1.06E-06	8.85E-08
119.12	6.45E-01	-7.74E-03	1.59E-06	9.11E-08
121.14	6.00E-01	-6.03E-03	1.06E-06	7.09E-08
123.12	6.15E-01	-1.03E-02	1.24E-06	1.21E-07

Note. DC:0Oe,F:133,AC:10,Mass:1.11E-01,Units:CGS

Table 21

(TlPbSrCaCuO) with Ba Doping Ba = 1.0

T	V'(ÊV)	V"(ÊV)	X'	X"
81.91	-46.225	-24.755	-6.60E-04	4.19E-04
82.71	-44.5015	-22.9366	-6.33E-04	3.88E-04
84.10	-41.4787	-19.9194	-5.92E-04	3.37E-04
85.85	-37.8197	-16.39	-5.50E-04	2.77E-04
87.09	-35.6676	-14.1996	-5.24E-04	2.40E-04
88.42	-33.35	-12.0359	-4.94E-04	2.04E-04
90.01	-30.6445	-9.7518	-4.79E-04	1.65E-04
91.40	-28.2004	-7.9608	-4.67E-04	1.35E-04
91.88	-27.3865	-7.317	-4.54E-04	1.24E-04
92.82	-25.7833	-6.3386	-4.36E-04	1.07E-04
93.68	-24.3273	-5.5252	-4.12E-04	9.35E-05
94.77	-22.3331	-4.6039	-3.78E-04	7.79E-05
95.79	-20.4708	-3.8591	-3.46E-04	6.53E-05
96.55	-19.0389	-3.3808	-3.22E-04	5.72E-05
97.51	-17.2926	-2.87E+00	-2.93E-04	4.86E-05
98.57	-15.1409	-2.37E+00	-2.56E-04	4.02E-05
99.83	-12.7635	-1.92E+00	-2.16E-04	3.25E-05
100.81	-10.7889	-1.59E+00	-1.83E-04	2.69E-05
101.73	-8.8743	-1.27E+00	-1.50E-04	2.14E-05
102.66	-7.0034	-1.01E+00	-1.19E-04	1.71E-05
103.61	-5.2684	-7.08E-01	-8.92E-05	1.20E-05
104.67	-3.5185	-4.05E-01	-5.95E-05	6.85E-06
105.61	-2.3069	-1.96E-01	-3.90E-05	3.31E-06
106.50	-1.3789	-7.66E-02	-2.33E-05	1.30E-06
107.52	-0.7045	-3.33E-02	-1.19E-05	5.64E-07
108.44	-2.85E-01	-9.97E-03	-4.82E-06	1.69E-07
109.46	2.91E-02	4.95E-02	4.93E-07	-8.38E-07
110.35	1.04E-01	5.48E-02	1.76E-06	-9.28E-07
111.32	1.94E-01	5.34E-02	3.29E-06	-9.04E-07
112.32	2.09E-01	5.12E-02	3.54E-06	-8.66E-07
113.29	2.84E-01	4.00E-02	4.81E-06	-6.76E-07
115.25	1.94E-01	8.59E-02	3.28E-06	-1.45E-06
117.22	1.95E-01	2.59E-02	3.29E-06	-4.38E-07
119.12	1.94E-01	5.39E-02	3.28E-06	-9.12E-07
121.08	1.64E-01	3.69E-02	2.78E-06	-6.24E-07
123.03	1.64E-01	6.44E-02	2.77E-06	-1.09E-06

