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Quasiperiodic Transition to Chaos in Ge; and Magnetic Susceptibility of High-$T_c$ Superconductors

Y. Kim

Ph.D. Thesis

April 1990

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Quasiperiodic Transition to Chaos in Ge; and
Magnetic Susceptibility of High-$T_c$ Superconductors

Youn-tae Kim

Ph. D. Thesis

April 1990

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Quasiperiodic Transition to Chaos in Ge; and
Magnetic Susceptibility of High-$T_c$ Superconductors

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Youngtae Kim
To my wife

Soon-Eun

and

my sons

Hyung-Suk, Hyung-Gil
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PREFACE

This thesis consists of the study of two topics in condensed matter physics: nonlinear dynamics in a Ge crystal; and magnetic properties of the high temperature superconductor $\text{YBa}_2\text{Cu}_3\text{O}_7$. These are given as two parts, each of which can be read independently.

The first part is concerned with the measurement of the universal spectrum of scaling indices of the critical attractor of the quasiperiodic route to chaos with golden mean winding number for the helical instability of the electron-hole plasma in a germanium crystal in a magnetic field. The second part consists of measurements of the complex ac magnetic susceptibility of the ceramic superconductor $\text{YBa}_2\text{Cu}_3\text{O}_7$, and together with theoretical models of the material, assumed to be composed of intergranular junctions and superconducting grains.

This thesis is a part of the fulfillment of the requirements for the degree of Doctor of Philosophy in the subject of Physics at the University of California at Berkeley.

March 26, 1990

Berkeley, CA

Youngtae Kim
Part I

Multifractal Structure of Attractors at the Quasiperiodic Transition to Chaos for Electron-Hole Plasma Instability in Ge
ABSTRACT

A recent theoretical development, the so-called $f(\alpha)$ spectrum, to characterize attractors of nonlinear dynamical systems was experimentally measured for quasi-periodic transition to chaos for helical instability of electron-hole plasma in a germanium crystal. The $f(\alpha)$ spectrum gives complete information of the multifractal, or interweaving fractal nature of scaling index $\alpha$, which is related to the probability distributions of states in a phase space on the attractors and thus the spectrum is unique for different routes to chaos. The $f(\alpha)$ spectrum measured from the time series of the quasiperiodic current oscillation of the plasma with irrational golden mean winding number at the onset of chaos is in good agreement with the prediction of a circlemap, which is a simple one-dimensional model of two interacting oscillators. The experimental power spectrum of the current oscillation also confirms that the transition to chaos is well described by the circlemap. Noise effects on the power spectrum and the scaling function $f(\alpha)$ are discussed by using the circlemap to compute the small deviation between the experiment and theory. It was concluded that the $f(\alpha)$ spectrum is a good quantity to characterize the attractors of dynamical systems in comparison with other dynamical quantities, but because of the sensitivity of the experimental $f(\alpha)$ spectrum to random noise the usefulness of the measurement is limited to very clean systems, i.e., those with very low intrinsic random noise.
CHAPTER 1. INTRODUCTION

Recent theoretical progress in nonlinear dynamics of dissipative systems has made it possible to understand a variety of nonlinear phenomena in nature\(^1\). Particularly deterministic chaos (whose name comes from the fact that simple, perfectly definite equations of motion can produce unpredictable, chaotic motions) has been studied extensively\(^2\). Discovery of various routes to chaos, period-doubling, quasiperiodicity, frequency locking, and intermittency, and theoretical predictions of the universality of each route are typical examples of the progress. Tests of the theoretical predictions have been done for many systems, for example, hydrodynamical\(^3\), chemical\(^4\), optical\(^5\), and solid-state systems\(^6\), and their experimental results have been found to be in reasonable agreement with the theoretical predictions. Recently it was found that spontaneous current oscillations due to helical instability of electron-hole plasma in a germanium crystal placed in parallel electric and magnetic fields showed chaotic dynamics and provided a good system to test the theory of nonlinear dynamics\(^7\).

In Part I of this thesis we observed the helical instability of electron-hole plasma in a Ge crystal to test the universal predictions of quasiperiodic transition to chaos. The quasiperiodic transition to chaos has been well studied theoretically by using a simple one-dimensional model, a circlemap\(^8\). This model makes predictions for the nature of frequency locking transition to chaos as well as quasiperiodic transition to chaos. The universal power spectrum and the \(f(\alpha)\) spectrum
are examples of universal properties of transition to chaos from quasiperiodic states. The $f(\alpha)$ spectrum is a recently developed theoretical concept which represents the probability distribution on attractors by a spectrum of scaling indices $\alpha$. The scaling index $\alpha$ is a kind of dimension for singularity of a probability $P(L)$ assigned to a piece of length $L$ and is defined as $P(L)\sim L^\alpha$. The $f(\alpha)$ is a fractal dimension of an attractor with a scaling index $\alpha$. The $f(\alpha)$ spectrum has been shown to be universal for the circlemap at the onset of quasiperiodic transition to chaos.

In this chapter the helical instability of electron-hole plasma in a semiconductor crystal placed in parallel electric and magnetic fields is reviewed in the section 1.1. This helical instability produces various routes to chaos as the electric field is increased. The Section 1.2 is a review of basic properties of chaos in dissipative systems. Three different routes to chaos are described in Section 1.3.

Chapter 2 discusses the sample preparation, experimental setup of apparatus, and the data acquisition for power spectrum and $f(\alpha)$ spectrum. Chapter 3 gives a review of the theoretical work on the circlemap and the universal scaling function $f(\alpha)$ at the onset of quasiperiodic transition to chaos. The experimental measurements of the power spectrum and $f(\alpha)$ spectrum are discussed in Chapter 4, where the method of analyzing the experimental data to obtain the spectra is discussed in detail. Chapter 5 describes the numerically computed random noise effect on the power spectrum and $f(\alpha)$ spectrum of the circlemap, and relates these to the experimental observation of noise-related features.
1.1. Helical Instability of Electron-Hole Plasma

A large number of instabilities, including the helical density instability which is the topic in this Section, in gaseous and in semiconductor plasmas have been discovered. Instabilities due to the Gunn effect and avalanche effect are the typical examples for semiconductor plasma. The helical plasma density instability was first discovered in 1958 by Lehnert in gaseous plasma. Ivanov and Ryvkin were the first who observed the same instability in semiconductors: small current oscillations of current when a large dc current was passed through a semiconductor in presence of a dc magnetic field. In a magnetic field, electrons and ions spiral along the magnetic field within the discharge column so the discharge column twists in a helical fashion which gives the name helical instability. The helical instability can be understood in the following way: when there is a superposed screw-type perturbation of positive (holes) and negative (electrons) carriers, a dc electric field \( E_0 \) separates the perturbed positive and negative helices axially as shown in Fig. 1.1(a). The resulting charge separation creates azimuthal electric fields, \( E_{r} \) and \( E_{\phi} \) shown in Fig. 1.1(b). These fields, together with the axial magnetic field \( B_0 \), cause the radial flow of both carriers which can produce a build-up of carrier densities near the surface shown in Fig. 1.1(c) and Fig. 1.1(d). This results in a growing helical perturbation when the influx of the carriers is sufficiently large to overcome the diffusion and recombination of the charge carriers. In Appendix A the classical mathematical description of this system by Hurwitz and McWhorter is given and the possible nonlinear mode interaction of
helical instability which gives rise to chaotic dynamics is also discussed.

1.2. Basic Properties of Chaos in Dissipative System

Most experimental systems, to some extent, dissipate energy provided from external sources to maintain motions of the systems. For generic dissipative systems the initial trajectory in phase space has very large dimension, which corresponds to the number of particles of the systems. As the system evolves, the phase trajectory typically elongates along some directions and contracts to other directions so the motion can be described with equations of motion with much lower degrees of freedom. The phase trajectory of the motion of the dissipative systems is usually a fractal object which means the dimension of the phase trajectory is not integer. Particularly when the system is chaotic, the nearby states on the trajectory diverge from one another exponentially in time so the trajectory of a steady state motion in phase space, which is called a limit set or an attractor, becomes a strange attractor which was coined by Ruell and Takens. The sensitive dependence of initial conditions of the strange attractor explains the difficulty of predicting the future accurately because the motion looks random or chaotic. Typically the strange attractor consists of an infinite number of folded surfaces contained within a bounded phase space and the fractal dimension of the strange attractor is an approximate measure of the number of degree of freedom necessary to describe the behavior of the system. Three universal routes for the low dimensional dissipative system to chaos have been well studied so far and those will be
1.3. Transitions to Chaos

1.3.1. Period-Doubling Transition

The period-doubling transition to chaos is most well-known in theory and experiment. The characteristics of this route to chaos can be found most simply by studying a logistic map, which is a special form of the quadratic map\(^{16}\), for a simple dynamical variable \(x_n\) and a control parameter \(\lambda\):

\[
x_{n+1} = \lambda \cdot x_n (1 - x_n).
\] (1.1)

The set of consecutive iterates \(\{x_n\}\) versus \(\lambda\) is the bifurcation diagram of the logistic map, which shows the development of the solutions of the map as \(\lambda\) increases, Fig. 1.2. The number of the solutions doubles as \(\lambda\) passes certain values. It should be mentioned that the map does not simply double the solutions but it oscillates between the solutions periodically. This is clearly shown in the power spectra of the solutions of the logistic map in Fig. 1.3. In the power spectra new subharmonic peaks, which are located at subharmonics of the previous peak, appear at each bifurcation point. The logistic map becomes chaotic when \(\lambda\) is greater than \(\lambda_\infty = 3.5699\ldots\) because the solution has an infinite period.

The period-doubling transition to chaos of the logistic map has been studied extensively by many authors using the renormalization group method\(^{17}\) and it was found that this period-doubling route to chaos occurs with some restrictions\(^{18}\) in one-dimensional map of first-order difference equations \(x_{n+1} = f(x_n)\) where \(f(x_n)\)
has only a single maximum in the unit interval $0 \leq x_n \leq 1$.

Many universal properties of the period-doubling transition to chaos have been discovered. For example, the value of $\lambda_n$ which is the control parameter at the onset of period $2^n$ scales as $\lambda_\infty - \lambda_n \propto \delta^{-n}$ for large $n$ where $\delta = 4.6692...$ is a universal constant. The power spectrum showed that the amplitudes of the odd subharmonics which appear after each bifurcation step are *in the mean* just the averaged amplitudes of the old components reduced by a universal factor $-13.5$ dB.

1.3.2. Intermittent Transition

In the fluid dynamics theory of turbulence the intermittency means random bursts of a turbulent motion on the background of laminar flow. In the nonlinear dynamics theory the intermittency means a similar thing, random alternations of chaotic and regular behavior in time evolution without involving any spatial degrees of freedom. The mechanism of intermittency can be understood with the logistic map and it occurs in the neighborhood of a tangent bifurcation\textsuperscript{19} where as period-doubling is related to the pitchfork bifurcations. For the logistic map the average time $t$ of regular behavior is given by $t \propto (\lambda_c - \lambda)^{-1/2}$ where $\lambda_c$ is the value of $\lambda$ at a tangent bifurcation\textsuperscript{19}.

1.3.3. Quasiperiodic and Frequency-Locking Transition

Landau first suggested that fluids going into the turbulent state develop
infinitely increasing number of incommensurate frequencies through Hopf bifurcations\textsuperscript{21}, which introduces a new fundamental frequency into the system\textsuperscript{22}. Later Ruell and Takens\textsuperscript{15} suggested a route to chaos which is much shorter than that proposed by Landau. They have shown that even two Hopf bifurcations could make the regular motion (or two interacting oscillators) highly unstable in favor of chaotic motion at a strange attractor.

The steady-state flow of two interacting oscillators in the phase space lies on a two-dimensional torus and depending on the frequency ratio of the two oscillators it shows frequency locking when the ratio is rational, or quasiperiodicity when the ratio is irrational. For a quasiperiodic flow the cross-section of its phase trajectory, so-called a Poincare section, is a closed one-dimensional curve. For a frequency locking its Poincare section consists of points because the flow repeats itself. The Poincare map of two oscillators whose motion is quasiperiodic or frequency locking can be described by a simple one-dimensional discrete time map of the circle, so-called a circlemap\textsuperscript{23}. The circlemap is given by:

\begin{equation}
\theta_{n+1} = f (\theta_n) = \theta_n + \Omega - \frac{K}{2\pi} \sin (2\pi \theta_n)
\end{equation}

where $\theta_n$ is the one dimensional dynamical variable, $K$ is a measure of nonlinearity, and $\Omega$ is the ratio of two oscillation frequencies in the absence of nonlinearity. Eq. 1.2 is sometimes called a sine circlemap, or sinemap because the nonlinearity comes from a sine function. The detailed properties of this circlemap will be discussed in Chapter 3.
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FIGURE CAPTIONS

Fig. 1.1. (a): Helical plasma density wave in a cylindrical semiconductor placed in a dc electric field \( E_0 \) and a dc magnetic field \( B_0 \) along the cylindrical axis is shown. The helix represents the perturbation of electron and hole density. (b): Various fields produced by the applied electric and magnetic field are shown. (c) and (d): Charge carrier densities built up by the induced fields shown in (b) are shown as a dotted line. The solid line shows an equilibrium charge density. This figure is from Ref. 13.

Fig. 1.2. Bifurcation diagram of a logistic map in Eq. 1.1 is shown. The letters on the \( \lambda \) axis correspond to the power spectra in Fig. 1.3.

Fig. 1.3. Power spectra of a period-doubling route to chaos: (a) period 1, \( \lambda = 2.9 \). (b) period 2, \( \lambda = 3.2 \). (c) period 4, \( \lambda = 3.455 \). (d) period 8, \( \lambda = 3.55 \). (e) chaos, \( \lambda = 3.6 \). (f) period 3, \( \lambda = 3.83 \).
Figure 1.1
Figure 1.2

Figure 1.3
CHAPTER 2. EXPERIMENTAL DETAILS

2.1. Sample

The sample used in the experiment is a n-type Ge crystal with net donor density $3.7 \times 10^{12}/\text{cm}^3$, difference between donor and acceptor density\(^1\). It was cut from a large, single crystal of Ge grown by Haller and Hansen at the Lawrence Berkeley Laboratory\(^2\). It is a rectangular-shaped rod with a dimension $1 \times 1 \times 10$ mm as shown in Fig. 2.1. The electron-hole plasma in a Ge crystal can be generated by double injection or optical pumping. In the experiment double injection method was used. For the double injection, a Li-diffused n\(^+\) contact for electron injection and a B-implanted p\(^+\) contact for hole injection was formed on opposite ends of the sample. When a dc bias voltage is applied to those ends shown in the Fig. 2.1, it produces the electron-hole plasma. The sample has additional 8 pairs of side contacts as shown in the Fig. 2.1 to apply an ac modulation to a current oscillation in the experiment. The side contact is 0.5 mm wide and spaced 1 mm and is formed by heavy P-implantation and Au-sputtering. Wires for the dc voltage bias were indium soldered and the wires were attached to the side contacts with silver epoxy. The sample was placed in a vertical metal dewar in which a horizontal dc magnetic field can be applied from an external split coil electro-magnet\(^1\). The magnet can be rotated around the dewar in the horizontal plane. The magnetic field intensity was measured with a rotating coil Gaussmeter and the magnet could provide a magnetic field up to 19 kG. Usually the dc magnetic field was almost paral-
lel to the sample axis within ± 4 degrees, and could be adjusted in orientation. During the experiment liquid nitrogen was used to cool the sample at 77 K, by immersion.

2.2. Experimental Setup

The experimental setup without an ac modulation of the current oscillation is shown in Fig. 2.2. The sample is connected to a low-noise dc power supply and a 100 Ω metal resistor in series. The low-noise dc power supply consists of a HP 6216A dc power supply and a high-precision voltage regulator made in my laboratory. The circuit diagram of the regulator is shown in Fig. 2.3. A feedback loop by using a power transistor regulates the dc output of a HP 6216A dc power supply from fluctuations of line voltages and loads; the output is constant to less than 20 μV. The dc output voltage can be varied from approximately 3 V to the maximum voltage, $V_{\text{max}} = 30$ V of the input power supply continuously. It was found that such a high degree of voltage regulation was very important to see the fine details of attractors without excess blurring of the trajectory. In the experiment there are three adjustable parameters: dc bias voltage $V_0$ across the sample, dc magnetic field $B_0$, and angle $\theta$ between $B_0$ and the sample axis. The magnetic field $B_0$ is essentially parallel to the sample axis as mentioned earlier. The experiment is done as follows: for a given value of $B_0$ and $\theta$, and $V_0$ sufficiently small, the ac component of the output voltage, $V_s$ in Fig. 2.2, is essentially zero. As $V_0$ is increased to a critical value, there is an abrupt onset of an ac voltage. This onset
of spontaneous plasma current oscillation forms a critical line in Fig. 2.4. The critical value of $V_0$ as a function of $B_0$ is shown as a lower line in Fig. 2.4. The second line in Fig. 2.4 is a line of onset of period-doubling transition and the top one is a critical line of period-doubling transition to strong chaos. Thus Fig. 2.4 is a measured phase diagram in $V_0 - B_0$ space for $\theta = -2^\circ$ and here the system went through mainly period doubling transition to chaos. The spontaneous current oscillation was monitored by observing the voltage across a $R_L = 100 \, \Omega$ metal resistor. The current oscillation $I(t)$ is a dynamical variable of the system. Tektronix differential amplifier AM 502 amplified the voltage and it was filtered by using a Ithaco dual filter model 4302 to remove harmonic distortions. The filtered signal was inputed to a HP 3585A analog spectrum analyzer and a Tektronix 468 digital oscilloscope to get power spectra and waveforms of the current oscillation respectively. They are connected to a DEC LSI-11 computer via a IEEE 488 bus in order to store the power spectra and the waveforms of the current oscillation onto a hard disk. The power spectra taken along the period-doubling route to chaos at $B_0 = 8.5 \, \text{kG}$ shown in Fig. 2.4 are given in Fig. 2.5 for increasing values of $V_0$. In Fig. 2.5(a) a sharp fundamental peak of the current oscillation with a frequency $f_0 \sim 120 \, \text{kHz}$ appears, along with harmonics. In Fig. 2.5(b) subharmonics at $1/2 f_0$ and $3/2 f_0$ appear which indicates a period 2 bifurcation appears. In Fig. 2.5(c) a period 4 bifurcation appears and further increase of $V_0$ (Fig. 2.5(d)) shows onset of weak chaos, which corresponds to broad peaks for $1/4$ subharmonics; a sharp period 8 bifurcation was not observed for this system owing to the presence of
extrinsic noise. As $V_0$ increases further the power spectrum shows a period 3 window (Fig. 2.5(e)) and strong chaos (Fig. 2.5(f)) which was indicated by the rise of background noise and disappearance of peaks. This sequence of dynamics of the system may be well understood from the bifurcation diagram and the power spectra of the logistic map shown in figures in Chapter 1, with the further assumption of additional noise not of deterministic origin.