Note. DC:00e,F:133,AC:10,Mass:7.73E-02,Units:CGS

APPENDIX B

AC susceptibility measurement results for Eu Doping

Table 22

(TlPbSrCaCuO) with Eu Doping Eu = 0.00

T	V'(ÊV)	V"(ÊV)	X'	X"
83.98	-120.159	-7.4232	-7.96E-04	4.92E-05
84.63	-118.515	-7.0658	-7.85E-04	4.68E-05
85.97	-115.073	-6.5513	-7.62E-04	4.34E-05
87.17	-112.661	-6.3717	-7.46E-04	4.22E-05
88.78	-109.559	-6.175	-7.26E-04	4.09E-05
89.97	-107.596	-6.0647	-7.13E-04	4.02E-05
90.60	-106.367	-5.9918	-7.05E-04	3.97E-05
91.39	-105.003	-5.9104	-6.96E-04	3.92E-05
92.32	-103.234	-5.8221	-6.84E-04	3.86E-05
93.20	-101.511	-5.7055	-6.72E-04	3.78E-05
94.25	-99.3826	-5.5883	-6.58E-04	3.70E-05
95.25	-97.359	-5.4885	-6.45E-04	3.64E-05
96.08	-95.4705	-5.3895	-6.32E-04	3.57E-05
97.03	-93.3571	-5.2771	-6.18E-04	3.50E-05
98.06	-90.7641	-5.14E+00	-6.01E-04	3.41E-05
99.53	-87.0169	-4.95E+00	-5.76E-04	3.28E-05
100.46	-84.1989	-4.81E+00	-5.58E-04	3.19E-05
101.37	-81.3509	-4.67E+00	-5.39E-04	3.09E-05
102.28	-78.2028	-4.53E+00	-5.18E-04	3.00E-05
103.22	-74.4846	-4.40E+00	-4.93E-04	2.91E-05
104.27	-69.5052	-4.32E+00	-4.60E-04	2.86E-05
105.21	-63.4866	-4.48E+00	-4.21E-04	2.96E-05
106.06	-55.7685	-4.87E+00	-3.69E-04	3.23E-05
107.10	-45.8	-5.27E+00	-3.03E-04	3.49E-05
107.99	-37.7135	-5.28E+00	-2.50E-04	3.50E-05
108.99	-2.96E+01	-4.95E+00	-1.96E-04	3.28E-05
109.88	-2.32E+01	-4.45E+00	-1.54E-04	2.95E-05
110.86	-1.71E+01	-3.72E+00	-1.13E-04	2.47E-05
111.82	-1.15E+01	-2.74E+00	-7.61E-05	1.81E-05
112.80	-6.84E+00	-1.60E+00	-4.53E-05	1.06E-05
114.77	-1.20E+00	-1.67E-01	-7.93E-06	1.11E-06
116.67	2.25E-01	-2.07E-03	1.49E-06	1.37E-08
118.53	2.55E-01	-4.92E-05	1.69E-06	3.26E-10
120.49	2.10E-01	-1.33E-03	1.39E-06	8.84E-09
122.38	2.40E-01	1.69E-03	1.59E-06	-1.12E-08

Note. DC:00e,F:133,AC:10,Mass:1.98E-01,Units:CGS

Table 23

(TlPbBaSrCaCuO) with Eu Doping Eu = 0.02

T	V'(ÊV)	V"(ÊV)	X'	X"
82.42	-10.1522	-4.544	-2.98E-04	1.43E-04
83.01	-9.5483	-3.9054	-2.37E-04	9.71E-05
84.43	-8.2366	-2.5611	-2.05E-04	6.37E-05
85.80	-7.1125	-1.6379	-1.77E-04	4.07E-05
87.23	-6.1645	-0.9392	-1.53E-04	2.34E-05
88.68	-5.3465	-0.5319	-1.33E-04	1.32E-05
89.95	-4.7497	-0.3424	-1.18E-04	8.52E-06
90.56	-4.4806	-0.2892	-1.11E-04	7.19E-06
91.40	-4.1514	-0.245	-1.03E-04	6.09E-06
92.30	-3.8819	-0.2138	-9.66E-05	5.32E-06
93.26	-3.6272	-0.1908	-9.02E-05	4.75E-06
94.27	-3.4324	-0.1804	-8.54E-05	4.49E-06
95.23	-3.2226	-0.1668	-8.02E-05	4.15E-06
96.06	-3.0877	-0.1609	-7.68E-05	4.00E-06
97.03	-2.9678	-1.49E-01	-7.38E-05	3.71E-06
98.04	-2.803	-1.39E-01	-6.97E-05	3.47E-06
99.53	-2.5481	-1.31E-01	-6.34E-05	3.26E-06
100.49	-2.3682	-1.22E-01	-5.89E-05	3.03E-06
101.37	-2.2483	-1.17E-01	-5.59E-05	2.92E-06
102.29	-2.0834	-1.09E-01	-5.18E-05	2.71E-06
103.22	-1.9186	-9.95E-02	-4.77E-05	2.47E-06
104.27	-1.7387	-9.04E-02	-4.32E-05	2.25E-06
105.19	-1.5137	-8.54E-02	-3.77E-05	2.12E-06
106.08	-1.3489	-7.61E-02	-3.36E-05	1.89E-06
107.10	-1.1841	-6.22E-02	-2.95E-05	1.55E-06
107.97	-9.74E-01	-5.85E-02	-2.42E-05	1.46E-06
108.97	-7.94E-01	-5.89E-02	-1.98E-05	1.46E-06
109.88	-5.99E-01	-4.55E-02	-1.49E-05	1.13E-06
110.85	-4.04E-01	-3.71E-02	-1.01E-05	9.22E-07
111.81	-2.10E-01	-2.62E-02	-5.21E-06	6.51E-07
112.80	-5.98E-02	-1.15E-02	-1.49E-06	2.87E-07
114.72	1.95E-01	3.40E-03	4.85E-06	-8.46E-08
116.66	3.00E-01	8.74E-03	7.46E-06	-2.17E-07
118.51	3.00E-01	7.74E-03	7.46E-06	-1.92E-07
120.45	3.30E-01	7.76E-03	8.21E-06	-1.93E-07
122.32	3.30E-01	1.83E-02	8.20E-06	-4.54E-07