The sample showed a variety of different routes to chaos when the angle between $B_0$ and the sample axis becomes larger, in contrast to simple current oscillations at small angles. For example, a large number of quasiperiodic routes were observed at $\theta = 5^\circ$ as shown in Fig. 2.6. The lower line represents the onset of the spontaneous current oscillation. The middle line is a line of onset of quasiperiodic or frequency locking transition from the spontaneous current oscillation, and the top line is a critical line of onset of strong chaos. The power spectra corresponding to the quasiperiodic route to chaos in Fig. 2.6 are shown in Fig. 2.7. As $V_0$ increased, there appeared another current oscillation with frequency $f_s$ in addition to the original current oscillation with frequency $f_0$ through a Hopf bifurcation. The ratio between two frequencies $f_s / f_0$ decreased slowly with increasing $V_0$. The motion of two interacting current oscillations is frequency locking when the ratio is rational, and quasiperiodic when the ratio is irrational. If $V_0$ becomes sufficiently large, the motion becomes chaotic as in Fig. 2.7(g) which shows typical properties of chaotic motions: increase of broad-band noise and broadening of peaks.
In order to do a controlled experiment on the interacting two oscillators an ac modulation voltage $V_1$ with an applied frequency $f_1$ was applied to a pair of side contacts just next to an injecting contact for holes (see Fig. 2.1). This situation was required in order for a minimum $V_1$ to accomplish a quasiperiodic transition to chaos and gave a best coupling to the longitudinal plasma current. The ac modulation voltage develops a local modulation of the spontaneous current oscillation of frequency $f_0$ so we can get a nonlinear interaction of two oscillators experimentally. The schematic diagram of the experimental setup with an ac modulation is given in Fig. 2.8. A low noise, very stable synthesizer/function generator HP3325A supplies an ac modulation with frequency $f_1$ and amplitude $V_1$ to the side contacts. The synthesizer also provides triggering pulse to a 12-bit analog-to-digital converter (ADC) with period $\tau_1 = 1/f_1$. Except for the application of the ac modulation the circuitry of the setup is the same as Fig. 2.2. The coupling between the spontaneous current oscillation and the ac modulation can be varied by changing the amplitude of ac voltage $V_1$ from the synthesizer. Because of this nonlinear coupling the changes of $f_1$ and $V_1$ continuously modify $f_0$, the intrinsic frequency of the spontaneous current oscillation. The frequency ratio, $f_1 / f_0$, which decides the types of motions of two-oscillator system could be monitored continuously at given $V_1$ and $V_0$ by using the spectrum analyzer. For data acquisition the signal is connected to a computer via a 12-bit analog-to-digital-converter (ADC) and this will be explained in detail in the next section.
2.3. Data Acquisition

The trajectories of motions in phase spaces can be reconstructed from experimental time series data by following Takens theorem\(^4\) which states that a \(n\)-dimensional phase trajectory can be constructed by using a time series of a single dynamical variable, say the plasma current \(I_i\). The so-called reconstructed \(n\)-dimensional phase trajectory \((I_i, I_{i+1}, I_{i+2}, \ldots, I_{i+n-1})\) for \(i = 1, 2, 3, \ldots\) is conjectured to have the same topology as the original \(n\)-dimensional phase trajectory where \(I_i = I(t+i\tau)\) and \(\tau\) is a sampling time of the experimental variable \(I(t)\). Takens theorem is true unless \(\tau\) is very small or very large where there is no correlation between data. When the sampling rate, \(1 / \tau\) is same as the frequency of the ac modulation, the reconstructed phase trajectory actually corresponds to a Poincare section of the motion.

As mentioned in the Section 2.2 a HP synthesizer provides triggering pulses to the ADC with period \(\tau_1 = 1 / f_1\) to get a time series data of the modulated current oscillation. Usually the triggering pulse is delayed in time with a HP 214B pulse generator to get a better reconstructed Poincare section which means one with less overlapped points and more circular on a Tektronix 611 display unit. The Poincare section could be displayed on a high resolution Tektronix 611 display unit by a program using time series data, or displayed in real time by an analog sample and hold circuit\(^5\). The most important thing in this experiment is to reduce ambient noise in the system to improve signal-to-noise (S/N) ratio of the signal. There are a large number of choices of parameter values, \((\theta, V_0, B_0)\). From the \(V_0\)
- $B_0$ phase diagrams for different $\theta$, the angle was determined (to $\pm 1^\circ$) which showed clean current oscillations in most of the phase space. The $B_0$ was fixed about 8kG which is about mid range of the maximum magnetic field of the magnet. In order to choose the optimum $V_0$ the frequency and power of the signal were measured by the spectrum analyzer as a function of $V_0$ as shown in Fig. 2.9, for this figure $\theta = 1^\circ$ and $B_0 = 8.092$ kG. In the range $V_0 = 10$ to 13 V the increasing rate of frequency and power of the oscillation was found to be small, and this maintained good frequency and power stability of the fundamental current oscillation. Fig. 2.10 shows the power spectrum of a clean spontaneous current oscillation taken at one of the optimum conditions found following the method discussed above; here $\theta = 1^\circ$, $B_0 = 7.9$ kG, and $V_0 = 13.5$ V. The frequency of the oscillation was around 200 kHz. As you can see from the figure, advantages of this system to do nonlinear dynamics experiment are:

(1) a large S/N ratio of $\approx 100$ dB shown in this power spectrum,

(2) frequency of the current oscillation has high stability which is approximately 5 parts in $10^5$,

(3) the frequency is low enough to allow digitization of time series by using a standard ADC and high enough to get a large number of data points over short time, so that drifts that cause extrinsic noise can be suppressed. For example, it took only 1 or 2 seconds to take 98,000 continuous samples of the time series of the current oscillation for each run.
REFERENCES. CHAPTER 2


FIGURE CAPTIONS

Fig. 2.1. Ge sample used in the experiment is illustrated. It is a rectangular-shaped rod with a dimension 1x1x10 mm. There are 8 pairs of side contacts. The electron-hole plasma is generated by applying a dc voltage along the sample in the presence of a parallel dc magnetic field.

Fig. 2.2. Schematic diagram of an experimental setup without an ac modulation of the current oscillation. The spontaneous current oscillation due to the helical plasma density instability is monitored by measuring the voltage across a 100 Ω metal film resistor, $R_L$.

Fig. 2.3. Circuit diagram of a high-precision dc voltage regulator. This circuit regulates the voltage output shown as $V_{\text{in}}$ from a commercial dc power supply. The regulated output can be from about 3 V to the maximum output of the input power supply, $V_{\text{max}} = 30$ V. The output voltage fluctuation is less than 20 μV.

Fig. 2.4. Measured phase diagram of the plasma current oscillation at $\theta = -2^\circ$ and $T = 77$ K in $V_0 - B_0$ space. The power spectra of the current oscillation along the route shown in the figure are given in Fig. 2.5. The lower line is a critical line of onset of spontaneous current oscillation. The middle line is a line of onset of period-doubling transition and the top one is a critical line of period-doubling transition to strong chaos.

Fig. 2.5. Power spectra of the spontaneous current oscillation along the period-doubling route to chaos shown in Fig. 2.4: (a) clean oscillation, (b) period 2, (c)
period 4, (d) weak chaos, (e) period 3, and (f) strong chaos.

Fig. 2.6. Measured phase diagram at $\theta = 5^\circ$ and $T = 77$ K. At this angle mostly quasiperiodic transitions to chaos were observed. The corresponding power spectra along the given path are in Fig. 2.7. The lower line is a critical line of onset of spontaneous current oscillation. The middle line is a line of onset of quasiperiodic or frequency locking transition and the top one is a critical line of onset of strong chaos.

Fig. 2.7. Power spectra of the spontaneous current oscillation of quasiperiodic transitions to chaos along the path shown in Fig. 2.6: (a) clean oscillation, (b) quasiperiodic, (c) 1/5 frequency locking, (d) quasiperiodic, (e) weak chaos, (f) period 2 bifurcation in 1/6 frequency locking, (g) chaos, and (h) clean oscillation.

Fig. 2.8. Schematic diagram of experimental setup with an ac modulation. This is the same as Fig. 2.2 except an ac modulation voltage across a pair of side contacts. To generate the Poincare section of the motions of the signal continuous time series of current data with a sampling rate $f_1$, which is the ac modulation frequency, is stored in a computer.

Fig. 2.9. The frequency and the power of a spontaneous current oscillation at $\theta = 1^\circ$ and $B_0 = 8.092$ kG as a function of $V_0$ to find optimum parameter values, $V_0$ and $B_0$.

Fig. 2.10. The power spectrum of a clean spontaneous current oscillation at the optimum parameter values: $\theta = 1^\circ$, $B_0 = 7.9$ kG, and $V_0 = 13.5$ V.
n-type Ge ($n_D - n_A = 3.7 \times 10^{12} \text{ cm}^{-3}$)

Figure 2.1

Figure 2.2
maximum current $I_m = \frac{650}{R_1}$ mA

Figure 2.3

Figure 2.4
Figure 2.5

(a) $V_0 = 15.0 \text{ V}$
(b) $15.5 \text{ V}$
(c) $18.6 \text{ V}$
(d) $18.8 \text{ V}$
(e) $20.1 \text{ V}$
(f) $21.2 \text{ V}$

$B_0 = 8.5 \text{ kG}, \ \theta = -2^\circ$
Figure 2.6
Figure 2.7

(a) $V_0 = 11.7 \text{ V}$

(b) $V_0 = 12.3 \text{ V}$

(c) $P(f) \text{ (dB)}$

(d) $P(f) \text{ (dB)}$

(e) $P(f) \text{ (dB)}$

(f) $P(f) \text{ (dB)}$

(g) $P(f) \text{ (dB)}$

(h) $P(f) \text{ (dB)}$

$B_0 = 8.5 \text{ kG}, \quad \theta = 5^\circ$
Figure 2.8
Figure 2.10
CHAPTER 3. THEORY

3.1. Circlemap

The circlemap which can describe the motions with quasiperiodicity or frequency locking is the mapping of a circle onto itself as given in Section 1.3.3:

\[ \theta_{n+1} = f(\theta_n) = \theta_n + \Omega - \frac{K}{2\pi} \sin(2\pi \theta_n) \]  

where \( \theta_n \) has modulo 1. The \( K \) is a measure of nonlinearity and \( \Omega \) is called the bare winding number. This is a limiting case of the more generalized two-dimensional annular mapping:

\[ \begin{align*}
\theta_{n+1} &= \theta_n + \Omega - \frac{K}{2\pi} \sin(2\pi \theta_n) + br_n \\
\rho_{n+1} &= br_n - \frac{K}{2\pi} \sin(2\pi \theta_n).
\end{align*} \]

For strongly dissipative systems the radial motion of the trajectory disappears so \( b \to 0 \) in Eq. 3.2 and this equation becomes equivalent to Eq. 3.1. The theoretical results to be used in this thesis have been derived from the one-dimensional circlemap, Eq. 3.1. Papers by Feigenbaum et al.\(^1\) and Rand et al.\(^2\) have studied properties of the circlemap within the renormalization group framework to see how a quasiperiodic motion with two incommensurate frequencies on a torus becomes chaotic if there is a nonlinear perturbation to the motion. When there is a nonlinear perturbation to the motion such as a term \( \sin(2\pi \theta) \) in Eq. 3.1, the actual rate of rotation of the state which is called a winding number \( \sigma \), on a torus is different from the bare winding number \( \Omega \) in the equation. If we increase the nonlinearity \( K, \Omega \) should be changed to keep the winding number \( \sigma \) fixed to a given number,
rational or irrational. In principle the winding number $\sigma$ is defined as the average shift of angle $\theta$ per iteration:

$$\sigma = \lim_{n \to \infty} \frac{f^n(\theta_0) - \theta_0}{n}.$$  \hspace{1cm} (3.3)

Using this definition we determine $\sigma$ of the circlemap numerically for given values of $K$ and $\Omega$. Fig. 3.1 shows the phase diagram of the circlemap for some rational values of $\sigma$ in $K - \Omega$ space. For rational $\sigma$ values where the motion is frequency locking, the range of $\Omega$ which gives the same $\sigma$ becomes larger with increasing $K$, and the boundary of this same $\sigma$ defined with the minimum and maximum value of $\Omega$ generates a V-shape which is called an Arnold tongue. The orders of occurrence of the frequency lockings are known to be given by the Farey sequences known in number theory. An $n$-Farey sequence is the increasing succession of rational numbers $p/q$ whose denominator are less than or equal to $n$. Here $p$ and $q$ are integers. For $n = 5$, the Farey sequence is $0/1, 1/5, 1/4, 1/3, 2/5, 1/2, 3/5, 2/3, 3/4, 4/5, and 1/1$. The next Farey sequence can be generated by adding a $(p+r)/(q+s)$ locking between $p/q$ and $r/s$ lockings of the previous sequence. In contrast to frequency locking motions the quasiperiodic motion with an irrational $\sigma$ values has only one $\Omega$ which corresponds to a given $\sigma$ and $K$ so Arnold tongues do not exist for quasiperiodic motions, i.e., they have infinitesimal widths.

The arguments mentioned above are true for $K \leq 1$ because the circlemap is differentiable in this region. At $K = 1$ the circlemap becomes nondifferentiable and
for \( K \geq 1 \) no unique inverse of the map exists. In Fig 3.1 Arnold tongues of frequency locking motions completely fill a \( K = 1 \) line and above \( K = 1 \) Arnold tongues with different \( \sigma \) overlap each other so \( \sigma \) is no longer uniquely defined. If \( K \) increases further, \( \sigma \) jumps around nearby winding numbers of overlapped tongues irregularly and it causes chaotic motions. For all quasiperiodic motions the motions become chaotic as soon as \( K \) reaches 1 so \( K = 1 \) is called the critical line in Fig. 3.2.

In this thesis I am interested in the quasiperiodic transition to chaos so behaviors of frequency lockings predicted by the circlemap will not be discussed further. It is well known that an irrational number can be approximated by a sequence of truncated continued fractions\(^6\) which are rational numbers:

\[
\sigma_g = \frac{1}{1 + \frac{1}{1 + \frac{1}{\ldots}}} = \frac{\sqrt{5} + 1}{2} = 0.6180\ldots
\]

(3.4)

where \( n_i \)'s are integers. Among irrational numbers we can think of one that is least approximated by rational numbers such that all \( n_i \)'s are 1;

\[
\sigma_g = \frac{1}{1 + \frac{1}{1 + \frac{1}{\ldots}}} = \frac{\sqrt{5} + 1}{2} = 0.6180\ldots
\]

(3.4)

This \( \sigma_g \) is the so-called golden mean and is approximated by the so-called Fibonacci numbers \( F_n \), which are defined by

\[
F_{n+1} = F_n + F_{n-1}; \ F_0 = 0, \ F_1 = 1; \ n = 0, 1, 2, \ldots
\]

(3.5)
and a sequence of rational numbers $\sigma_n$:

$$\sigma_n = \frac{F_n}{F_{n+1}} = \frac{F_n}{F_{n-1} + F_n}$$

(3.6)

which converges to the golden mean $\sigma_g$ with increasing $n$ such as

$$\sigma_g = \lim_{n \to \infty} \sigma_n .$$

(3.7)

Fig. 3.2 shows a calculated quasiperiodic route of the circlemap with golden mean winding number in $K - \Omega$ space. $\Omega_n$'s for small $n$ determined first from the circlemap in order to calculate $\Omega_g$ for a given $K$ and by using the fact that $\Omega_n$ converges to $\Omega_g$ for large $n$ following Eq. 3.8, $\Omega_g(K)$ can be extrapolated. As shown in the figure the route is not a straight line and $\Omega_g$ is monotonically decreasing with increasing $K$. At $K = 1$, I computed $\Omega_g = 0.6066610634701...$ by using $\Omega_n$'s up to $n=18$.

Quasiperiodic transition to chaos for golden mean winding number at $K = 1$ has been studied extensively by many authors\textsuperscript{1,2,3,7} and many universal properties of the transition we will discuss below have been predicted:

(1) The bare winding number $\Omega_n$ which corresponds to the winding number $\sigma_n$ approaches a constant as $n$ increases,

$$\lim_{n \to \infty} \frac{\Omega_n - \Omega_{n-1}}{\Omega_{n+1} - \Omega_n} = \delta$$

(3.8a)

where

$$\delta = -(\sigma_g)^2 = -2.61803... \text{ for } 0 \leq K < 1$$

(3.8b)

$$= -2.83362... \text{ for } K = 1 .$$
(2) The distance \( d_n \) from \( \theta = 0 \) to the nearest element of a cycle with \( \sigma_n \) scales as

\[
\lim_{n \to \infty} \frac{d_n}{d_{n+1}} = \beta ; \quad d_n = f^{F_n}(0) - F_{n-1}
\]

(3.9a)

where

\[
\beta = -(\sigma_g)^{-1} = -1.618... \quad \text{for } 0 \leq K < 1
\]

\[
= -1.289... \quad \text{for } K = 1.
\]

(3.9b)

(3) The power spectrum \( S(\omega) \) of \( \theta \) defined as

\[
S(\omega) = \frac{1}{\sqrt{N}} \sum_{j=0}^{N-1} f^j(\theta) \exp(-2\pi j \omega)
\]

(3.10)

displays a self-similar band structure between the main peaks which occur at every power of golden mean \( \sigma_g \). Fig. 3.3 shows a power spectrum \( P(f) \) and a normalized power spectrum \( P(f)/f^2 \) of the circlemap where \( P(f) = 20 \log_{10} S(\omega) \) and \( \omega = 2\pi f \). The self-similar band structure is more clearly seen in the normalized power spectrum. The appearance of peaks in the power spectrum at powers of Fibonacci numbers reflects that the motion is almost periodic after \( F_n \) iterations.

(4) The scaling function \( f(\alpha) \) of this route has an universal spectrum; \( f(\alpha) \) will be defined and discussed in more detail in the next section.

The properties mentioned above are universal in the sense that they hold as long as the nonlinearity in the circlemap is quadratic in \( \theta \). Experimentally the power spectrum of quasiperiodic motion with \( \sigma_g \) and \( K = 1 \) can be mostly easily tested and many experiments measured power spectra at near golden mean values\(^8\)-\(^11\). It turned out that few experimental spectra agree reasonably well with the theoretical prediction. Therefore it is still of importance to experimentally test
if more diverse systems follow predictions of the circlemap.

3.2. $f(\alpha)$ Spectrum

There have been much efforts to characterize the attractors of the motions of different dynamical systems. These attractors are usually very complicated fractal objects so lots of new concepts of dimensions were introduced for this purpose. Very recently this complicated, or multifractal nature of attractors has been fully understood with a scaling function $f(\alpha)$. In this section the meaning and the method to calculate scaling function $f(\alpha)$ is explained by using the circlemap.

One characterization of an attractor is to specify the probability distribution of states on the attractor in phase space. The most frequently used dimension in dynamical system theory is a Hausdorff dimension or fractal dimension $D_0$ defined as,

$$D_0 = \lim_{\varepsilon \to 0} \frac{\log N(\varepsilon)}{\log (1/\varepsilon)}$$

(3.11)

where $N(\varepsilon)$ is the number of non-empty boxes of size $\varepsilon$ required to cover the attractor. After the introduction of this fractal dimension a large number of other dimensions, such as information dimension, and correlation dimension, were introduced for possibly fuller or more readily calculable descriptions of a attractor. The disadvantage of these dimensions is that the structure of an attractor cannot be completely characterized by a single dimension because the local probability distribution on some part of the attractor is not perfectly the same as that of other part. The dimensions mentioned previously represent the average properties of an
attractor. Most attractors from experiments and theories have an interwoven structure of many different fractal dimensions.

The scaling function \( f(\alpha) \) of an attractor is a fractal dimension of the scaling index \( \alpha \) and \( f(\alpha) \) constitutes a spectrum of dimensions. This concept was first introduced as the generalization of fractal dimensions by Halsey et al\(^{14} \). To understand the \( f(\alpha) \) spectrum let us use the circlemap. The circlemap with \( \Omega_g = 0.606661063... \) and \( K = 1 \) generated the time series of \( N_{\text{total}} = 10^4 \) consecutive values of \( \theta \) from \( \theta_0 = 0 \). The circle is then divided into \( n_{\text{cell}} = 10^3 \) cells with size \( L = 1/n_{\text{cell}} = 10^{-3} \) and the number of \( \theta \) in the i-th cell located between \((i-1)L\) and \(iL\), which is denoted as \( N_i(L) \), is calculated. The probability \( P_i(L) \) of the i-th cell is defined as

\[
P_i(L) = \lim_{L \to 0} \frac{N_i(L)}{N_{\text{total}}} , \quad (3.12a)
\]

\[
N_{\text{total}} = \sum_{i=0}^{n_{\text{cell}}} N_i(L) . \quad (3.12b)
\]

Fig. 3.4 shows a calculated probability distribution on the circle of the circlemap with winding number \( \sigma_g \) and \( K = 1 \) by following the method described above. The figure shows a very complicated probability distribution which does not look self-similar. The scaling index \( \alpha_i \) is a kind of dimension for singularity of a probability \( P_i(L) \) assigned to the i-th cell of length \( L \) and is defined as

\[
P_i(L) \propto L^\alpha_i \quad \text{as} \quad L \to 0 . \quad (3.13)
\]

The probability distribution can be represented by the scaling indices assigned to each cell. The cells in valleys in the figure scale quite differently from those in
peaks as $L$ varies. The probability distribution in Fig. 3.5(a) of the circlemap therefore can be decomposed into subsets with scaling index whose value is between $\alpha$ and $\alpha + d\alpha$. Fig. 3.5(b) and 3.5(c)\textsuperscript{15} show schematically distributions of two different values of $\alpha$ on the circle. For sharp peaks as in Fig. 3.5(b) the probability distribution grows slowly with increasing $L$ so $\alpha$ is small. For valleys as in Fig. 3.5(c) the probability increases very rapidly with increasing $L$ so $\alpha$ is large. As shown in bottom of Fig. 3.5(b) and 3.5(c) the scaling indices between $\alpha$ and $\alpha + d\alpha$ are distributed over the circle and they form a structure with fractal dimension $f(\alpha)$ which can be calculated by using Eq. 3.11. This means that the probability distribution of the circlemap in Fig. 3.4 can be represented by the spectrum of fractal dimensions $f(\alpha)$ which characterizes the distributions of different $\alpha$.

Now I will discuss an algebraic method of calculating $f(\alpha)$ from a probability distribution. If the attractor is divided into pieces of size $L$ as explained before, the distribution of scaling indices on the attractor can be assumed to have the form\textsuperscript{14}:

$$N(\alpha) = d\alpha \rho(\alpha) L^{-f(\alpha)}$$  \hspace{1cm} (3.14)

where $N(\alpha)$ is a number of boxes with scaling indices between $\alpha$ and $\alpha + d\alpha$ and $\rho(\alpha)$ is a continuous function of $\alpha$. Halsey et al\textsuperscript{14} introduced a partition function $\Gamma(q,L)$ which is defined as

$$\Gamma(q,L) = \sum_{i=1}^{n_{\text{cell}}} P_i(L)^q$$  \hspace{1cm} (3.15)

where summation is over all cells on the attractor. The parameter $q$ is an arbitrary number and controls the contribution of the specific value of $\alpha$ to the partition function. If $q$ has a large positive value, boxes with large probability, or dense-
density cells, dominate the partition function. If \( q \) has a large negative value, cells with small probability, or rare-density, dominate the partition function. Summation in the definition of \( \Gamma(q,L) \) can be changed into an integral form by using Eq. 3.13 and Eq. 3.14 as follows:

\[
\Gamma(q,L) = \int d\alpha N(\alpha) P(L,\alpha)^q 
\]

or,

\[
\Gamma(q,L) = \int d\alpha \rho(\alpha) L^{q\alpha - f(\alpha)}. 
\]

For \( L \ll 1 \), the integrand in Eq. 3.16(b) is dominated by the value of \( \alpha \) that minimizes the exponent, \( q\alpha - f(\alpha) \). The conditions that minimize the exponent give the relations:

\[
\alpha = \frac{d \tau(q)}{dq}, 
\]

and

\[
f(\alpha) = q\alpha - \tau(q) 
\]

where \( \tau(q) \) is the exponent defined as;

\[
\tau(q) = q\alpha - f(\alpha) = (q-1)D_q. 
\]

\( D_q \) is the generalized dimension coined by Hentschel and Procaccia\(^{13} \). For example \( D_0 \) is a usual fractal dimension, \( D_1 = \) information dimension, and \( D_2 = \) correlation dimension, etc. The \( f(\alpha) \) spectrum has a maximum value at \( q = 0 \) and \( f(\alpha) = D_0 \), the fractal dimension of the attractor. It has an infinite slope at \( q = \pm \infty \).

Halsey \textit{et al}\(^{14} \) have discussed a numerical method of calculating the \( f(\alpha) \) spectra for models of different dynamical systems. They used a recursive method for the spectrum of quasiperiodic transition to chaos of the circlemap with golden
mean winding number. Starting from $\theta_0 = 0$ the time series of $\theta_i$ generated up to $i = F_{17} = 2584$ and Markovian partitions $l_i = \theta_i + F_{16} - \theta_i$ were defined as natural scales with probability $P_i = 1/F_{17} = 1/2584$ instead of the partitions of equal size discussed above. The partition function is defined as,

$$\Gamma(q, \tau, l) = \sum_{i=1}^{F_{16}} \frac{(P_i)^q}{(l_i)^q}$$

(3.19)

and $\tau(q)$ is defined by

$$\Gamma(q, \tau(q), l) = 1.$$  

(3.20)

The $\tau(q)$ can be calculated numerically by using Newton’s method even if its convergence for large $q$ is slow. To improve the convergence of $\tau(q)$, a ratio trick that assumes the relation

$$\frac{\Gamma(q, \tau, l(F_{17}))}{\Gamma(q, \tau, l(F_{16}))} = 1$$

(3.21)

is used. After $\tau(q)$’s are obtained the $f(\alpha)$ spectrum is determined by using Eq. 3.17(b).

The universal properties of the circlemap for quasiperiodic transition to chaos with golden mean winding number discussed in Section 3.1 determine $\alpha_{-\infty}$ analytically. As in Eq. 3.9 the distance around $\theta = 0$ which corresponds to the most rarefied region on the circle scales down by a universal factor $\beta = -1.289...$ at $K = 1$ so $l_{-\infty} \sim \beta^{-n}$ when $\theta_i$ is truncated at $F_n$. The corresponding probability scales as $P_{-\infty} \sim 1/F_n - (\sigma_g)^n$, leading to

$$\alpha_{-\infty} = D_{-\infty} = \frac{\log \sigma_g}{\log \beta^{-1}} = 1.8980...$$

(3.22)
At $\theta = 0$ the circlemap for $K=1$ has a zero slope with a cubic inflection points so $\theta = 0$ maps onto the most concentrated region and therefore $I_\infty \sim \beta^{-3n}$. The probability scales as $P_\infty \sim 1/F_n \sim (\sigma_g)^n$ so that we get

$$
\alpha_\infty = D_\infty = \frac{\log \sigma_g}{\log \beta^{-3}} = 0.6326... 
$$

The numerically calculated $f(\alpha)$ spectrum of the circlemap following the recursive method explained above with $i = F_{17} = 2584$ is shown in Fig. 3.6. To show the uniqueness of the $f(\alpha)$ spectrum of quasiperiodic transition to chaos the spectrum of period-doubling transition to chaos from the logistic map in Eq. 1.1 is shown at the left. The $f(\alpha)$ curve is unique at the onset of quasiperiodic transition to chaos with golden mean winding number. The values of $\alpha_{\pm\infty}$ in the figure agree quite well with the analytical values 0.6326 and 1.8980 and the fractal dimension $D_0$ of the circlemap which is $f(\alpha)$ at $q = 0$ is in excellent agreement with the theoretical value 1.

The recursive method to calculate the spectrum is almost impossible to apply to experimental data. Markovian partitions defined from experimental data cannot have such fine details as the circlemap has because of finite resolution of measured data. Extrinsic noise makes the measured partitions even worse. Noise particularly affects the fine details of the partitions significantly and smooths them. This smoothing consequently reduces the range of $\alpha$ and gives significant deviation of experimentally measured $f(\alpha)$ spectrum from the universal spectrum. In next chapter I will discuss a better method of calculating the $f(\alpha)$ spectrum from
experimental data.
REFERENCES. CHAPTER 3


FIGURE CAPTIONS

Fig. 3.1. Phase diagram of the circlemap in $K - \Omega$ space. Arnold tongues of dominant frequency lockings are shown. This is from Ref. 3.

Fig. 3.2. Quasiperiodic route with golden mean winding number $\sigma_g$. The bare winding number $\Omega_g$ was found to be $0.0.606661063...$ from the numerical calculation.

Fig. 3.3. Power spectrum of circlemap with $\sigma_g$ and $K = 1$. A fast Fourier transform was used to compute the power spectrum $P(f)$ in (a) and the normalized power spectrum $P(f)/f^2$ in (b) from $2^{11}$ values of $\theta$ generated from the circlemap.

Fig. 3.4. Probability distribution on circle of a circlemap of quasiperiodicity with $\sigma_g$ and $K = 1$. The cell size is $1 \times 10^{-3}$ and total number of $\theta$ used here is $1 \times 10^4$.

Fig. 3.5. Decomposition of a probability distribution into subsets with different scaling indices $\alpha$. The scaling index $\alpha$ is a kind of dimension for singularity of a probability and a distribution of same value of $\alpha$ in (b) and (c) forms a structure with a fractal dimension $f(\alpha)$.

Fig. 3.6. The numerically obtained $f(\alpha)$ spectrum of the circlemap with $\sigma_g$ and $K = 1$ by using a recursive method in Ref. 14. To show the uniqueness of the $f(\alpha)$ spectrum of quasiperiodic transition to chaos the spectrum of period-doubling transition to chaos is shown at the left.
Figure 3.3
Figure 3.4
Figure 3.5
Figure 3.6

The graph shows the function $f(\alpha)$ plotted against $\alpha$. The x-axis represents $\alpha$ ranging from 0.2 to 2.0, while the y-axis represents $f(\alpha)$ ranging from 0 to 1.2.

- **Quasiperiodicity**: Indicated by an arrow on the right side of the graph.
- **Period-doubling**: Indicated by an arrow on the left side of the graph.
CHAPTER 4. EXPERIMENTAL RESULTS AND DISCUSSIONS

In this chapter I will present experimentally measured power spectra and the $f(\alpha)$ spectrum of the helical instability of electron-hole plasma in Ge crystal at the onset of chaos from quasiperiodic motion with golden mean winding number. As discussed in Chapter 2 the ac modulation voltage from a HP synthesizer/function generator is applied to the side contacts of the sample. The ac modulation provides a second oscillator in addition to the spontaneous current oscillation as a first oscillator. To increase the S/N ratio of the quasiperiodic signal arising from the interaction between the ac modulation voltage and the spontaneous current oscillation the optimum values of experimental parameters $(V_0, B_0, \theta)$ for the spontaneous current oscillation were used. In the experiment these were $V_0 \sim 12$ V, $B_0 \sim 8$ kG, and $\theta = 1^\circ$ to first order; the value of $V_0$ was refined as below. For the ac modulation experiment there are two additional parameters; frequency $f_1$ and voltage $V_1$ of the ac modulation. The frequency $f_0$ of the spontaneous current oscillation was about $170 - 180$ kHz and the ratio of two frequencies $f_1/f_0$ was set closely to golden mean value ($\sigma_g = 0.6180...$) so $f_1$ should be about $100$ kHz. To find the optimum values of parameter values this time the phase diagrams of the motion in $V_0-V_1$ space for a given $f_1$ were plotted out. They were mapped by repeating the steps of varying $V_0$ continuously and fixing $V_1$ to a new value so as to maintain the desired ratio of $f_1/f_0$. The real-time image of the Poincare section were generated on a Tektronix CRT display unit by an sample and hold analog circuit and
the power spectrum of the signal were used to determine the types of the motion of the signal, (either frequency locking or quasiperiodicity) and its winding number which is $f_1/f_0$. The frequency ratio was determined by a LSI-11 computer by reading frequencies from a spectrum analyzer through IEEE 488 bus. Near a quasiperiodic route with winding number $\sigma_g$ many Arnold tongues of frequency locking with ratio of successive Fibonacci numbers, for example $2/3, 3/5, 5/8, 8/13, 13/21$, could be observed; see Fig. 4.1. Depending on the choice of parameter values there are substantial difference in the phase diagrams; the width of the Arnold tongues of Fibonacci frequency locking, the maximum number of Fibonacci frequency lockings, and appearance of other type of frequency lockings differing from the Farey sequence. When frequency lockings from Farey sequence near $\sigma_g$ appeared, the width of the Arnold tongues of Fibonacci frequency locking was usually small so the quasiperiodic route with $\sigma_g$ was hard to locate. The maximum number of Fibonacci frequency lockings is important because it indicates a good S/N ratio. At the optimum condition the $13/21$ locking was clearly observed but for other conditions it was extremely difficult to distinguish from nearby quasiperiodic states.

The phase diagram measured at the optimum value of parameters is shown in Fig. 4.1, here $\theta = 1^\circ, B_0 = 8.092$ kG, $f_1 = 107.0$ kHz, and $V_0 \sim 12$ V. The range of $V_1$, which corresponds to magnitude of nonlinearity, is from 0 to 1.5 V. The dotted line is the quasiperiodic route to chaos with winding number $\sigma_g$ as determined from measuring $f_1/f_0 = 0.6180...$ to 2 part in $10^4$. The solid horizontal line C in
the figure is the critical line, e.g., $K = 1$ line in the circlemap, above which the motions of the signal are chaotic. This critical line was chosen by watching the Poincare section of quasiperiodic attractors which has a smooth and continuous trajectory below the critical line. The Poincare section just above the critical line begins to wrinkle, indicating the breakup of the invariant torus. This method was more sensitive from the appearance of weak chaos in the power spectrum. Far beyond the critical line the motion becomes strongly chaotic and the section develops quite complicated folding structure and eventually destroys the torus.

In this chapter the measurement of the power spectrum and the $f(\alpha)$ spectrum at a crossing point of the critical line and the dotted line in Fig. 4.1 will be discussed to test the universality of the circlemap; it will be assumed that this crossing point corresponds to $K = 1$ and $\Omega = \Omega_g$ in the theory.

4.1. Power spectra

The power spectra were taken from the HP analog spectrum analyzer (model 3585A) and were stored onto a hard disk of a LSI-11 computer for averaging. Averaging is very useful to see the details of low frequency peaks which are usually buried in background noise. It reduces the background noise and also partially compensates for the fluctuation of peak positions due to fluctuation of the ratio $f_1/f_0$. I found that the averaged ratio could be maintained to $\sigma_g$ to approximately 2 parts in $10^4$ although $f_1$ has an stability of 1 part in $10^6$, and $f_0$ of about 1 part in $10^5$. Fig. 4.2(a) shows the measured power spectrum $P(f)$, and Fig. 4.2(b)
shows the normalized power spectrum $P(f)/f^2$ of the current oscillation $I(t)$ as a function of frequency at the onset of quasiperiodic transition to chaos ($V_0 = 11.820$ V, $V_1 = 1.038$ V, $f_1 = 107.0$ kHz, $B_0 = 8.092$ kG) for $f_1/f_0$ set as closely as possible to $\sigma_g$. The power spectrum is an average of 10 power spectra taken at essentially the same condition. The peak positions in the power spectrum matched excellently up to 6-th power of $\sigma_g$ (see Fig. 4.2(a)). This indicates the accuracy of setting of frequency ratio during the data acquisition. The normalized power spectrum shows the universality of the quasiperiodic motion more clearly because it shows self-similar band structures between the power of $\sigma_g$ as shown in Fig. 3.3(b). Near $\sigma_g$, 15 peaks could be resolved and their magnitude agreed with the circlemap within 15%. This confirms that the transition to chaos by quasiperiodicity for the plasma oscillation is reasonably well described by the circlemap. The deviation between experimental and theoretical spectrum becomes larger when the magnitude of peaks is smaller and the frequency is lower and it seems to be due to noise in the system. This was confirmed from study of noise effect on power spectrum and $f(\alpha)$ spectrum of circlemap and it will be discussed later in Chapter 5.

4.2 $f(\alpha)$ Spectrum

Although the power spectrum can partially test the validity of the circlemap to describe the motion the real test comes from comparison of $f(\alpha)$ spectrum. As described in Section 2.3 a consecutive time series of the 98000 quasiperiodic
current oscillation $I(t)$, sampled at every triggering pulse with period $\tau_1 = 1/f_1$, were taken at the parameter values given above for each run of the experiments. The n-dimensional Poincare section can be reconstructed from the experimental time series by plotting points $(I(t_i), I(t_{i+1}), I(t_{i+2}), \ldots, I(t_{i+n-1})$) where $t_i = i \tau_1$. Fig. 4.3 shows the reconstructed two-dimensional Poincare sections, plots of $I(t_i)$ vs. $I(t_{i+1})$, below, on, and above the critical line. Below the critical line the Poincare section is smooth although distribution of points along the attractor is not uniform which indicates that it is a multifractal object where the scaling index $\alpha$ is distributed. On the critical line the attractor develops small scales foldings at several places and the distribution of points becomes more clustered. Above the critical line it develops larger scale foldings and its shape is more distorted indicating the breakup of the invariant torus.

Now I will discuss methods to derive a $f(\alpha)$ spectrum from an experimental time series of a single dynamical variable. The first method has been reported by Jensen et al. It is called a recurrence time method. Starting from the point $x_i$ on the reconstructed attractor, the recurrence time, $m_i$, is the number of steps along the time series required before a point returns to within a given length $l$ of the starting point. The probability around the starting point is then assumed to be defined as,

$$P_i(l) = (m_i)^{-1}$$

(4.1)

and the partition function defined in Eq. 3.15 is estimated as,

$$\Gamma(q,l) = \langle P_s(L)^{q-1} \rangle_{av} = \langle m_i^{1-q} \rangle_{av}$$

(4.2)
where the averaging is over consecutive elements of a trajectory on the attractor.

The averaging in Eq. 4.2 is to smooth the fluctuation of recurrence times and give an efficient calculation of \( f(\alpha) \) spectrum in spite of using a small number of data for the calculation. The \( \tau(q) \) is derived from the partition function by

\[
\Gamma(q,l) = l \tau(q)
\]  \hspace{1cm} (4.3)

from Eq. 3.18, or the slope of a \( \log \Gamma(q,l) \) vs. \( \log l \) curve

\[
\tau(q) = \lim_{l \to 0} \frac{\log \Gamma(q,l)}{\log l} \hspace{1cm} (4.4)
\]

By varying \( q \) we can get corresponding \( \tau(q) \). Afterwards the \( f(\alpha) \) spectrum can be determined from \( \tau(q) \) by using Eq. 3.17(a) and 3.17(b).

The second method differs from the recurrence time method only in defining probability. Instead of using recurrence time method it uses a literal meaning of probability so I will call it the averaging method; this method was developed by Gwinn and Westervelt. The attractor is divided into segments of length \( l \). The probability of the segment at \( s \) along the attractor is defined as

\[
P_s(l) = \frac{N_s(l)}{N_{\text{total}}}
\]  \hspace{1cm} (4.5)

where \( N_{\text{total}} \) is the total number of points on the attractor and \( N_s(l) \) is the number of points on the segment at \( s \). Except for this difference later steps to derive the \( f(\alpha) \) spectrum are the same as the recurrence time method. Fig. 4.4 shows \( \log \Gamma(q,l) \) vs. \( \log l \) plots for \( q = 2 \) and \( -3 \) by using consecutive values of 5000 \( \theta \) from the circlemap with \( K = 1, \sigma_g, \) and \( \theta_0 = 0 \). Fig. 4.4(a) was generated by using the recurrence time method and Fig. 4.4(b) used the averaging method. As shown in the figure, the former makes steps or wigglings but the latter is much smoother.
and has a longer straight portion of the curves which means the slope \( \tau(q) \) can be determined easily over large range of \( l \). Such worse features shown in Fig. 4.4(a) comes from the fact that the equivalence of probabilities \( P_i(l) \) and inverse of the recurrence time \( m_i \) is not generally valid for a limited number of data. Particularly lack of the equivalence becomes clear when the data is contaminated by noise.

Another thing we should think about to calculate \( f(\alpha) \) spectrum is the definition of length \( l \). In experiments the attractors are reconstructed by the method explained above for a given embedding dimension. In the recurrence time method length was defined by using Euclidean geometry:

\[
l = \left( \sum_{i=1}^{N_d} [x_m(i) - x_n(i)]^2 \right)^{1/2}
\]

(4.6)

when the embedding embedding is \( N_d \). For the averaging method the best way to define length is to measure the distance along the attractor, not along a straight line between points.