Note. DC:0Oe,F:133,AC:10,Mass:5.26E-02,Units:CGS

Table 24

(TlPbBaSrCaCuO) with Eu Doping Eu = 0.04

T	V'(ÊV)	V"(ÊV)	X'	X"
80.61	-155.419	-146.105	-3.69E-03	3.04E-03
81.23	-146.007	-138.979	-2.84E-03	2.71E-03
82.50	-126.582	-122.758	-2.46E-03	2.39E-03
83.87	-108.539	-105.015	-2.11E-03	2.04E-03
85.28	-91.769	-86.8098	-1.79E-03	1.69E-03
86.71	-77.0779	-68.9757	-1.50E-03	1.34E-03
88.04	-66.1364	-54.6475	-1.29E-03	1.06E-03
89.45	-56.7574	-41.8984	-1.11E-03	8.16E-04
90.98	-48.0506	-29.3191	-9.35E-04	5.71E-04
91.37	-45.6751	-26.1796	-8.89E-04	5.10E-04
92.36	-41.3244	-20.1613	-8.05E-04	3.93E-04
93.16	-38.083	-1.59E+01	-7.41E-04	3.10E-04
94.17	-34.6176	-11.6134	-6.74E-04	2.26E-04
95.18	-31.4398	-8.0189	-6.12E-04	1.56E-04
96.01	-29.2161	-5.64E+00	-5.69E-04	1.10E-04
96.94	-26.9866	-3.58E+00	-5.25E-04	6.97E-05
97.99	-24.9421	-2.10E+00	-4.86E-04	4.09E-05
99.49	-22.6149	-1.35E+00	-4.40E-04	2.63E-05
100.44	-19.7548	-1.04E+00	-3.85E-04	2.03E-05
101.33	-18.3162	-9.48E-01	-3.57E-04	1.85E-05
102.22	-16.8322	-8.79E-01	-3.28E-04	1.71E-05
103.17	-15.0785	-7.92E-01	-2.94E-04	1.54E-05
104.24	-13.5191	-7.42E-01	-2.63E-04	1.44E-05
105.14	-11.8551	-6.74E-01	-2.31E-04	1.31E-05
106.04	-9.97E+00	-6.32E-01	-1.94E-04	1.23E-05
107.07	-8.21E+00	-5.84E-01	-1.60E-04	1.14E-05
107.94	-6.37E+00	-5.79E-01	-1.24E-04	1.13E-05
108.96	-4.72E+00	-4.67E-01	-9.18E-05	9.10E-06
109.84	-3.25E+00	-3.14E-01	-6.33E-05	6.12E-06
110.81	-2.04E+00	-2.25E-01	-3.96E-05	4.38E-06
111.81	-1.18E+00	-1.26E-01	-2.30E-05	2.45E-06
112.77	-1.80E-01	-2.64E-03	-3.50E-06	5.14E-08
114.08	2.24E-01	3.94E-02	4.37E-06	-7.67E-07
116.70	2.09E-01	5.52E-02	6.07E-06	-1.07E-06
118.56	1.94E-01	5.89E-02	9.78E-06	-1.15E-06
120.52	2.99E-01	6.52E-02	2.58E-05	-1.27E-06