In the calculation of the experimental \( f(\alpha) \) spectrum in this section the averaging method with definition of distance along the attractor was used. In order to define the distances a high-probability path along the attractor should be generated. This path was generated with a computer program in the Appendix. The program first assigns the minimum length scale, which is the diameter of a circle used to find the path and determines the starting point on the attractor. Then it checks whether the starting point lies in a high probability region near the point. If not, it finds the point near the center of the highest probability region and assigns
the point as a new starting point. After the starting point is decided, it puts circles with a given diameter around that and searches the next nearby position of the new circle in which maximum number of points is contained. After the new position of the circle is found, the previous step is repeated until the circle returns all the way to the starting point.

To find the path, grids can be used instead of circles but I found that using circles gave a smoother probability distribution along the path than using grids because grids have only four choices of direction. The size of circle in this algorithm is very important. If it is too small, the probability distribution is affected significantly by noise. If it is too large, details of the probability distribution cannot be resolved. The criterion of choosing an appropriate size is to set the smallest probability, or the smallest number of points in a circle. If the smallest probability found by using a certain size of circle is smaller than the criterion, increase the size and repeat running the program again until it is larger than the criterion.

Fig. 4.5 is the high-probability path found for the critical attractor shown in Fig. 4.3(b) by using the program explained above. When the whole length of the path is assumed to be one, the size of cells used is about $1.3 \times 10^{-3}$. The total number of points along the path is about 85000 and Fig. 4.6 shows the number of points in each cell, e.g., the probability distribution, along the path. The figure shows many peaks and valleys as in Fig. 3.4 of the circlemap but it is hard to see the direct correlation of the two figures. Once the probability distribution is determined, the calculation of $f(\alpha)$ spectrum is straightforward by applying the
averaging method.

The plots of log $\Gamma(q,l)$ vs. log $l$ for $q = -2, 0, \text{and } 4$ are shown in Fig. 4.7. Similar plots were generated by varying $q$ from $-4$ to $8$ with increasing $0.1$ at a time. The slope of the curve gives $\tau(q)$. I found four things from the plots:

(1) When the absolute value of $q$ is large, the plots show two or three different slopes depending on the values of $L$.

(2) Only one slope shown with a straight line is $\tau(q)$ which corresponds to that of the circlemap; and it will be called a right scaling region.

(3) As the absolute value of $q$ increases, $L$ range of right scaling decreases. The rate of decrease of range $L$ for negative $q$ is much faster than for positive $q$.

(4) For negative $q$, small $L$ has $\tau(q)$ of value $q - 1$. This is due to noise which randomizes details of probability distribution of small scales. The next chapter about noise effects on $\Gamma(q,L)$ for different $q$ will explain the above properties.

The measured $f(\alpha)$ spectrum from the experimental data is shown in Fig. 4.8 as dots with error bars together with the spectrum computed from the circlemap at the onset of the quasiperiodic transition with golden mean winding number to chaos. The range of $q$ used to get $\tau(q)$ from the experimental data was $-4 \leq q \leq 8$. The error bars were estimated by varying the fitting range of $L$. As shown in the figure, the experimental spectrum for positive $q$ (left part of the $f(\alpha)$ spectrum) are in good agreement with the theory. For negative $q$ (right part of the spectrum) the scaling range for $\Gamma(q,L)$ became narrower as the magnitude of $q$ increased. This
fact explains the large error bars in that region. This seems to be mainly due to noise and the intrinsic limitation of stability of frequency $f_0$ of the current oscillation and the deviation of the ratio $f_1/f_0$ from $\sigma_g$ during the experiments. The deviation of the measured spectrum from that of the circlemap in the negative $q$ region is less than 5%. Although a slight decrease in driving amplitude of ac voltage $V_1$ from the critical value reduced the range of $\alpha$ noticeably we could not see the collapse of the $f(\alpha)$ spectrum under the critical line which was predicted by Arneodo and Holschneider\textsuperscript{4}. 
REFERENCES. CHAPTER 4


4. This result has been reported earlier: Y. Kim, Phys. Rev. A39, 4801 (1989).

FIGURE CAPTIONS

Fig. 4.1. Phase diagram of the electron-hole plasma instability in Ge driven by an ac voltage. \( B_0 = 8.092 \text{ kG}, f_1 = 107.0 \text{ kHz} \). The Arnold tongues of frequency locking with Fibonacci numbers are shown. The dotted line shows the quasiperiodic route to chaos with a winding number \( \sigma_g \). A horizontal solid line \( C \) is the critical line above which quasiperiodic attractors are chaotic. The crossing point \( X \) corresponds to transition to chaos at the golden mean winding number.

Fig. 4.2. (a) Experimental power spectrum \( P(f) \) and (b) normalized power spectrum \( P(f)/f^2 \) of current oscillation \( I(t) \) at the onset of quasiperiodic transition to chaos. The sample was driven at \( V_0 = 11.820 \text{ V}, V_1 = 1.028 \text{ V}, f_1 = 107.0 \text{ kHz} \), and \( B_0 = 8.092 \text{ kG} \). Winding number was about \( \sigma_g = 0.618034\ldots \). Compare this spectra with those in Fig. 3.3.

Fig. 4.3. Reconstructed two-dimensional Poincare sections, plots of \( I(t) \) vs. \( I(t+\tau_1) \) where \( \tau_1 = 1/f_1 \), of a quasiperiodic trajectory with a winding number \( \sigma_g : f_1 = 107.0 \) kHz, \( B_0 = 8.092 \text{ kG} \), and (a) below \( (V_0 = 11.854 \text{ V}, V_1 = 0.998 \text{ V}) \), (b) on \( (V_0 = 11.820 \text{ V}, V_1 = 1.028 \text{ V}) \), and (c) above \( (V_0 = 11.773 \text{ V}, V_1 = 1.077 \text{ V}) \) the critical line of Fig. 4.1.

Fig. 4.4. Comparison of calculation methods of a \( f(\alpha) \) spectrum for 5000 data points from the circlemap with \( \sigma_g \) and \( K = 1 \). (a) recurrence time method, (b) averaging method.
Fig. 4.5. High-probability path of the critical attractor shown in Fig. 4.3(b). It was found by using a program explained in Section 4.2, and given in the Appendix.

Fig. 4.6. Probability distribution along the high-probability path shown in Fig. 4.5. The size of cells is about $1.3 \times 10^{-3}$ when the total length of the path is assumed to be 1. The total number of points along the path is about 85000. The computer program is given in the Appendix.

Fig. 4.7. Plots of $\log \Gamma(q,l)$ vs. $\log l$ for $q = -2, 0, \text{ and } 4$ for the probability distribution shown in Fig. 4.6.

Fig. 4.8. The $f(\alpha)$ spectrum calculated by using the probability distribution shown in Fig. 4.6 is shown as dots with error bars. The solid line is the $f(\alpha)$ spectrum computed from the circlemap.
\[ f_1 = 107.0 \text{ kHz} \]

\[ f_1/f_0 = 3/5 \]

\[ \text{Figure 4.1 XBL 8810-3687} \]
Figure 4.2
Figure 4.3
Figure 4.4
Figure 4.5

Figure 4.6
Figure 4.7
Figure 4.8
CHAPTER 5. NOISE EFFECTS

Recent years there have been several experiments which reported \( f(\alpha) \) spectra of quasiperiodic\(^1\) and period-doubling\(^4,6\) transitions to chaos. Dynamical systems of the experiments are; a forced Rayleigh-Bernard system\(^1,6\), a driven diode resonator\(^4\), a driven relaxational oscillator\(^3\), electronic transport in Ge at low temperature\(^2\), and helical wave instability of electron-hole plasma in Ge\(^5\). Among many chaos experiments these are a few cases for which the \( f(\alpha) \) spectrum has been measured. Also the quality of agreement of the measured spectrum with the circlemap prediction was generally not satisfying. It is therefore still of interest to measure the \( f(\alpha) \) spectrum for other physical systems. In this Chapter the noise effect on the power spectrum \( P(f) \) and the \( f(\alpha) \) spectrum of the circlemap is discussed to understand the difficulties of high-precision experimental measurements of the spectra and from this numerical study I will show that the reduction of noise in experiments is essential to derive accurately the \( f(\alpha) \) spectrum of the system of interest.

5.1. Power spectra

During experiments on quasiperiodic or period-doubling motions the measured winding number \( f_1/f_0 \), or the nonlinear coupling \( V_1 \) in the experiment can fluctuate due to noise in the system, or electronic instruments. Noise from these sources contaminates the measured data and deviates the measured spectra from that of a noiseless system. In this chapter noise is generated by a computer pro-
gram using a random number generator and is added to the values of $\theta$ generated from the circlemap for with $K = 1$ and $\Omega = \Omega_q$ in order to simulate the noise in the experimental system. For every iteration of the circlemap noise is added as follows:

$$\theta_{n+1} = f(\theta_n) + noise = f(\theta_n) + \epsilon \xi_n$$

(5.1)

where the quantity $\xi_n$ is a random variable with an even distribution of unit width, and $\epsilon$ is a parameter that controls the amplitude of the noise. The circlemap is denoted here by $f(\theta)$. Our results show that there is no preference of noise form: application of Gaussian noise with standard deviation $\epsilon$ and zero mean to the circlemap has the same effect on the spectra as Eq. 5.1.

Fig. 5.1 shows the noise effect on frequency lockings. To see the effect on frequency lockings $\theta_n$ for 1000 iterations of the circlemap for frequency lockings $8/13$ and $13/21$ at each of 200 increments of $\sigma$ were calculated for $K = 1$ and $\Omega = 0.60488...$ and $0.60729...$ respectively. The figure shows that when the noise amplitude $\epsilon$ increases, higher order frequency lockings first disappear completely whereas lower order frequency lockings sustain. This implies that experimentally higher order frequency lockings cannot be distinguished from quasiperiodic trajectory in the presence of noise because the dots on the Poincare section broadened into lines with some width and finally they touch each other to form a continuous curve. In our experiment frequency lockings whose order is higher than $13/21$ could not be observed. If the noise can be reduced further, higher order frequency lockings will be distinguished from quasiperiodic trajectories. This indicates the
clue to choose the optimum parameter values in the experiment.

The normalized power spectra of circlemap with $K = 1$ and golden mean winding number for different noise levels are shown in Fig. 5.2. The figure shows just one band structure between $\sigma_g$ and $\sigma_g^2$. We can see three effects of noise on power spectrum:

(1) Because of increase of background noise level weak peaks are buried in the background so the number of peaks is reduced. Fig. 5.2(a) has more weaker peaks than Fig. 5.2(b) and 5.2(c).

(2) Increase of noise also kills moderate amplitude peaks before they are buried in the background. This further reduces the number of peaks above the background.

(3) The magnitude of weaker peaks decreases faster than that of stronger peaks with increasing noise. This explains the fact that the deviation between measured magnitude of peaks and that of theory is larger for weaker peaks.

For moderate noise level the positions of peaks in the power spectrum are almost the same as those with no noise.

5.2. $f(\alpha)$ spectrum

The $f(\alpha)$ spectrum is directly related to the probability distribution of points on the attractor. It can be expected that the presence of noise may smooth the sharp valleys or peaks in Fig. 3.4 because it randomizes the small scale structures in the probability distribution. Therefore for small length scales the probability distribution is characterized by the noise, and not by the properties of the
underlying noise-free distribution. Fig. 5.3 illustrates the noise effect on the probability distribution for the circlemap at the onset of chaos of golden mean quasi-periodic transition. As shown in the figure, the noise fills in valleys and broadens the weaker peaks, but it appears to have little effect on the sharp peaks which corresponds to densely populated areas. When noise dominates the signal, the probability at small scales is uniform and $P(L) \propto L$ holds true. This implies that for small scales

$$\Gamma(q,L) = \left\langle P_s(L)^{q-1} \right\rangle_{av} \propto L^{q-1}$$

(5.2)

for $L \ll 1$ from Eq. 4.2, therefore $\tau(q) \sim d\log \Gamma(q,L) / d\log L$ has a value of $q-1$. The log $\Gamma(q,L)$ vs. log $L$ curves therefore must show a change of slope, $\tau(q)$, from $q-1$ to a right scaling value due to existence of noise as $L$ increases.

The Fig. 5.4 shows the noise effect on the plots of log $\Gamma(q,L)$ vs. log $L$ for $q = -5$ and 5. Each curve has distinct two values of $\tau(q)$. The slope for large $L$ has right scaling but another for small $L$ has $q-1$, which is the noise characteristic. For negative $q$ the right scaling region decreases very rapidly with increasing noise level whereas the right scaling region for positive $q$ survives quite well for large noise. Same reduction of the range of right scaling regions has been reported for the calculation of a correlation dimension of attractors. Therefore the reduction of scaling ranges is a generic property of the dynamical systems and its reduction rate is larger for higher order dimensions. It should be noticed that scaling exponents $\tau(q)$ in the plots do not change except narrowing of the scaling ranges even if noise contaminated the time series. This can explain the poor
agreement and the large error bars of the measured \( f(\alpha) \) spectrum of attractors of noisy experimental data. When noise is present in the experimental system, it causes destruction of the underlying fine structure of the attractor so randomization of the probability distribution at small scales occurs. The effect of noise also appears in the \( \tau(q) \) by showing the change of slopes in the log \( \Gamma(q,L) \) vs. log \( L \) curves. When the noise amount in the system is moderately small, its \( f(\alpha) \) spectrum can be extracted from the noise-contaminated data but details of the spectrum for negative \( q \) may be lost more than for positive \( q \). When the noise is relatively large, the scaling region is very small or almost none so that determination of the \( f(\alpha) \) spectrum would be impossibly difficult.
REFERENCES. CHAPTER 5


FIGURE CAPTIONS

Fig. 5.1. Noise effect on frequency lockings. (a) 8/13 locking: \( K = 1, \Omega = 0.60488... \) was used. The locking structure is broadened by noise but is sustained. (b) 13/21 locking: \( K = 1, \Omega = 0.60729... \) was used. The locking structure completely disappears when \( \varepsilon \) is greater than about \( 2\times10^{-3} \).

Fig. 5.2. The normalized power spectra of circlemap at \( K = 1 \) and golden mean winding number for different noise levels; (a) \( \varepsilon = 0 \) (no noise), (b) \( \varepsilon = 10^{-4} \), and (c) \( \varepsilon = 10^{-3} \).

Fig. 5.3. Probability distribution of the circlemap at \( K = 1 \) and golden mean winding number for different noise levels; (a) \( \varepsilon = 0 \) (no noise), (b) \( \varepsilon = 10^{-4} \), and (c) \( \varepsilon = 10^{-3} \).

Fig. 5.4. Log \( \Gamma(q, L) \) as a function of log \( L \) for different noise levels and different \( q \). The curves are generated from \( 10^4 \) values of \( \theta_n \) from the circlemap at \( K = 1.0 \) and \( \Omega_g \). The noise amplitude \( \varepsilon \) for the curves in (a) and (b) is: (1) 0 (no noise), (2) \( 2\times10^{-5} \), (3) \( 2\times10^{-4} \), and (4) \( 2\times10^{-3} \), (5) \( 2\times10^{-2} \).
Figure 5.1
Figure 5.2

(a)

\( \log \frac{P(f)}{f^2} \)

(b)

\( \log \frac{P(f)}{f^2} \)

(c)

\( \log \frac{P(f)}{f^2} \)
Figure 5.3
Figure 5.4

(a) \( q = 5.0 \)

(b) \( q = -5.0 \)
CHAPTER 6. CONCLUSIONS

The helical density instability of electron-hole plasma in a n-type Ge crystal under ac modulation has been studied in order to compare it with the universal predictions of power spectrum and \( f(\alpha) \) spectrum of the transition from quasiperiodicity to chaos of a simple one-dimensional model, the circlemap. The chaos phenomena of the helical instability in this crystal was discovered previously in our laboratory by Held and Jeffries. The spontaneous current oscillation due to the helical instability above the threshold dc voltage across the sample provides a signal with a large S/N ratio and a highly stable frequency. It was found that the ac modulation voltage across side probes produced a quasiperiodic motion of the current through the sample. The winding number and the nonlinearity of the quasiperiodicity could be controlled by changing the frequency and the voltage of the ac modulation respectively. Because of a convenient and stable frequency ratio this system provides an excellent test of universal properties of the quasiperiodic prediction to chaos predicted by the circlemap. The measured phase diagrams showed the general features of the circlemap: Arnold tongues of frequency locking motions; quasiperiodic motions in between the Arnold tongues; and a critical line above which begins folding the Poincare section.

By adjusting the winding number of the current signal, which is the ratio of the ac modulation frequency to the frequency of the spontaneous current oscillation around golden mean \( \sigma_g = (\sqrt{5} + 1)/2 \), the phase diagram showed Arnold
tongues of frequency locking with Fibonacci sequence. During the experiment the winding number could be set to within 2 parts in \(10^4\). For the optimum values of parameters the quasiperiodic route with golden mean winding number was found to be located in between the boundary of Arnold tongues of 8/13 and 13/21 frequency locking. The measured power spectrum taken at the point of quasiperiodic transition to chaos showed the predicted self-similar band structure between the powers of golden mean value. For the high frequency region the magnitudes and positions of peaks in the power spectrum agreed excellently with those of the circlemap but the agreement was poorer for the low frequency region. The source of the disagreement was later clarified as non deterministic system noise which continuously contaminated the measured current signal.

The \(f(\alpha)\) spectrum has been recently introduced theoretically to exploit the interwoven multifractal structure of singularities of the attractor. The probability distribution of the points on the attractor can be characterized by the scaling index \(\alpha\) and its distribution on the attractor has a fractal dimension \(f(\alpha)\). The \(f(\alpha)\) spectrum of the transition from quasiperiodicity to chaos of the circlemap is uniquely determined. It was predicted that the spectrum for any amplitude below the critical amplitude \((K = 1)\) collapses to a point but the spectrum at the critical amplitude has a smooth parabolic curve. A large number of digitized data of continuous time series of the experimental quasiperiodic signal from the plasma with the critical amplitude and the golden mean winding number were taken to reconstruct the attractor. By using a program of finding a high-probability path along the
reconstructed two-dimensional attractor, the attractor was replaced by the path and the probability distribution was calculated along the path. This method turned out to be better than the recurrence time method by reducing the wigglings or steps in the calculation of the scaling exponent $\tau(q)$. This is due to lack of equivalence of probability to the inverse of the recurrence time. We found that the measured scaling exponent $\tau(q)$ which is a slope of $\log \Gamma(q,l)$ vs. $\log l$ curve had two or three different values depending on the value of $q$ and only one of them produced the $f(\alpha)$ spectrum predicted by the circlemap. As the value of $q$ is decreasing, the range of distance of having right scaling, i.e., theoretical exponents becomes smaller. This is related to the poor agreement and large error estimation of the measured $f(\alpha)$ spectrum with the universal one. The measured $f(\alpha)$ spectrum except the consistent small deviations for negative $q$ from the analysis method explained above are in good agreement with the circlemap.