Note. DC:00e,F:133,AC:10,Mass:6.72E-02,Units:CGS

Table 25

(TlPbBaSrCaCuO) with Eu Doping Eu = 0.06

T	V'(ÊV)	V"(ÊV)	X'	X"
82.79	-13.6101	-7.5587	-2.98E-04	1.65E-04
83.26	-13.1342	-6.4648	-2.87E-04	1.41E-04
84.53	-11.8348	-3.5506	-2.59E-04	7.77E-05
85.81	-10.5443	-1.8528	-2.31E-04	4.05E-05
87.23	-9.0611	-0.8758	-1.98E-04	1.92E-05
88.72	-7.718	-0.4678	-1.69E-04	1.02E-05
89.99	-6.5804	-0.3234	-1.44E-04	7.08E-06
90.58	-5.8612	-0.2703	-1.28E-04	5.91E-06
91.40	-5.2016	-0.2408	-1.14E-04	5.27E-06
92.44	-4.3773	-0.1939	-9.58E-05	4.24E-06
93.26	-3.7485	-0.1204	-8.20E-05	2.63E-06
94.21	-3.1038	-9.82E-02	-6.79E-05	2.15E-06
95.21	-2.5186	-0.1025	-5.51E-05	2.24E-06
96.01	-2.2934	-0.1095	-5.02E-05	2.40E-06
96.99	-1.9794	-5.26E-02	-4.33E-05	1.15E-06
98.04	-1.7239	-7.66E-02	-3.77E-05	1.68E-06
99.61	-1.4997	-3.07E-02	-3.28E-05	6.71E-07
100.54	-1.3193	-5.05E-02	-2.89E-05	1.11E-06
101.44	-1.26	-9.99E-03	-2.76E-05	2.19E-07
102.36	-1.0952	1.89E-03	-2.40E-05	-4.13E-08
103.31	-0.9301	-2.33E-04	-2.03E-05	5.10E-09
104.36	-0.8093	-4.71E-02	-1.77E-05	1.03E-06
105.26	-0.7199	-1.06E-02	-1.58E-05	2.31E-07
106.13	-0.5703	1.45E-02	-1.25E-05	-3.18E-07
107.18	-0.4349	-1.11E-02	-9.51E-06	2.43E-07
108.05	-2.85E-01	7.52E-03	-6.24E-06	-1.65E-07
109.06	-2.25E-01	-4.27E-04	-4.92E-06	9.35E-09
109.95	-1.05E-01	3.17E-03	-2.30E-06	-6.93E-08
110.91	-3.05E-02	2.60E-02	-6.66E-07	-5.68E-07
111.88	1.42E-02	4.53E-02	3.11E-07	-9.90E-07
112.84	1.64E-01	4.84E-02	3.59E-06	-1.06E-06
114.78	1.50E-01	2.26E-02	3.27E-06	-4.95E-07
116.67	2.69E-01	5.67E-02	5.89E-06	-1.24E-06
118.57	2.39E-01	4.87E-02	5.23E-06	-1.07E-06
120.53	2.99E-01	6.17E-02	6.54E-06	-1.35E-06
122.32	2.70E-01	2.22E-02	5.90E-06	-4.86E-07

Note. DC:0Oe,F:133,AC:10,Mass:5.98E-02,Units:CGS

Table 26

(TlPbBaSrCaCuO) with Eu Doping Eu = 0.08

T	V'(ÊV)	V"(ÊV)	X'	X"
81.29	-40.3308	-51.3017	-4.54E-04	5.54E-04
82.07	-37.5859	-43.5381	-4.33E-04	4.43E-04
83.44	-33.3301	-29.5107	-4.06E-04	3.52E-04
85.00	-29.3138	-17.2272	-3.84E-04	2.25E-04
86.29	-26.4464	-9.6057	-3.46E-04	1.26E-04
87.54	-24.2561	-5.3049	-3.17E-04	6.94E-05
89.11	-21.2899	-2.4839	-2.79E-04	3.25E-05
90.54	-18.5347	-1.3237	-2.42E-04	1.73E-05
91.39	-16.8603	-0.9884	-2.21E-04	1.29E-05
92.36	-14.9585	-0.7892	-1.96E-04	1.03E-05
93.22	-13.1903	-0.6713	-1.73E-04	8.78E-06
94.15	-11.5116	-5.79E-01	-1.51E-04	7.57E-06
95.14	-9.698	-0.4833	-1.27E-04	6.32E-06
96.06	-8.1991	-0.4072	-1.07E-04	5.33E-06
96.96	-7.015	-3.48E-01	-9.18E-05	4.55E-06
98.00	-6.0558	-2.94E-01	-7.92E-05	3.85E-06
99.60	-4.9765	-2.43E-01	-6.51E-05	3.18E-06
100.56	-4.5268	-2.23E-01	-5.92E-05	2.91E-06
101.47	-4.0921	-2.04E-01	-5.35E-05	2.67E-06
102.38	-3.7023	-1.89E-01	-4.84E-05	2.47E-06
103.31	-3.3425	-1.70E-01	-4.37E-05	2.22E-06
104.38	-2.8779	-1.48E-01	-3.77E-05	1.93E-06
105.29	-2.4881	-1.32E-01	-3.26E-05	1.73E-06
106.18	-2.1132	-1.22E-01	-2.76E-05	1.59E-06
107.20	-1.6783	-1.10E-01	-2.20E-05	1.44E-06
108.07	-1.27E+00	-9.32E-02	-1.67E-05	1.22E-06
109.09	-8.69E-01	-7.92E-02	-1.14E-05	1.04E-06
109.96	-5.69E-01	-5.34E-02	-7.45E-06	6.99E-07
110.95	-2.55E-01	-3.39E-02	-3.33E-06	4.44E-07
111.91	-8.97E-02	-1.91E-02	-1.17E-06	2.49E-07
112.89	1.52E-02	-1.32E-02	1.99E-07	1.73E-07
114.88	1.80E-01	7.64E-03	2.35E-06	-1.00E-07
116.76	3.00E-01	6.74E-03	3.92E-06	-8.81E-08
118.63	3.30E-01	8.76E-03	4.32E-06	-1.15E-07
120.55	3.60E-01	1.13E-02	4.71E-06	-1.48E-07
122.47	3.15E-01	1.10E-02	4.12E-06	-1.44E-07