At decreased the values of $q$, the $\tau(q)$ and $f(\alpha)$ are determined by low probability areas on the attractor which are more sensitive to noise. The noise effect on the power spectrum and the $f(\alpha)$ spectrum obtained from the experimental data was studied from circlemap data with added random fluctuations. The noise was found to smooth the fine structure of the probability distribution and give uniform density at small scales. For the power spectrum the background noise increased to kill weak peaks so the low frequency structure are distorted severely from the noise-free spectrum. The $\tau(q)$ plots showed the noise-dependent $q - 1$ value for the smaller length region and the right scaling value underlying the circlemap for the
larger length region. In comparison with positive $q$ the negative $q$ has shorter scaling region of $\tau(q)$ and is very sensitive to noise as shown in the analysis of the experimental data. This leads to the conclusion that the $f(\alpha)$ spectrum is an important dynamical quantity of characterizing the attractor in comparison with other dynamical quantities, but it is still very difficult to measure because of the sensitivity to extrinsic non deterministic noise found in many real physical systems.
APPENDIX A. MODELS FOR HELICAL WAVE INSTABILITY$^{1,2,3}$

The following differential equations of motion were proposed to describe the motion of electrons and holes in the semiconductor crystals:

\[ J_e = ne \mu_e E + eD_e \nabla n - \mu_e J_e \times B \]  
\[ J_h = pe \mu_h E - eD_h \nabla p + \mu_h J_h \times B \]  
\[ \frac{\partial n}{\partial t} = \frac{1}{e} (\nabla \cdot J_e) + \gamma \]  
\[ \frac{\partial p}{\partial t} = - \frac{1}{e} (\nabla \cdot J_h) + \gamma \]  
\[ \nabla \cdot E = - \frac{e}{\varepsilon} (n - p) \]

where $J_{e,h}$ is the local electron (hole) current density, $n$ is the local electron density, $p$ is the local hole density, $E$ is the local electric field, $B$ is the applied dc magnetic field, $e$ is the magnitude of the electronic charge, $\mu_{e,h}$ is the electron (hole) mobility, $D_{e,h}$ is the electron (hole) diffusion constant, $\varepsilon$ is the dielectric constant of the sample, and $\gamma$ is the rate of net charge carrier generation. The boundary condition of the equations of motion for an infinitely long cylindrical sample along z-axis at the cylindrical surfaces is:

\[ J_e = J_h = esn \]  

where $s$ is the surface recombination rate. When the perturbation of charge density of electron-hole plasma occurs, the carrier densities and the electric field can be expanded about the equilibrium values:

\[ n = n_0 + n_1(t); \quad p = p_0 + p_1(t) \]  
\[ E = E_0 - \nabla \psi_1(t) \]

It has been shown that the first order terms lead to a helical density wave,
\[ n_1 = p_1 = N_1(r) \exp (i \omega t - ikz - im \phi) \quad (A1.4a) \]
\[ \psi_1 = \psi_1(r) \exp (i \omega t - ikz - im \phi) \quad (A1.4b) \]

by substituting Eq. A1.3 into Eq. A1.1. Above the threshold values of the applied electric and magnetic field, the helical density wave is unstable and spontaneous oscillations occur.

Nonlinear behavior of the model can be considered a superposition of different modes of the helical wave in which the time dependence is not assumed to be periodic:\(^3\):

\[ n_1 = p_1 = \sum_{k,m} C_{km}(t) N_{km}(r) \exp (-ikz - im \phi) + c.c. \quad (A1.5a) \]
\[ \psi_1 = \sum_{k,m} C_{km}(t) \psi_{km}(r) \exp (-ikz - im \phi) + c.c. \quad (A1.5b) \]

where c.c. means complex conjugates. This equations result in a differential equation of the form

\[ \frac{\partial C_{km}}{\partial t} = M_{km} C_{km} + \sum_{k=k_1+k_2} M'_{k_1k_2m_1m_2} C_{k_1m_1} C_{k_2m_2} \quad (A1.6) \]
\[ + \sum_{k=k_1-k_2} M'_{k_1k_2m_1m_2} C_{k_1m_1} C_{k_2m_2} \]

where \( M \) and \( M' \) are independent of time, and only \( M \) is complex. This equation describes a wave-wave interaction in which waves with different wave vector \( k \) and rotation number \( m \) can couple nonlinearly. Wersinger et al.\(^4\) studied numerically a special case of Eq. A1.6, the evolution of an undamped wave coupled to two damped waves, and they found that the system undergoes a period doubling transition to chaos.
REFERENCES. APPENDIX A


APPENDIX B. COMPUTER PROGRAMS

Program 1. Ridge

This FORTRAN program finds a high-probability path of a two-dimensional reconstructed attractor. The attractor is reconstructed from the experimental data stored in a computer. The program calculates and stores the probabilities on the attractor for the calculation of a $f(\alpha)$ spectrum.

c
dimension idata(98000),n(100),x(5000),y(5000),m(5000)
character filename*20

c
reading data

c
print*, 'filename of a time series of data ?'
read*, filename
open (unit=2, file=filename, status='old', access='sequential', form='formatted')
rewind 2
print*, 'How many data ?'
read*, ndata
do 10 i=1,ndata+1
  read(2,*) idata(i)
do 10 i=1,ndata+1
  read(2,*) idata(i)
if(idata(i).gt.max) max=idata(i)
if(idata(i).lt.min) min=idata(i)
10 continue
close(unit=2)
c
open graphic window

c
27 call initgraph('window')
call window(0.,0.,1.,1.)
call clear()
call scale(float(min)-100.,float(max)+100.,float(min)-100.,
  float(max)+100.)
call border()
c
find a ridge line

c
print*, 'radius of circle ?'
read*, orad
print*, 'starting point ?'
read*, ist
do 20 i=1,500
  if(i.ne.ist) go to 21
call boxat(float(idata(i)),float(idata(i+1)))
go to 20
21 call dotat(float(idata(i)),float(idata(i+1)))
20 continue
call move(float(min),0.)
call pendown()
call move(float(min)+float(2*nrad),0.)
call penup()
print*, 'all right (0-yes, 1-no) ?'
read*, nresp
if(nresp.eq.1) go to 27

c define no of different directions
c
pi=2.*asin(1.)
ndir=24
rotat=2.*pi/float(ndir)
c
print*, 'filename of ridge data ?'
read*,filename
open(unit=3,file=filename,status='new',form='formatted')
write(3,51) nrad,ndata
51 format(i6,i8)
x(I)=float(idata(ist))
y(I)=float(idata(ist+1))
x0=x(1)
y0=y(1)
phase=0.
do 22 i=1,ndata
    dst=(x0-float(idata(i)))**2+(y0-float(idata(i+1)))**2
    dst=sqrt(dst)
    if(dst.le.float(nrad)*0.99) then
        m(1)=m(1)+1
    endif
22 continue
nridge=1
print*, 'initial position:', x0,y0,m(1)
write(3,52)nridge,x(I),y(I),m(1)
call move(x0,y0)
call pendown()
40 max=0
if(nridge.eq.1) then
    nin=2
    nfn=ndir/2
else
    nin=-1*(ndir/4)
    nfn=ndir/4
endif
i=nin,nfn
23 do 30 jj=2,nridge
    x1=x0+float(2*nrad)*cos(rotat*(i-1)+phase)
    y1=y0+float(2*nrad)*sin(rotat*(i-1)+phase)
    if(nridge.le.2) go to 39
    do 34 jj=2,nridge
        ddst=(x(jj)-x1)**2+(y(jj)-y1)**2
        ddst=sqrt(ddst)
        if(ddst.lt.float(nrad)*0.99) then
            m(1)=m(1)+1
        endif
34 continue
30 continue
print*, 'initial position:', x0,y0,m(1)
write(3,52)nridge,x(I),y(I),m(1)
call move(x0,y0)
call pendown()
print*, 'overlapping'
go to 30
endif

34 continue
39 n(i)=0
do 32 j=1,ndata
dst=(x1-float(idata(j)))**2+(y1-float(idata(j+1)))**2
dst=sqrt(dst)
if(dst.le.float(nrad)) n(i)=n(i)+1
32 continue
if(n(i).gt.max) then
max=n(i)
temx0=x1
temy0=y1
imax=i
endif
30 continue
x0=temx0
y0=temy0
phase=phase+rotat*(imax-1)
if(abs(phase).gt.2.*pi) then
phase=phase-2.*pi*int(phase/(2.*pi))
endif
nridge=nridge+1
x(nridge)=x0
y(nridge)=y0
m(nridge)=max
print*,nridge,x0,y0,max.phase*180./pi
write(3,52) nridge,x(nridge),y(nridge),m(nridge)
52 format(i6,f12.3,f12.3,i6)
call move(x0,y0)
if((x0-x(1))**2+(y0-y(1)))**2.lt.float(nrad)**2) then
print*, 'ridge-finding finished'
go to 100
else
go to 40
endif
100 close(unit=3)
c
c draw density distribution along the ridge
c
call window(-0.2,0.,1.2,1.)
call clearO
maxy=0
do 62 i=1,nridge
if(m(i).gt.maxy) maxy=m(i)
62 continue
ymax=float(maxy)*4./3.
print*, 'scale : x = 0 *.nridge,' y = 0 *,ymax
call scale(0.,float(nridge),0.,ymax)
call borderO
xtic=float(nridge)/10.
ytic=ymax/10.
call axes(0.,0.,xtic,ytic)
do 60 i=1,nridge
   call move(float(i),float(m(i)))
   print*,nridge,x0,y0,max,phase*180./pi
   write(3,52) nridge,x(nridge),y(nridge),m(nridge)
52   format(i6,f12.3,f12.3,i6)
   call move(x0,y0)
   if((x0-x(1))**2+(y0-y(1))**2<lt.float(nrad)**2) then
      print*,"ridge-finding finished"
      go to 100
   else
      go to 40
   endif
100  close(unit=3)
c
  draw density distribution along the ridge
  call window(-0.2,0.,1.2,1.)
  call clear()
  maxy=0
  do 62 i=1,nridge
      if(m(i).gt.maxy) maxy=m(i)
62  continue
  ymax=float(maxy)*4./3.
  print*,"scale : X=",nridge," Y=",ymax
  call scale(0.,float(nridge),0.,ymax)
  call border()
  xtic=float(nridge)/10.
  ytic=ymax/10.
  call axes(0.,0.,xtic,ytic)
  do 60 i=1,nridge
      call move(float(i),float(m(i)))
      if(i.eq.1) call pendown()
60  continue
  call penup()
  print*,"0 to quit"
  read*,number
  call exitgraph()
call exit
end
Program 2. Gamma

This program calculates partition functions $\Gamma(q,l)$ for a given $q$ as a function of $l$ by using a probability distribution on the attractor obtained from the program 1 (ridge) in Appendix.

c
dimension x(1000),y(1000),m(1000),idata(96000)
dimension prob(5000)
double precision partial,prob,gamma,sumgam,q
character filename1*20, filename2*20
c
reading data
c
print*, 'filename of a probability distribution ?'
read*, filename1
open(unit=2,file=filename1,status='old',form='formatted')
read(2,1) minl,ndata
1 format(i6,i8)
npoint=0
do 3 i=1,2000
      read(2,5,end=9) x(i),y(i),m(i)
5 format(6x,2f12.3,i6)
      no=no+1
      npoint=npoint+m(i)
3 continue
9 close(unit=2)
c
print*, 'filename of a time series of data ?'
read*, filename2
open(unit=2,file=filename2,status='old',form='formatted')
do 10 i=1,5000
   read(2,11) idata(i)
11 format(i8)
10 continue
close(unit=2)
c
calculate partition functions
c
open(unit=3,file='gamma.dat',status='unknown',form='formatted')
c
size=2.*float(minl)
do 20 i=1,7
   radius=float(2*i-1)*size
   write(3,21) radius
21 format(1x,e15.7)
do 22 l=1,5000
   prob(l)=0.
22 continue
do 30 j=1,5000
mtotal=0

```
do 31 k=1,no
  d=(float(idata(j))-x(k))**2+(float(idata(j+1))-y(k))**2
  d=sqrt(d)
  if(d.lt.float(minl)) then
    kpos=k
    go to 33
  endif
  continue
 31 continue
```

```
do 32 l=1,i
  if(l.eq.1) then
    m1=m(kpos)
    m2=0
    m3=0
  else
    m1=0
  endif
  if(kpos+l.gt.no) then
    m2=m(kpos+l-no)
  else
    m2=m(kpos+l)
  endif
  if(kpos-l.lt.1) then
    m3=m(na+kpos-l)
  else
    m3=m(kpos-l)
  endif
  mtotal=mtotal+m1+m2+m3
  partial=float(mtotal)/float(npoint)
  prob(j)=prob(j)+partial
  30 continue
```

```
c set q and calculate Gamma(q,nradius)
c```

```
do 40 n=-30,30
  q=float(n)*1.d0
  sumgam=0.
  do 50 nn=1,5000
    gamma=prob(nn)**(q-1.)
    sumgam=sumgam+gamma
  50 continue
  sumgam=sumgam/5.d3
  write(3,52) q,sumgam
  52 format(1x,dI5.7,5x,dI5.7)
```

```
do 20 continue
  close(unit=3)
  stop
end
```
Part II

AC Magnetic Susceptibility Measurements of Granular Superconductor $\text{YBa}_2\text{Cu}_3\text{O}_7$
The two components of the complex ac magnetic susceptibility of sintered pellet and powdered samples of the ceramic high \( T_c \) superconductor \( \text{YBa}_2\text{Cu}_3\text{O}_7 \) were measured as a function of magnitude of dc and ac magnetic fields, frequency of ac magnetic field, and temperature. The measured magnetic susceptibilities show strong dependence on applied magnetic fields and temperature for cylindrical pellet samples. They are also strongly dependent on sample preparations: for the powdered samples, the dependences are quite different from those for sintered pellet samples. The magnetic susceptibilities show hysteresis at large dc magnetic fields and low temperatures; the hysteresis is larger for small ac magnetic fields.

A modified critical state model is developed to explain the measured data for the ac magnetic susceptibility. In the model, the susceptibility is calculated by assuming contributions from two components, which we refer to as arising from two types of local regions in the sample: (i) superconducting grains and (ii) intergranular weak links, including Josephson junction. The intergranular component was found to be responsible for the strong dependence on small ac and dc magnetic fields at relatively low temperature. A new phenomenological form of critical current density as a function of magnetic field for the intergranular region is proposed to explain the experimental results. The hysteresis effects are explained by using a modified critical state model with the assumption of irreversible shielding currents in addition to reversible currents. This irreversible critical current is believed to arise from flux pinning of magnetic vortices in intergranular regions.
As the temperature is increased the vortices begin to penetrate into grains, so the temperature dependence of the ac magnetic susceptibility shows an interplay of both regions. This modified critical state model with two components, intergranular regions and superconducting grain regions, is compared in detail with the experimental results. Excellent agreement is obtained for much of the data, which is the principal result for this part of the thesis.
CHAPTER 1. INTRODUCTION

Since the first discovery of superconductivity in mercury in 1911 there has been much progress in studies of basic properties and applications of various superconducting materials. From the point of view of experiments the number of superconductors has increased tremendously and their basic properties such as critical temperatures, and critical currents have been improved steadily. From the theoretical viewpoint the nature of superconductivity has been the interest of many physicists for more than three decades. Bardeen et al\textsuperscript{1} could finally explain the mystery of superconductivity by assuming Cooper pairs, which are pairs of electrons coupled by phonons. In spite of this progress, superconductors have not been used extensively in industry mainly because of the required extremely low critical temperatures (4 K for Hg, 23 K for a Nb compound). There has been much effort to increase the superconducting critical temperature, both experimentally – in synthesizing and investigating novel materials, and theoretically – in the search for novel superconducting mechanisms. In 1986 Bednorz and Müller\textsuperscript{2} reported a possible high-$T_C$ superconductivity near 40 K in the Ba-La-Cu-O System and they opened a new door to high-$T_C$ superconductivity. More oxide superconductors were discovered by various groups and they have record-high critical temperatures such as 90 K for $\text{YBa}_2\text{Cu}_3\text{O}_7$, 110 K for $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$, and 120 K for $\text{Tl}_2\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$. These high-$T_C$ superconductors have some common generic properties of low temperature, conventional superconductors\textsuperscript{6}, but they also have novel properties.
In this chapter the basic properties of high-$T_c$ superconductors will be discussed briefly in Section 1.1. In Section 1.2 the magnetic susceptibility of magnetic materials including superconductors will be introduced. In Chapter 2, two present models which seem to be related to the observed magnetic properties of high $T_c$ superconductors will be described. In Chapter 3 are given experimental details of the measurements of the two components of the complex ac magnetic susceptibility of $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO) samples as a function of ac and dc magnetic fields, temperature, and frequency of ac magnetic fields. In Chapter 4, the measured susceptibility data are presented and are discussed in terms of numerical results of a modified critical state model. In Chapter 5, we conclude something about the underlying mechanisms of the magnetic properties of the granular superconductor YBCO from the experimental results and numerical calculations.

1.1. High-$T_c$ Superconductors

The new high-$T_c$ superconductors\textsuperscript{7} are a class of superconducting copper oxides. In Table 1.1 there are given some of commonly studied high-$T_c$ Superconducting materials. These compounds are usually made through cycles of calcinizations by mixing elements with the desired atomic ratios and oxygen annealings. The superconducting properties are quite sensitive to the method of preparation and annealing. The oxygen content in the samples is critical and difficult to control. These oxide superconductors have a layered perovskite-type structure, or a $\text{K}_2\text{NiF}_4$ structure. Except for the $(\text{La-Sr})_2\text{CuO}_4$ system, they have a distorted
oxygen-defect perovskite structure shown in Fig. 1.1 for superconducting orthorhombic YBCO.

<table>
<thead>
<tr>
<th>Compounds</th>
<th>Transition Temperature (K)</th>
<th>Number of Cu-O Layers</th>
</tr>
</thead>
<tbody>
<tr>
<td>(La–Sr)$_2$CuO$_4$</td>
<td>40</td>
<td>1</td>
</tr>
<tr>
<td>YBa$_2$Cu$_3$O$_7$</td>
<td>95</td>
<td>2</td>
</tr>
<tr>
<td>YBa$_2$Cu$_4$O$_8$</td>
<td>81</td>
<td>2</td>
</tr>
<tr>
<td>Bi$_2$Sr$_2$CuO$_6$</td>
<td>12</td>
<td>1</td>
</tr>
<tr>
<td>Bi$_2$Sr$_2$CaCu$_2$O$_8$</td>
<td>90</td>
<td>2</td>
</tr>
<tr>
<td>Bi$_2$Sr$_2$Ca$_2$Cu$<em>3$O$</em>{10}$</td>
<td>110</td>
<td>3</td>
</tr>
<tr>
<td>Tl$_2$Ba$_2$CuO$_6$</td>
<td>90</td>
<td>1</td>
</tr>
<tr>
<td>Tl$_2$Ba$_2$CaCu$_2$O$_8$</td>
<td>110</td>
<td>2</td>
</tr>
<tr>
<td>Tl$_2$Ba$_2$Ca$_2$Cu$<em>3$O$</em>{10}$</td>
<td>122</td>
<td>3</td>
</tr>
</tbody>
</table>

Table 1.1. High-$T_C$ superconductors (from Ref. 9)

Neutron and X-ray diffraction studies indicate the simultaneous presence of both two and one-dimensional features in the Cu–O structures as shown in Fig. 1.1. It would appear that two-dimensional Cu–O layers in a-b planes play a crucial role in the origin of the superconductivity of these compounds and the linear Cu–O chains along c-axis are required for the superconductivity of LaSrCuO and YBCO compounds through coupling to the Cu–O layers, but not necessary for other compounds which have no linear chains. In the case of YBCO compounds
the replacement of Y by rare earths usually results in materials with similar high-
$T_C$ which indicates no correlation of $T_c$ with the magnetism of the rare earth ions.
The replacement of Cu by Ni produces a dramatic reduction of $T_c$ which may be
an important clue to the underlying electronic mechanism. With the availability of
single crystals of these compounds many workers have reported strong anisotropy
effects in physical properties such as electrical resistivity, magnetization, or criti-
cal currents. It was inferred that the interplanar coherence length is much shorter
than the in-plane coherence length.

All of the materials in the table exhibit the classical features of low tempera-
ture superconductors: zero electrical resistance and the Meissner effect below
superconducting transition temperature $T_c$. The electrical resistance versus tem-
perature curve for a YBCO sample is shown in Fig. 1.2.10 However the high-$T_C$
superconductors have unique features in comparisons to old superconductors. For
example, the critical current density is extremely low ($\approx 10^3$ A/cm$^2$) for bulk
polycrystalline ceramic materials and two to three orders of magnitude higher for
thin films. The charge carrier density ($\approx 10^{21}$/cm$^3$), superconducting energy gap
($\approx 20$ meV), and coherence length ($\approx 1.2$ nm at $T \to 0$) determined from the
experiments are very small. Another important property of these compounds is
the lack of (or very small) isotope effect in case of the replacement of O$^{16}$ by O$^{18}$.
Sintered samples of those compounds consist of a polycrystalline array of grains
by the nature of the ceramic material. Even if they are single crystals, there are
stacking defaults and twin boundaries. After the discovery of high $T_C$
superconductivity the effects of grains on magnetic and electrical properties were widely discussed in the literature. The difference between field-cooled (FC) and zero-field-cooled (ZFC) magnetic susceptibility and the logarithmic decay of remnant magnetization in time, which are similar to predictions for a spin glass, were reported and related with the granular property. The grain boundaries induce intergranular Josephson or weak-link junctions and these affect dramatically the diamagnetic properties at very low magnetic fields. These junctions couple with the superconducting grains and trap magnetic fluxons as the magnetic field increases. There is a de Almeida-Thouless line separating metastable from stable regions which indicates spin-glass-like behavior.

Many theoretical speculations on the mechanism of high $T_C$ superconductivity, including the Bardeen-Cooper-Schrieffer (BCS) theory of low $T_C$ superconductors, have been attempted to explain experimental results. In many instances the data reflect extrinsic properties due to poor quality of samples such as inhomogeneities, grain boundaries, or defects, rather than intrinsic superconducting properties. Therefore more systematic and detailed experiments are necessary to help establish a theoretical model of the high $T_C$ superconductors.

1.2. Magnetic Susceptibility

The macroscopic magnetic properties of matter can be described in terms of three magnetic field vectors: the magnetic induction $B$, the magnetization $M$, and the magnetic field intensity $H$. In the MKS system of units, the relation between
these vectors is

$$B = \mu_0 (H + M) = \mu H$$

(1.1)

where $\mu_0 = 4\pi \times 10^{-7}$ H/m is the permeability of free space and $\mu$ is the permeability of the magnetic material. The magnitude of the relative permeability is defined as

$$\kappa_m = \mu / \mu_0$$

(1.2)

and the magnetic susceptibility $\chi$ is defined from $\kappa_m$ as

$$\kappa_m = 1 + \chi .$$

(1.3)

The magnetization is then related with $H$ by

$$M = \chi H .$$

(1.4)

The magnetic susceptibility is used as a criterion to distinguish paramagnetic, diamagnetic, and ferromagnetic materials. For paramagnetic materials $\chi$ will normally be slightly larger than zero; for diamagnetic materials smaller than zero; and for ferromagnetic materials much greater than zero. When the vectors $B$, $M$, and $H$ are parallel, the permeability or susceptibility is a constant. In anisotropic materials $B$ and $H$ are not parallel, so the susceptibility is given as a tensor.

If a dc magnetic field is applied to a magnetic material (say, in the form of a long cylinder), the applied field induces a magnetization and the $dc$ magnetic susceptibility $\chi_0$ is defined as

$$\chi_0 = \frac{M}{H}$$

(1.5)

where $M$ is the magnetization measured from experiments. Here only equilibrium states were considered and the time required to establish the equilibrium is
assumed to be negligibly small when the magnetic field is changed. However when an ac magnetic field of sufficiently high frequency is applied, the magnetization of the material is unable to follow the changes of the applied field, but instead lags in phase. It is convenient to describe the phenomena in a manner analogous to that used for the static case. If the applied field has the form

\[ H(t) = H_0 + H_1 \cos \omega t, \]  

(1.6)

the magnetization may be represented by

\[ M(t) = M_0 + M_1 \cos (\omega t - \phi) \]  

(1.7)

where \( H_0 \) is a dc magnetic field, \( M_0 \) is the equilibrium value of the magnetization in this dc field, and \( \phi \) is the phase angle by which the magnetization lags the field; \( \phi \) is determined by the ac losses of the material. In analogy to Eq. 1.5, we define

\[ \chi' = \frac{M_1 \cos \phi}{H_1}, \]  

(1.8a)

and

\[ \chi'' = \frac{M_1 \sin \phi}{H_1} \]  

(1.8b)

so that \( \chi'' / \chi' = \tan \phi \). Then Eq. 1.7 becomes

\[ M(t) = \chi_0 H + \chi' H_1 \cos \omega t + \chi'' H_1 \sin \omega t. \]  

(1.9)

The complex notation can be used, so that

\[ H(t) = H_0 + H_1 e^{i \omega t} \]  

(1.10a)

and

\[ M(t) = M_0 + \chi H_1 e^{i \omega t} \]  

(1.10b)

where
\[ \chi = \chi' - i \chi'' = |\chi| e^{-i\phi}. \] (1.11)

Both \(\chi'\) and \(\chi''\) are dependent on the frequency \(\omega\) as well as the magnitude of the dc and ac fields. Particularly, \(\chi''\) is related to the energy absorbed by the material from the ac field. Since the work done on the specimen is given by \(dW = -H\,dM\), the energy \(A\) absorbed per second per unit volume is given by

\[ A = \frac{\omega}{2\pi} \int_{cycle} H\,dM = \frac{\omega}{2} \chi'' H^2. \] (1.12)

At very low frequency \(\chi' \to \chi_0\) and \(\chi'' \to 0\), that is, \(M\) and \(H\) are in phase.

Superconductors have a diamagnetic property below critical magnetic fields. When the magnetic field is applied above \(T_C\) and then the superconductor is cooled below \(T_C\), the magnetic field inside the specimen is expelled so that the magnetic induction \(B\) is zero. This flux expulsion is called the Meissner effect. When the specimen is cooled below \(T_C\) without applying a magnetic field and then the magnetic field is applied, the superconductor generates a shielding current to exclude the magnetic flux. That is called the dc magnetic shielding. For ordinary (type I) superconductors the magnetic susceptibility measured with field-cooled (FC) and zero-field-cooled (ZFC) cases are same. Type II superconductors and high \(T_C\) superconductors in particular have been reported to have different values of the magnetic susceptibility\(^{11}\) for FC and ZFC cases as shown in Fig. 1.3. From this it was argued that high \(T_C\) superconductors form superconducting glasses, proposed by Ebner and Stroud\(^{12}\) in a calculation of the diamagnetic susceptibility of superconducting clusters. Some important consequences they predicted were: a large difference between dc and ac diamagnetic susceptibilities at low
temperatures; and a difference between FC and ZFC susceptibilities. Further evidence for glassy superconductivity comes from the existence of intergranular Josephson junctions or weak links junctions. The high $T_C$ superconductors have a extremely short coherence length (≈ 1.2 nm) of the order of the size of the unit cell, and this short coherence length lowers the pair potential at surfaces and the interfaces. This leads to the appearance of internal Josephson junctions responsible for the glassy behavior. Particularly, it was suggested that twin boundaries form intergrain Josephson junctions and these junctions form loops that divide superconducting grains into weakly coupled superconducting domains.

The ac loss mechanism of high temperature superconductors, which have two critical magnetic fields and allow magnetic field penetration without destroying the superconductivity, has been studied extensively mainly because of their potential for industrial applications. There are basically two loss mechanisms: hysteretic loss and flux-flow loss. When an ac magnetic field is applied, fluxons sweep in and out each cycle from the specimen and a layer of bulk current is induced analogous to the skin effect in a normal conductor. This causes the hysteretic loss, owing to the pinning of the fluxons by impurities or defects. The flux-flow loss appears due to flux-flow resistivity. In an ideal type II superconductor an electric field appears by a flow of the flux lines when the flux lines are no longer pinned. The vortex structure is approximated by an inner core that is normal, surrounded by a superconducting region. The ac loss then occurs as Joule heating in this essentially normal core region.
REFERENCES. CHAPTER 1


7. There are several review books about high $T_c$ superconductors even though it is still a fast expanding area. For example see, C. P. Poole, Jr., Copper Oxide Superconductors (Wiley, New York, 1988) and references therein.


FIGURE CAPTIONS

Fig. 1.1. Structures of the orthorhombic (superconducting) and tetragonal (normal) \( \text{YBa}_2\text{Cu}_3\text{O}_7 \) unit cells. This figure is adapted from Ref. 8. Oxygens are randomly dispersed over the basal plane sites in the tetragonal structure. Thermal vibration ellipsoids are shown for the atoms.

Fig. 1.2. Resistivity of a single-phase \( \text{YBa}_2\text{Cu}_3\text{O}_7 \) sample as a function of temperature. This is from Ref. 10.

Fig. 1.3. (a) Magnetization versus temperature for a single crystal of \( \text{YBa}_2\text{Cu}_3\text{O}_7 \). The open circles represent dc screening and crosses represent Meissner effect. This is from Ref. 15. (b) Magnetization hysteresis loops at 10 K for a single crystal of \( \text{YBa}_2\text{Cu}_3\text{O}_7 \). This is from Ref. 15.
Figure 1.1

Figure 1.2
Figure 1.3
CHAPTER 2. THEORY

An important feature of the copper oxide superconductors is that in the sintered form they are polycrystalline ceramics. Even the single crystal structure of the YBa$_2$Cu$_3$O$_7$ class of materials is in fact frequently interrupted by twin boundaries separating microcrystals, and their crystal axes are rotated to interchange the a and b axes of the superconducting orthorhombic structure$^1$. For polycrystalline ceramic pellet materials (used in this thesis) inhomogeneous structures are commonly observed and the length scale of granularity is of order of 1 μm. It is assumed that the sample consists of grains of relatively good crystalline material which are separated by nonstoichiometric materials which may be superconducting with lower $T_C$ or even normal$^1$, or even insulating. Depending on the nature of separating materials the intergranular coupling can be modeled as Josephson junctions or superconducting-normal-superconducting (SNS) type weak links$^2$. This intergranular coupling seemed to characterize the rather unique magnetic behavior of the high $T_C$ superconductors.

In this chapter two theoretical models are described to show how the intergranular coupling or the granularity can affect the magnetic property of these oxide superconductors significantly even in a small magnetic field.

2.1. Josephson Junctions Model

The theory of the Josephson junction is well established and can be found in textbooks on superconductivity$^2$. In this section the property of the Josephson
junction in the presence of a magnetic field will be described briefly using Gaussian units. Fig. 2.1 shows the geometry of a simple Josephson tunneling junction normal to the x axis. When the thickness of the interfacial material is thin enough to give rise to tunneling of superconducting electron pairs, the supercurrent density flows in one direction and is given by

\[ J(T) = J_0(T) \sin \gamma \]  

when there is no applied magnetic field. \( J_0(T) \) is the tunnel current density at a given temperature \( T \). It is the maximum current density and is called the critical current and has, according to the Ambegaokar-Baratoff theory, the value

\[ J_0(T) = \frac{\pi \Delta(T)}{2 e R_n} \tanh \frac{\Delta(T)}{2 kT} \]  

where \( R_n \) is the tunneling resistance per unit area of the junction at normal state and \( \Delta(T) \) is the superconducting energy gap at \( T \). The quantity \( \gamma \) is the phase difference between superconductors \( S_1 \) and \( S_2 \) on opposite sides of the junction and is given by

\[ \gamma = (\phi_2 - \phi_1) - \frac{2 \pi}{\Phi_0} \int_{1}^{2} A_x \, dx \]  

where \( \phi \) is the phase of the superconducting electrons; \( A_x \) is the component of a vector magnetic potential along the current direction (x axis in the figure) and \( \Phi_0 = \frac{hc}{2e} = 2 \times 10^{-7} \text{ Gcm}^2 \), the flux quantum.

Consider that we apply a magnetic field \( H \) parallel to the z axis to the junction in the figure. Suppose that the magnetic field penetrates the interfacial barrier by the distance \( \lambda_f \), then
\( \gamma(y) - \gamma(0) = \frac{2 \pi \Phi(y)}{\Phi_0} = \frac{2 \pi H (2\lambda_f + d) y}{\Phi_0} \) \hspace{1cm} (2.4)

The superconducting current through the junction is modulated along the \( y \) axis so the total current \( I \) will be, on integration,

\[ I = YZ J_0 \sin \gamma(0) \frac{\sin (\pi \Phi / \Phi_0)}{\pi \Phi / \Phi_0} \hspace{1cm} (2.5) \]

where \( \Phi = H Y (2\lambda_f + d) \). The maximum supercurrent through the junction as a function of applied field is

\[ I_{\text{max}} = YZ J_0 \left[ \frac{\sin (\pi \Phi / \Phi_0)}{\pi \Phi / \Phi_0} \right] \hspace{1cm} (2.6) \]

since the sine function cannot exceed unity. Fig. 2.2 shows a diffraction-like pattern of the maximum supercurrent through a Josephson junction in a dc magnetic field \( H \).

If the tunneling current is not negligible, as is assumed above, the tunneling current tends to screen the magnetic field from the junction region. Then we have

\[ \frac{d^2 \gamma}{dy^2} = \frac{1}{\lambda_f^2} \sin \gamma \hspace{1cm} (2.7a) \]

where

\[ \lambda_f = \left[ \frac{c \Phi_0}{8 \pi^2 J_0 (2\lambda_s + d)} \right]^{1/2} \hspace{1cm} (2.7b) \]

is called an effective Josephson junction penetration depth.

\[ \lambda_s = \left( \frac{mc^2}{4\pi n_s e^2} \right)^{1/2} \hspace{1cm} (2.7c) \]

is the bulk or London penetration depth of the superconductors \( S_1 \) and \( S_2 \) where \( n_s \) is the density of superconducting electrons and \( e \) is the electronic charge.
Early measurements of a point contact junction of YBCO did indeed show behavior qualitatively like Fig. 2.2, but with poorly defined minima. Other data on simple contacts on bulk YBCO material also showed crude data, somewhat like Fig. 2.2.

Other measurements of critical current of high $T_C$ superconductors did not show the diffraction-like pattern shown in Fig. 2.2. This was attributed to a distribution of Josephson junction areas. Different areas produces dips of $I_{\text{max}}$ at different magnetic fields so that the distribution of the junction areas smooths the pattern. If the distribution is broad, one cannot see the dips. Instead $I_{\text{max}}$ is monotonically decreasing with increasing magnetic field, which is typically observed in the measured critical current as a function of magnetic fields for the slabs of sintered YBCO. The extreme sensitivity of magnetic and electronic transport properties observed for high $T_C$ superconductors at relatively low magnetic fields is believed to be related to that of the critical current of Josephson junctions with the distribution of the junction areas. Some experiments have been explained successfully by applying this model. However, if the applied field is larger or the temperature is higher, the penetration depth of junctions $\lambda_J$ is larger than the size of the junctions so that the above model breaks down, and the superconducting grains themselves play a dominant role in the magnetic behavior.