Note. DC:0Oe,F:133,AC:10 (Arb. Units),Units:CGS

Table 27

(TlPbBaSrCaCuO) with Eu Doping Eu = 0.10

T	V'(ÊV)	V"(ÊV)	X'	X"
82.17	-9.3321	-0.8225	-1.22E-04	1.08E-05
82.73	-8.9141	-0.7032	-1.17E-04	9.20E-06
83.89	-7.9122	-0.5157	-1.04E-04	6.75E-06
85.40	-6.6542	-0.3932	-8.71E-05	5.14E-06
86.77	-5.6801	-0.3332	-7.43E-05	4.36E-06
88.00	-4.8858	-0.2833	-6.39E-05	3.71E-06
89.55	-3.9266	-0.2296	-5.14E-05	3.00E-06
91.02	-3.0723	-0.1836	-4.02E-05	2.40E-06
91.44	-2.8024	-0.1714	-3.67E-05	2.24E-06
92.30	-2.3828	-0.1466	-3.12E-05	1.92E-06
93.18	-1.9482	-0.122	-2.55E-05	1.60E-06
94.31	-1.4385	-9.71E-02	-1.88E-05	1.27E-06
95.23	-1.0489	-7.18E-02	-1.37E-05	9.40E-07
96.02	-0.779	-6.56E-02	-1.02E-05	8.58E-07
96.99	-0.4793	-4.59E-02	-6.27E-06	6.00E-07
98.06	-0.2693	-4.32E-02	-3.52E-06	5.65E-07
99.49	-7.45E-02	-2.73E-02	-9.75E-07	3.57E-07
100.42		-2.74E-02	7.91E-07	3.59E-07
101.33	0.1203	-1.59E-02	1.57E-06	2.08E-07
102.26	0.2103	-1.43E-02	2.75E-06	1.88E-07
103.19	0.2102	-1.03E-02	2.75E-06	1.35E-07
104.24	0.3003	-1.33E-02	3.93E-06	1.74E-07
105.16	0.3002	-1.08E-02	3.93E-06	1.41E-07
106.04	0.3452	-8.48E-03	4.52E-06	1.11E-07
107.07	0.4051	-4.31E-04	5.30E-06	5.63E-09
107.94	4.50E-01	3.35E-03	5.89E-06	-4.39E-08
108.96	4.80E-01	3.78E-04	6.28E-06	-4.95E-09
109.83	5.55E-01	3.19E-03	7.26E-06	-4.17E-08
110.83	5.25E-01	2.16E-03	6.87E-06	-2.83E-08
111.81	5.55E-01	6.19E-03	7.26E-06	-8.09E-08
112.77	6.30E-01	1.45E-02	8.24E-06	-1.90E-07
114.77	6.90E-01	9.54E-03	9.03E-06	-1.25E-07
116.66	7.50E-01	1.41E-02	9.81E-06	-1.84E-07
118.51	7.80E-01	1.31E-02	1.02E-05	-1.72E-07
120.45	7.65E-01	1.64E-02	1.00E-05	-2.14E-07
122.38	8.10E-01	1.26E-02	1.06E-05	-1.65E-07

Note. DC:00e,F:133,AC:10,(Arb. Units),Units: CGS