2.2. Critical State Model

The critical state model was an ad hoc proposal by Bean to explain the hys-
teresis of magnetization measurements of hard superconductors. In these materials the pinning force on the magnetic flux (fluxons) is so strong that it prevents any substantial vortex motion and associated electrical resistance. As briefly mentioned in Section 1.2, for a type II superconductor the flux vortices thread through it when the applied field is larger than the lower critical field $H_1$. When there is no pinning force, the Lorentz force density given by

$$F = J \times \frac{B}{c}$$ (2.8)

acts between the current in the superconductor and the flux vortices and the flux lines tend to move transverse to the current. Here $c$ is a speed of light. This is the origin of the energy dissipation due to flux-flow. It was found experimentally that the flux vortices move in bundles. From the practical point of view, this flux-flow loss can have a disastrous sudden onset in high field superconducting magnets. Fortunately some low temperature type II superconductors have been developed with some properties which prevent the Lorentz force from moving the flux vortices. Such a mechanism is called a pinning force since it pins the vortices to fixed locations in the material. Pinning usually results from any spatial inhomogeneity of the material, such as grain boundaries, impurities, or voids. If the pinning is sufficiently strong, the vortex motion is negligible and the superconductor material will remain essentially a good superconductor up to very high current and fields, up to $H_{c2} \approx 10^5$ Oe. If the pinning force is smaller than the driving Lorentz force, the vortices move in a steady motion and the superconductor has a flux-flow resistivity similar to that of a normal conductor.
If we consider a cylindrically shaped superconductor shown in Fig. 2.3, the applied magnetic field $H_{\text{ext}}$ along the $z$ axis produces a driving force density $\alpha$ on the vortices which is given by

$$
\alpha = J \times \frac{B}{c} = (\nabla \times H) \times \frac{B}{4\pi}
$$

(2.9)

where $H$ is the local magnetic field inside the superconductor. The Maxwell equation relating the current density to the magnetic field,

$$
\nabla \times H = \frac{4\pi}{c} J
$$

(2.10)

is used in Eq. 2.9. For the geometry of Fig. 2.3, Eq. 2.10 can be written

$$
\frac{dH}{dr} = -\frac{4\pi}{c} J
$$

(2.11)

so there is an outward force on each vortex given by Eq. 2.11. Summed over all vortices, Eq. 2.9 gives a force per unit volume of

$$
\alpha = \frac{B}{4\pi} \frac{dH}{dr}
$$

(2.12)

Let us consider meaning of the current $J$ in the above equations. When we apply a magnetic field $H_{\text{ext}}$ which is larger than $H_{c1}$, a low critical field, flux vortices enter the wall; however this flux is canceled by a magnetic shielding current in the wall. This induced shielding current density is given by Eq. 2.11 and it is large if $H(r)$ drops rapidly in the cylinder. If the force due to the induced shielding current density is larger than the maximum available pinning force $\alpha_c$, the Bean model assumes that the vortices will penetrate further into the specimen, tending to reduce the gradient term. This process will continue until the driving force is smaller than $\alpha_c$ everywhere inside the cylinder. This situation is called
the critical state and the induced current density inside is called a critical current density \( J_c \). Bean first suggested that \( J_c \) is constant and independent of \( H \) but this assumption implies an infinite critical current in zero field.

2.3. Kim-Anderson Version of the Model

This difficulty was avoided by Kim et al. by assuming

\[
J_c (H) \propto \frac{1}{|H| + H_s}
\]  

(2.13)

where \( H \) is a local magnetic field inside the superconductor and \( H_s \) is an empirical parameter which prevents an infinite current density at \( H = 0 \). \( H_s \) is assumed to be a constant.

A simple critical current model with a current of the form of Eq. 2.13 of Kim et al. has been recently used for high \( T_C \) superconductors to explain their nonlinear magnetic response. In the model the superconductors are assumed to consist of superconducting grains and intergranular junctions (see Fig. 2.3). The model will be discussed more fully in the Appendix A. Here we outline the essential ideas, so that the experimental details can be discussed in the next chapter.

When the superposed parallel dc and ac magnetic fields are parallel to each other and to the long axis of the cylindrical sample of radius \( R \), we define the total applied field

\[
H(t) = H_0 + H_1 \cos \omega t
\]

(2.14)

and the field-dependent critical intergranular junction current density \( J_{cJ} \); and the critical grain current density \( J_{cg} \). Here \( H_0 \) and \( H_1 \) are the magnitudes of the
applied dc and ac magnetic field respectively. Here the critical current density is more generalized and is given by these two equations:

\[ \begin{align*}
J_{cJ} &= \alpha_J \frac{c}{|H| + H_{sJ}} \\
J_{cg} &= \alpha_g \frac{c}{|H| + H_{sg}}
\end{align*} \]  

(2.15a) (2.15b)

where \( H_{sJ}, H_{sg} \) are assumed constants and \( \alpha_J, \alpha_g \) are assumed pinning force densities for intergranular junctions and grains respectively. The \( \alpha_J \) and \( \alpha_g \) are assumed to be independent of \( H \) in Kim-Anderson version of the model. The equations of motion of critical state are then given from Eq. 2.13 by the two separate equations, corresponding to the two separate components, intergranular and grains, respectively:

\[ \begin{align*}
\frac{dH_J}{dr} &= \pm \frac{4\pi}{c} J_{cJ} \\
\frac{dH_g}{dr} &= \pm \frac{4\pi}{c} J_{cg}
\end{align*} \]  

(2.16a) (2.16b)

The ± sign in the equation accounts for the out- or inward-motion of vortices with increasing and decreasing applied magnetic field respectively; this follows from Bean’s original assumption that the direction of the initial shielding current is determined by the most recent algebraic sign of the emf induced by the applied field.

The complex ac magnetic susceptibility components are then defined in MKS units as

\[ \chi' = \frac{\omega}{\pi \mu_0 H_1} \left[ \frac{2\pi/\omega}{\int_0^{2\pi/\omega} <B(t)> \cos(\omega t) dt} \right] - 1, \]  

(2.17a)
\[ \chi'' = \frac{\omega}{\pi \mu_0 H_1} \int_0^{2\pi/\omega} <B(t)> \sin(\omega t) dt, \tag{2.17b} \]

\[ <B(t)> = <B_j(t)> + <B_g(t)>, \tag{2.17c} \]

\[ <B_j(t)> = \int [\mu_j H_j(r) d^3r] / \int d^3r \tag{2.17d} \]

\[ <B_g(t)> = \int [\mu_g H_g(r) d^3r] / \int d^3r \tag{2.17e} \]

is the averaged magnetic induction over the sample, for the two components respectively. In the above equations \( \mu_J \) and \( \mu_g \) are the permeability of intergranular and grains region respectively and they will be disussed in detail in Chapter 4.

The critical state equation (Eq. 2.16) can be solved numerically to find \( H(r) \) (see Appendix B); with the solutions of \( H(r) \) the susceptibility from Eq. 2.17 can also be computed numerically. The measured susceptibility is then the summation of the contribution of the two components: intergranular and grain. The details and the extension of the model calculation will be presented in Chapter 4 and in Appendix A for a cylindrical superconducting sample. When the magnetic field is sufficiently large, the field can fully penetrate into the sample and the minimum field which penetrates into the center is called the penetration field \( H^* \). This penetration field is related to \( \alpha \) and \( R \) as follows:

\[ H^* = \left[ 8 \pi \alpha R + H_s^2 \right]^{1/2} - H_s. \tag{2.18} \]

This equation will be applied only to the intergranular region and generalization of Eq. 2.18 for the grains and intergranular regions will be given in Eq. 4.13b in Section 4.4. Eq. 2.18 may be used to replace the assumed model parameters \( \alpha, H_s, \) and \( R \) by \( H^*, H_s, \) and \( R \); this is useful because the cylinder radius \( R \) is known experimentally and \( H^* \) can often be easily obtained from experimental data. As
an example Fig. 2.4 shows a numerical solution of Eq. 2.16 for an infinitely long cylindrical superconductor with a radius \( R = 0.085 \text{ cm} \), \( H^* = 5.39 \text{ Oe} \), and \( H_s = 5.0 \text{ Oe} \) when \( H_0 = 2.0 \text{ Oe} \) and \( H_1 = 3.0 \text{ Oe} \), which values of parameters are likely to explain the magnetic behavior of the intergranular junction. The ac magnetic susceptibility can be calculated numerically by using Eq. 2.17 from the computed time sequence of field patterns inside the material per cycle of the applied ac magnetic field.

The analytical details of local magnetic field distributions for a cylindrical superconducting specimen are discussed in Appendix B with computer programs in Appendix C which calculate ac magnetic susceptibility as functions of \( H_0 \) and \( H_1 \). It should be noted that we have the same values of ac susceptibility for samples with different radius if \( H^* \) and \( H_s \), are the same. In Eq. 2.18 increase of \( R \) decreases \( \alpha \) but the field distribution inside the larger \( R \) sample are exactly the same in units of nondimensional \( r/R \) as that of smaller \( R \). Therefore this produces the same values of ac susceptibility.

One can comment that a complete critical state model of a granular two-component superconductor can determine the complex ac magnetic susceptibility and the dc magnetization, if magnetic field dependence of critical current density is known as well as the relative contributions of the two components. These questions concerning the critical state model will be discussed in more detail in our modified model in Chapter 4, where experimental measurements of the ac susceptibilities will be presented.
REFERENCES. CHAPTER 2


FIGURE CAPTIONS

Fig. 2.1. Schematic diagram of a simple Josephson tunneling junction. \( S_1 \) and \( S_2 \) are superconductors. \( I \) is an insulator.

Fig. 2.2. Dependence of maximum supercurrent through a Josephson junction upon the flux threading the junction.

Fig. 2.3. Cylindrical geometry of the bulk superconducting sample of radius \( R \). The axis of the cylinder is along the \( z \) axis. Magnetic field \( H(t) \) is applied to the \( z \) axis. The superconducting grains are approximated by cylinders of radius \( R_g \), aligned along the \( z \)-direction parallel to the applied field \( H(t) \).

Fig. 2.4. Computed time sequence of the local magnetic field \( H(r) \) inside a cylindrical superconductor. The fields are calculated numerically from Eq. 2.16 for the parameter values, \( R = 0.085 \) cm, \( H^* = 5.39 \) Oe, \( H_s = 5.0 \) Oe, \( H_0 = 2.0 \) Oe, and \( H_1 = 3.0 \) Oe. The magnetic field is given by \( H = H_0 + H_1 \cos(\pi n/10) \), where \( n = 0, 1, 2, \ldots, 20 \). (a): \( H \) is decreasing; \( n=0 \to 10 \), (b): \( H \) is increasing; \( n=10 \to 20 \).
Figure 2.1

Figure 2.2

Figure 2.3
Figure 2.4

(a) 

(b)
CHAPTER 3. EXPERIMENTAL PROCEDURES†

3.1. Samples

In Table 3.2 the various $\text{YBa}_2\text{Cu}_3\text{O}_7$ samples used in the experiments are listed. National Superconductor and Colorado Superconductor Inc. are producing high quality polycrystalline single-phase ceramic pellets with a disc-shape or rectangular bar-shape. They produced YBCO samples by following a conventional method: sintered from precursor chemicals of high grade, calcined and sintered, then annealed in flowing oxygen. All samples have superconducting transition temperatures of well above liquid nitrogen temperature.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Shape Description</th>
<th>Size</th>
<th>Maker</th>
</tr>
</thead>
<tbody>
<tr>
<td>#1</td>
<td>Sintered cylinder</td>
<td>$R=0.085\text{ cm}, 0.95\text{ cm long}$</td>
<td>Colorado Superconductor</td>
</tr>
<tr>
<td>#2</td>
<td>Sintered cylinder</td>
<td>$R=0.085\text{ cm}, 1.05\text{ cm long}$</td>
<td>National Superconductor</td>
</tr>
<tr>
<td>#3</td>
<td>Sintered cylinder</td>
<td>$R=0.085\text{ cm}, 0.90\text{ cm long}$</td>
<td>A. Zettl</td>
</tr>
<tr>
<td>#4</td>
<td>Powder from #2</td>
<td>$R=0.10\text{ cm}, 1.03\text{ cm long}$</td>
<td>National Superconductor</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Powder size~10\text{ μm}</td>
<td></td>
</tr>
</tbody>
</table>

Table 3.2. $\text{YBa}_2\text{Cu}_3\text{O}_7$ samples used in the experiments

The pellets are cut into rectangular shapes with a diamond saw and they are rolled on a very fine carborundum paper #400 to make cylindrical samples with a radius

† In this chapter MKS units are used.
of 0.085 cm. Powdered sample #4 comes from grinding part of the pellet used for the sample #2 with an agate mortar and this powder is packed in a quartz tube with an approximate 2 mm inner diameter. An optical microscope shows a distribution of size of powders but the average size is approximately 10μm which is comparable to the size of superconducting grains. Sample #3 was grown in a quartz tube with an inner diameter of 2 mm and it was rolled on carborundum paper to get the final radius. The density of the cylindrical samples was measured and they all showed approximately 4.0 g/cm which is about 65% of the theoretical density from X-ray experiments. That indicates that the samples used have almost the same packing factor. The measured resistive superconducting transition temperature of all samples was approximately 90 ± 2 K and was measured exactly from the temperature dependence of the susceptibility, discussed below.

3.2. Experimental Setup

Ac magnetic susceptibility can be measured in various ways. Among those most commonly used one is a two-coil method which employs an ac magnetic field coil and a pickup coil. A typical circuit is shown in Fig. 3.1. This method usually requires compensation of the induced signal for the case of no sample. The compensation coil with identical size and shape as the pickup coil, but connected in opposite phase, reduces the output signal voltage to essentially zero, so that the measured signal with a sample is only from the sample. The induced signal voltage $V_s$, which is a complex voltage with magnitude and phase, in the pickup coil
due to the sample is given by\textsuperscript{1}

\[ V_s = -L^* l \frac{dM}{dt} \]  

(3.1)

where \( L^* \) is the mutual inductance between the pickup coil and the sample with the magnetization \( M \), and \( l \) is the sample length. This quantity can be calculated numerically from the formula in Ref. 1 and is closely related with the geometry of the pickup coil and the sample shape. If the ac field is assumed to be uniform, \( M = \chi H \) where \( \chi \) is the complex ac magnetic susceptibility and \( H = H_1 \cos 2\pi ft \) the applied ac magnetic field. Rearranging Eq. 3.1 we get the final formula in MKS units

\[ \chi = \frac{V_s}{L^* l 2\pi f H_1} \]  

(3.2)

As shown in the Fig. 3.1, a lock-in amplifier measures the in-phase and out-of-phase voltage signal induced and using Eq. 3.2 the real and imaginary part of ac susceptibility of the magnetic sample is determined from the in-phase and out-of-phase voltage respectively.

This two-coil susceptometer is mostly used for the measurements of ac magnetic susceptibility but it has disadvantages. First, the frequency range of usual commercial lock-in amplifiers is less than 100 kHz. Second, the susceptibility is very sensitive to phase variation during dc magnetic or ac magnetic field sweep so the design of coils geometry is critical to the accurate measurements. The inductive coupling of various coils should be minimized.
The method used in the experiments in this thesis is a one-coil method shown in Fig. 3.2. This consists of a single coil around the cylindrical sample, which acts as an ac field coil; it is connected to an HP 4192A impedance analyzer. An outer coaxial dc field coil is driven by an HP 3325A function generator. When a sinusoidal current $I = I_1 \cos 2\pi ft$ is driven through the pickup coil by the impedance analyzer, the induced voltage across the pickup coil without a sample is given by

$$V_0 = -L_s \frac{dl}{dt} + I R_s,$$  \hfill (3.3)

where $L_s$ and $R_s$ are the self inductance and resistance of the pickup coil respectively. From Eqs. 3.1 and 3.2 the sample contribution to the voltage is

$$V_s = L^* I \chi 2\pi f H_1.$$  \hfill (3.4)

The field $H_1$ is proportional to the current in the coil

$$H_1 = c I_1$$  \hfill (3.5)

where $c$ is a proportionality constant which be readily computed. The total voltage is given then by

$$V_0 + V_s = i 2\pi f I_1 (L_s + cL^* \chi') + 2\pi f I_1 (R_s - 2\pi fc L^* \chi'')$$  \hfill (3.6a)

which can be rearranged as

$$= i 2\pi f I_1 (L_s + \Delta L) + 2\pi f I_1 (R_s + \Delta R)$$  \hfill (3.6b)

which defines $\Delta L$ and $\Delta R$; they are the change of inductance and resistance respectively of the pickup coil due to the presence of sample, and $i = \sqrt{-1}$.

From the definition of the ac magnetic susceptibility $\chi \equiv \chi' - i \chi''$, the ac susceptibility components are given by
\[ \chi' = \frac{\Delta L}{c \ L^* \ l}, \]  
\[ \chi'' = \frac{\Delta R}{2\pi fc \ L^* \ l}. \]  

(3.7a)  
(3.7b)

The impedance analyzer (model HP 4192A) is designed to measure a wide range of complex impedance parameters simultaneously, that is, \( L \) and \( R \), \( |Z| \) and \( \theta \), or \( C \) and \( Y \), etc, for a frequency from 0.5 Hz to 13 MHz. The impedance measurement function of the HP 4192A is based on the vector-voltage-current ratio measurement method. In this method, the impedance of the pickup coil is determined by measuring the vector ratio between the applied ac signal voltage and the current flowing through the coil. As shown in Fig. 3.3, a voltage-to-current converter amplifier which has a range resistor feedback circuit is employed to detect the vector current through the coil. The \( I-V \) converter causes a current to flow through the range resistor \( R_r \) equal to the current through the coil. Thus, the output voltage of the \( I-V \) converter is equal to the product of coil current and the range resistor value. Accordingly, the coil impedance is determined from the ac signal voltage, the output voltage of the \( I-V \) converter, and the range resistor value. The auto-balance bridge circuit employed in the HP 4192A permits the impedance measurement from 5 Hz up to the 13 MHz region. Fig. 3.4 shows the auto-balance bridge circuit. The ac signal source applies a signal \( e_s \) to the coil and causes a current \( i_x \) to flow through the coil. This yields the current \( i_r \) which flows through the range resistor \( R_r \). The variable amplitude-phase oscillator applies a signal \( e_r \) of the same frequency as the ac signal to the range resistor. Currents \( i_x \) and \( i_r \) can
be balanced by controlling the output of the variable amplitude-phase oscillator. The null detector detects the difference \( i_d = i_x - i_r \). When the variable amplitude-phase oscillator is adjusted for \( i_d = 0 \), the impedance \( Z_x \) of the coil is calculated from the vector voltages as follows:

\[
Z_x = R_r \frac{e_s}{e_r}.
\]  

(3.8)

The amplitude-phase oscillator from the null detector output automatically balances the bridge to give the impedance values. During the measurements of magnetic susceptibility \( L \) and \( R \) of the pickup coil were measured.

<table>
<thead>
<tr>
<th>Pickup Coils</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Coil</strong></td>
</tr>
<tr>
<td>#1</td>
</tr>
<tr>
<td>#2</td>
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<tr>
<td>#3</td>
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<tr>
<td>#4</td>
</tr>
<tr>
<td>#5</td>
</tr>
</tbody>
</table>

*Table 3.3. Pickup coils used in the experiment for different frequency and ac magnetic field ranges; \( l \) = coil length.*

The resolution of the impedance analyzer is strongly dependent on the complex impedance of the coils used for the experiment. For easiness of change of samples the pickup coils were wrapped around quartz tubes with an outer diameter
of 3.0 mm and an inner diameter of 1.8 mm with #40 insulated copper magnet wire. Solenoid pickup coils with different number of turns, length, and layers were tested to get the best resolution and they are listed in Table 3.3. As the number of turns of a coil increases, the stray capacitance and inductance increases. Thus the pickup coil behaves as a resonant circuit. The resonant frequency of the coil is decreasing with increasing the number of turns and the resonant frequency is an agent which limits the useful frequency range of the given pickup coil as shown in the frequency column in Table 3.3.

A schematic diagram of the experimental setup of the one-coil ac susceptometer is shown in Fig. 3.5. It consists of a quartz tube with a pickup coil wrapped around it, a dc magnetic field coil, and a mu-metal shield. The whole assembly is immersed in a liquid nitrogen dewar to cool it at 77 K. The sample is inserted into the quartz tube from the top and placed in the center of the pickup coil. For each experimental run, the impedance analyzer measures the series inductance \( L \) and resistance \( R \) of the pickup coil with a sample; and data are recorded by computer in a file. Then an otherwise identical run is made without the sample. This procedure is done for varying parameter values of the dc field, ac field, frequency of ac field, and temperature. As temperature lowers below \( T_c \), \( L \) decreases due to Meissner effect or dc shielding effect but \( R \) increases due to ac loss of superconductors.

In order to measure temperature dependence of ac susceptibility a simple cryogenic setup was used. Instead of the quartz tube in Fig. 3.5, the new sample
tube shown in Fig. 3.6 was made. The pickup coil was wrapped around a short piece of (highly heat-conductive) quartz tube. The top of the quartz tube is attached to a very thin stainless steel tube to minimize heat conduction from the top; the diameter of the tube is 1/8 inch and its wall thickness is $10^{-2}$ inch. The bottom of the quartz tube is attached to an oxygen-free copper rod with a heater. The heater is made of #36 Manganin wire of 27 $\Omega$ total resistance. A small amount of heat sink compound was used between the copper rod and the quartz tube to increase the heat transfer to the sample and to reduce the thermal gradient of the sample. This sample tube is then inserted into a another thin wall stainless steel tube with 1/4 inch diameter. The sample tube is contacted to the outer stainless steel tube by flexible brass springs soldered around the copper rod.

With this cryogenic design the temperature can be changed from 77 K to room temperature by increasing the heater current. The temperature controller was not necessary to maintain a constant temperature. Without a controller temperature was quite stable within 0.5 K. A thin copper-constantan thermocouple was inserted through the sample tube to read the sample temperature. The thermocouple was pressed onto the top of the sample whose top was coated with a silicone heat sink compound (type z9) manufactured by GC Electronics Co. As will be seen in Chapter 4, the superconducting transition temperature was found to be about 90 K for all samples.

3.3. Data Acquisition
A block diagram of the system is shown in Fig. 3.7. The data acquisition during the experiments is done automatically by a computer, LSI-11. Various experimental devices are connected to a IEEE 488 controller in the computer. The impedance analyzer (HP 4192A) used in the experiment can be programmed to control the frequency $f$ of the ac field as well as the amplitude $H_1$. The frequency and ac source voltage (or ac source current) across the pickup coil can be read directly so that frequency and magnitude of ac magnetic field to the sample can be stored into a file on the hard disk of the computer. The impedance analyzer also reads the complex impedance of the coil, here the inductance and resistance which can be stored in the file. The dc magnetic field is provided by a synthesizer (HP 3325A) for fields up to $H_0 = 40$ Oe. For higher fields a dc power supply (HP 6274B) is used. The dc source is connected to a digital multimeter (Keithley 197) to read the current through the dc coil. From the geometry of the dc coil the dc magnetic field is calculated. For the dc power supply, the dc output current can be changed with an input voltage to programmable control of the power supply output voltage. This input voltage is supplied from the digital-to-analog-converter (DAC) controlled by the computer. For the HP 3325A synthesizer the computer can program the sweep time and sweep voltage, so we can have a slow dc field scan; this is very useful for measuring the dependence of $\chi'$ and $\chi''$ on $H_0$. The temperature of the sample is read by another digital multimeter (Keithley 197) via a Copper-Constantan thermocouple. The thermocouple has a 77 K reference junction and the program automatically converts the TC voltage into absolute
temperature. A very stable dc power supply described in Part I of this thesis is used to supply the dc voltage to the heater. With this power supply the temperature can be set at fixed value. In order to decrease or increase temperatures continuously a ramp voltage with a large sweep period from the synthesizer is used and the temperature is monitored continuously with the thermocouple described above. When the time of lowering the temperature from 100 K to 77K was greater than 1 hour, thermal hysteresis in susceptibility was negligible (less than 1 %). But the for sweep time of 30 min, thermal lag gave rise to the hysteresis of 5%.

The experimental results for each run are stored as independent files on hard disk. Each file contains the sample name, the frequency of ac field, magnitudes of applied ac and dc field, the inductance and resistance of the pickup coil, the temperature of the sample, and the computer clock time. The computer programs to take data are given in Appendix C.

The two components of the complex ac susceptibility $\chi'$ and $\chi''$ were calculated by using Eqs. 3.7a and 3.7b respectively. In the equations $\Delta L$ and $\Delta R$ are the difference of $L$ and $R$ of the pickup coil with the sample from those of the same coil without the sample, but with all other parameters the same. $\Delta L$ is always negative for the superconductors because of diamagnetic property of the sample below $T_c$. $\Delta R$ is positive because of ac loss of the superconductors. It is easy to calculate $\Delta L$ and $\Delta R$ for $H_0$ or $H_1$ scan at fixed $T$ because the impedance values in the data file of the empty coil were not changed as the magnetic field changes. We obtained $\Delta L$ or $\Delta R$ of the sample by subtracting a constant, which is
$L$ or $R$ of the empty coil, from $L$ or $R$ of the coil with the sample. For temperature scan $R$ of the empty coil decreases linearly as $T$ decreases but $L$ remained almost constant. In order to get $\Delta L$ or $\Delta R$ from the files with and without the sample for $T$ scan we interpolated the impedance of the empty coil so that $T$ of the impedance of the coil with the sample is the same as that of the empty coil. Other parameters for the files above are exactly the same.

The precise measurement of magnetic susceptibility involves several calibrations and corrections. The first one is the consideration of the demagnetization factor, which is a geometrical property of the sample shape. When the cylinder is not sufficiently long, the measured susceptibility $\chi$ should be corrected for demagnetization fields

$$\chi_{in} = \frac{\chi}{1 - D\chi},$$

where $\chi_{in}$ is the intrinsic susceptibility, corrected for the demagnetization and $D$ is the demagnetization factor ($0 < D < 1$). The values of $D$ for uniformly magnetized cylinders are given in Ref. 1 and 2. For the cylindrical samples in Table 3.2 the demagnetization factor is only about 0.04 because of large ratio of length/radius. Since the susceptibility is a complex quantity, we can rewrite Eq. 3.9 into

$$\chi'_{in} = \frac{\chi' - D(\chi'^2 + \chi''^2)}{(1 - \chi'D)^2 + D^2\chi''^2}$$

(3.10a)

and

$$\chi''_{in} = \frac{\chi''}{(1 - \chi'D)^2 + D^2\chi''^2}.$$  

(3.10b)
The susceptibility data given in Chapter 4 are corrected using Eqs. 3.10.

There is also a correction to the measured susceptibility owing to the resonance of the pickup coil. As shown in Fig. 3.8, equivalent circuit of the pickup coil is a combination of a capacitor, inductor, and resistor. As the number of turns of the coil increases, the capacitance as well as the inductance increases so that the resonance frequency becomes smaller. The impedance analyzer does not know the equivalent circuit of the coil so we should deduce the intrinsic $L$ and $R$ of the coil from the impedance outputs. When the measuring frequency range is assumed to be much smaller than the resonant frequency of the coil ($\omega \ll \sqrt{\frac{1}{L \cdot C}}$), the intrinsic impedance is approximately given by the following linearization,

\[
\Delta L = (1 + \omega^2 L_{in}) \Delta L_{in} - 2R_{in} C_{in} (1 + 2\omega^2 L_{in} C_{in}) \Delta R_{in} \tag{3.11a}
\]

and

\[
\Delta R = 2\omega^2 R_{in} C_{in} \Delta L_{in} + (1 + 2\omega^2 L_{in} C_{in}) \Delta R_{in} \tag{3.11b}
\]

where $C_{in}$ is a parallel parasitic capacitance of the coil, $\omega = 2\pi f$, and $f$ is the frequency of ac field. The susceptibility data given in Chapter 4 have been corrected using Eqs. 3.11.

In Chapter 4 the corrected susceptibility measurements on the YBCO samples as functions of many variables such as temperature and magnetic fields will be presented.
REFERENCES. CHAPTER 3


FIGURE CAPTIONS

Fig. 3.1. Typical circuit of ac magnetic susceptometer using a two-coil method. The pickup coil(3) and the ac magnetic field coil(2) surround a cylindrical sample(5) coaxially. It has usually a compensation coil(4) which is ideally identical to the pickup coil to compensate the signal when there is no sample. The outer coaxial solenoid coil(1) is for applying dc magnetic field.

Fig. 3.2. Schematic diagram of one-coil ac magnetic susceptometer used in this thesis. Here the coil(2) around a cylindrical sample(3) produces an ac magnetic field and also serves as a pickup coil for an impedance analyzer. The outer coaxial solenoid coil(1) is for applying a dc magnetic field which can be slowly ramped. Data are taken with and without the sample in place, but under otherwise identical condition.

Fig. 3.3. Vector-voltage-current ratio measurement method for HP 4192A impedance analyzer using the range resistor amplifier. \( R_r \) is the range resistor and DUT means the device under test. In the experiment the DUT is the coil(2) in Fig. 3.2.

Fig. 3.4. Schematic diagram of the auto-balance bridge circuit of HP 4192A impedance analyzer.

Fig. 3.5. Schematic of the experimental ac susceptometer apparatus. See also Fig. 3.2 for detection and recording system. 1: glass dewar, 2: dc magnet coil, 3: pickup coil, 4: superconducting sample, 5: 3 mm OD quartz tube, 6: \( \mu \)-metal mag-
netic shield, 7: BNC connector for pickup coil, 8: connector for other wires, 9: liquid nitrogen, 10: plastic cap.

**Fig. 3.6.** Simple cryogenic insert system for ac susceptometer. This assembly of a sample tube and outer stainless steel tube is fitted into the ac susceptometer in Fig. 3.5. 1: 1/4 inch OD stainless steel tube, 2: 1/8 inch OD stainless steel tube, 3: superconducting sample, 4: pickup coil, 5: heater, 6: copper rod, 7: brass springs, 8: plastic cap, 9: liquid nitrogen, 10: thermocouple.

**Fig. 3.7.** Block diagram of data acquisition system. DMM: Keithley 197 digital multimeter. Data, which are $L$, $R$ of the pickup coil, $H_0$, $H_1$, $f$, and $T$, are stored in a file on hard disk.

**Fig. 3.8.** Equivalent circuit of a pickup coil. $L$ and $R$ are the measured quantities by an impedance analyzer. $L_{in}$ and $R_{in}$ are the intrinsic quantities of the impedance of a pickup coil.
Figure 3.1

Figure 3.2
\[ i_x = \frac{e_s}{Z_x} = \frac{e_r}{R_r} \quad \therefore \quad Z_x = R_r \frac{e_s}{e_r} \]

**Figure 3.3**

\[ f_r = f_s \]

**Figure 3.4**
Figure 3.5
Figure 3.7

Figure 3.8
CHAPTER 4. EXPERIMENTAL RESULTS AND DISCUSSIONS

In this chapter we will present experimental data of ac magnetic susceptibility as functions of applied magnetic fields, temperature, type of samples, and frequency. Although many authors have reported ac magnetic susceptibility measurements of high-temperature superconductors$^{1-11}$, a complete picture of the behavior of the ac susceptibility has not yet emerged because of lack of systematic studies. These data are related to the granular nature of high-temperature superconductors; the theoretical models discussed in Chapter 2 will be used to try to explain the measured ac susceptibility data. Throughout this chapter theoretical calculations of ac susceptibility using a modified critical state model with two components are presented and compared with the experimental data. The modified critical state model can explain satisfactorily the data in spite of its simplicity. A hysteresis of the ac susceptibility observed and described in this chapter has not been previously reported, although that of the dc magnetization $M(H)$ is quite commonly reported as shown in Fig. 1.3. The possible origin and magnetic behavior of the hysteresis is discussed in terms of the modified critical state model.

4.1. Magnetic Field Dependence of AC Magnetic Susceptibility

Complex ac magnetic susceptibilities were measured by applying ac and dc magnetic fields at fixed temperature as discussed in Chapter 3. The cylindrical samples described in Table 3.2 were used and the sample was placed in the coil
quartz tube in the center in Fig. 3.5 for fixed temperature at 77 K, or else placed in the sample tube in Fig. 3.6 for measuring data at higher temperatures by applying a constant heater current through the heater. For best resolution and stability of the HP impedance analyzer we used approximately 10 kHz, but frequencies from several kHz to several 100 kHz did not give any significant change of ac susceptibility. Coils #4 or #5 in Table 3.3 which cover the optimum frequency range mentioned above were used for the most of measurements.

In what follows we will show all the experimental data (upper figures, Figs. 4.1, ... , 4.4) for three different temperatures, and the corresponding model calculations (lower figures, Figs. 4.1, ... , 4.4). A quick overview of the figures shows good agreement as the reader can easily ascertain. The details are now presented.

The experimental data have been measured by applying only an ac magnetic field at room temperature and cooling the sample below $T_c$ and then applying a constant or scanned dc magnetic field. In Fig. 4.1 to Fig. 4.3, the upper figures show experimental data for complex ac susceptibilities, $\chi'$ and $\chi''$, and are shown as a function of a scanned dc magnetic field $H_0$ (a,b), and as a function of a scanned ac magnetic field $H_1$ (c,d). The lower figures show the corresponding calculated susceptibility (e-h) from the modified critical state model, to be fully discussed in Section 4.1. The same sintered polycrystalline cylindrical rod of YBCO (sample #2 in Table 3.3 from National Superconductor, Inc.) and coil #4 in Table 3.3 were used for the data in Figs. 4.1, 4.2, and 4.3. Fig. 4.4 shows similar plots for sample #3 from A. Zettl and coil #4. The data show some common
features:

(1) $\chi'$ as a function of $H_0$ (Figs. 4.1(a), 4.2(a), 4.3(a))

At $H_0 = 0$ and $H_1 \leq 0.03$ Oe, $\chi' \approx -1$, i.e., fully diamagnetic. Then $\chi'$ increases monotonically as $H_0$ increases for small $H_1$. The rate of increase of $\chi'$ is strongly dependent on the values of $H_1$ and temperature $T$. The higher $H_1$ is, the larger the increase rate of $\chi'$ is. The higher $T$ is, the larger $\chi'$ and the increase rate of $\chi'$ are. For relatively large $H_1$ and $H_0$, $\chi'$ is saturated at $\chi' \approx -0.5$ so that $\chi'$ is constant but is still negative. A very interesting feature of $\chi'$ appears when $H_1$ is large, for example $H_1 \geq 5$ Oe at $T = 78.3$ K (Fig. 4.1(a)): the minimum of $\chi'$ is located at nonzero $H_0$, in this case at $H_0 \approx 5$ Oe. This is quite unique and later in this section it will be shown that the critical state model produces exactly the same result. Another important feature is the appearance of hysteresis of $\chi'(H_0)$ as $H_0$ is scanned up and down. When $T$ is low, hysteresis is barely observed, Fig. 4.1(a). But for higher $T$ hysteresis is clearly observed and is larger for smaller $H_1$, Fig. 4.3(a). Hysteresis of dc magnetic measurements such as dc magnetization has been reported by many authors but the observation of hysteresis of ac magnetic susceptibility has not been reported yet. We believe that this is the first time the hysteresis phenomena of ac magnetic susceptibility of high temperature superconductors has been reported and discussed, as below in Section 4.3 in more detail; it cannot be easily explained by the standard critical state model.

(2) $\chi''$ as a function of $H_0$ (Figs. 4.1(b), 4.2(b), 4.3(b))
At $H_0 = 0$ and $H_1 \leq 0.03$ Oe, $\chi''$ is essentially zero, i.e., there is essentially no loss. However $\chi''$ shows much more complicated behavior as $H_0$ and $H_1$ vary. When the value of $H_1$ is small, $\chi''(H_0)$ increases monotonically within our measurement range of $H_0$. For larger $H_1$, $\chi''(H_0)$ develops a maximum at a certain value of $H_0$ whose value becomes smaller and smaller and finally zero as $H_1$ is made larger, Fig. 4.1(b). Hysteresis of $\chi''(H_0)$, when $H_0$ is scanned up and down, is also larger with increasing $T$ (eg., Fig. 4.3(b)), and is similar to be behavior of $\chi'(H_0)$.

(3) $\chi'$ as a function of $H_1$ (Figs. 4.1(c), 4.2(c), 4.3(c))

The $\chi'$ dependence on $H_1$ shows a monotonic increase of $\chi'$ with increase of $H_1$ that is strongly dependent on a value of $H_0$. The larger the applied dc magnetic field is, the larger the $\chi'$ is. The initial rate of increase of $\chi'(H_1)$ is larger for a larger dc field. It should be noticed that at large $H_1$ field $\chi'$ can be lower, i.e. more diamagnetic shielding, for a higher $H_0$ than for a lower $H_0$; specifically in Fig. 4.1(c) $\chi'$ for $H_0 = 2$ or 5 Oe is lower than that for $H_0 = 0$ Oe at $H_1 \geq 5$ or 7.5 Oe respectively. $\chi'$ for large $H_0$ is saturated ($\chi' = -0.5$) for large $H_1$. An interesting phenomenon is that there is no magnetic hysteresis in $\chi'(H_1)$ as $H_1$ is scanned up and down; this is in contrast to $\chi''(H_0)$.

(4) $\chi''$ as a function of $H_1$ (Figs. 4.1(d), 4.2(d), 4.3(d))

When there is no dc magnetic field, $\chi''$ increases monotonically at low $H_1$ and then it has a maximum at intermediate ac field $H_1^{\text{max}}$. At larger $H_1$, $\chi''$
decreases monotonically. For nonzero $H_0$, $\chi''(H_1)$ has in general two local maxima; for example, in Fig. 4.3(d) for $H_0 = 2$ Oe there are local maxima at $H_1 \approx 1$ and 4 Oe. As $H_0$ increases, the peak value of the maxima of lower $H_1$ becomes larger than that of the maxima of larger $H_1$; this can be seen in Fig. 4.3(d). $H_1^{\text{max}}$ is strongly dependent on $T$ and $H_1^{\text{max}}(T)$ is larger when $T$ is lower. No hysteresis of $\chi''(H_1)$ is observed as $H_1$ is scanned, similar to $\chi'(H_1)$. Later in this section we show that $H_1^{\text{max}}$ for $H_0 = 0$ Oe in the figures is approximately the penetration field $H_J^*$ of the so-called intergranular junctions, which is one of parameters used in the critical state model.

The sample dependence of $\chi$ was checked by using sample #3 (A. Zettl group) in Fig. 4.4 which has essentially the same geometry and density as sample #2. Fig. 4.4 shows the measured complex ac magnetic susceptibility of sample #3 as functions of $H_0$ and $H_1$ at $T = 78.3$ K. One can see that Fig. 4.4 is rather similar to Fig. 4.3 in general and this suggests that this magnetic behavior of $\chi$ is generic for the high temperature superconductor YBCO. As will be shown, the data of Fig. 4.4 can be understood from the same model as that for Fig. 4.3, but with somewhat different parameter values; see Table 4.3.

It should be noted that the experimental data in this section show interesting behavior mainly at low magnetic fields, in the range 0 to 20 Oe. It has been predicted that this large sensitivity of the susceptibility to low field is mainly due to the intergranular junctions. At higher field the susceptibility is expected to show quite different behavior because the field penetrates fully into the junctions.
region and starts to penetrate into the superconducting grains. This change of the magnetic response of the YBCO superconductor will be discussed in Section 4.3. The superconducting grains have a very large penetration field and shielding current density. If we can increase $H_1$ up to $H_g^*$, the penetration field of the grains, another $\chi''$ peak will appear due to energy loss related to fluxon motion into and out of the grains. This will provide a clear evidence of existence of the two components in YBCO sample and it has been observed by Lam$^{24}$.

4.1.1. Modified Critical State Model

In this section we now more explicitly describe a modified critical state model that we use to compare to the data in this thesis and hence to interpret it. The model predictions are shown as the lower figures in Figs. 4.1, 4.2, 4.3, 4.4. As explained in Chapter 2, high temperature superconductors are assumed to consist of superconducting grains surrounded by intergranular junction materials and they behave as type II superconductors. The critical state model in Section 2.2 can describe magnetic behavior of such a dirty superconductor and can be extended to apply to two components, intergranular and intragranular. Here we apply the two-component critical state model to explain the measurements of ac susceptibility shown in this section; in addition we modify the magnetic field dependence of the critical current density to get better fit of the data. Generalization of the critical current density can be obtained by introducing an exponent $\beta$ such as in the following expression:
\[ J_{cJ}(H) = \alpha_J \frac{c}{(|H| + H_{sj})^{\beta_J}} \]  
(4.1a)

\[ J_{cg}(H) = \alpha_g \frac{c}{(|H| + H_{sg})^{\beta_g}} \]  
(4.1b)

The Bean or Kim-Anderson current density is a special case of Eq. 4.1 when \( \beta = 0 \) or 1 respectively. This modification of the critical state model does not change the description in Section 2.3 except for change of the definition of the penetration field given by Eq. 2.18 into:

\[ H_J^* = \left[ 4\pi(1+\beta_J)\alpha_J R + H_{sj}^{1+\beta_J} \right] \frac{1}{1+\beta_J} - H_{sj} \]  
(4.2a)

\[ H_g^* = \left[ 4\pi(1+\beta_g)\alpha_g R_g + H_{sg}^{1+\beta_g} \right] \frac{1}{1+\beta_g} - H_{sg} \]  
(4.2b)

The bulk sample, a long cylinder of radius \( R \), is assumed to be composed of long cylindrical superconducting grains (intragranular) with radius \( R_g \), embedded in intergranular materials, which generally give weak links, such as Josephson junctions, or SNS junctions, possibly point contacts. We refer to this region by the various equivalent wordings, junctions, intergranular, weak links and do not have yet definite knowledge of their exact nature. The grains are aligned along the axis of the bulk cylindrical sample which is parallel to the applied magnetic field; see Fig. 2.3. If we assume that the intergranular material is nonmagnetic, we can define the effective temperature-dependent permeability\textsuperscript{12} as

\[ \mu_{\text{eff}} = f_n + f_s \left[ \frac{2I_1(R_g/\lambda_g)}{(R_g/\lambda_g)I_0(R_g/\lambda_g)} \right] , \]  
(4.3a)

\[ B_J = \mu_{\text{eff}} H \]  
(4.3b)

where \( f_n \) and \( f_s = 1-f_n \) are the fraction of intergranular material and
superconducting grains, respectively. $I_0$ and $I_1$ are modified Bessel functions of the first kind and $\lambda_g$ is the London penetration depth of superconducting grains. The effective permeability $\mu_{\text{eff}}$ is the same as $\mu_f$ in Eq. 2.17d for the intergranular region.

The first part in the right side of Eq. 4.3a is related to the pure intergranular region. The second part is related to the induced magnetic moment of the superconducting grains due to field penetration by temperature-dependent $\lambda_g$. Therefore $\mu_{\text{eff}}$ for the whole sample reflects the contribution from the pure intergranular region and the field-penetrated part of the grains.

It was reported\textsuperscript{13,14} that an expression for $\lambda_g(T)$ from two-fluid approximation\textsuperscript{15} for superconducting grains

$$\lambda_g(T) = \lambda_g(0) \left[ 1 - (T/T_c)^4 \right]^{-1/2}$$

(4.4)

could explain quite well experimental data when anisotropy along the axes is neglected. The reported value\textsuperscript{16} of $\lambda_g(0)$ from measurement of the magnetic susceptibility for YBCO crystal is approximately 1400 Å for the field perpendicular to the $a$-$b$ plane and 4200 Å for the field perpendicular to the $c$ plane. The expression of $\lambda_g(T)$ in Eq. 4.4 is strongly temperature-dependent and $\lambda_g(T)$ decreases very fast to $\lambda_g(0)$ as temperature decreases below $T_c$. Therefore the superconducting grains will show perfect diamagnetism except for their surfaces within the penetration depth $\lambda_g(T)$ for a small applied magnetic field. At low temperature imagine that magnetic property of high temperature superconductors at low mag-
The magnetic field is dominated by that of the intergranular materials. To test it the critical state model was first used for only a single component, the intergranular material.

As mentioned earlier many parameters which will be used in the model need to be chosen. As shown in Fig. 4.1 to Fig. 4.3, there are two sets of measured ac susceptibility data, $\chi(H_0)$ and $\chi(H_1)$, for each temperature. Particularly it was found from the model calculations that the $H_1^{\text{max}}$ value of $\chi''(H_1)$ peak for $H_0 = 0$ Oe on a $\chi''$ versus $H_1$ curve is almost same as $H_J^*$, the penetration field of intergranular material. Thus $H_J^*$ is easily obtained from the $\chi''$ peak in data plots (eg., Figs. 4.1(d), 4.2(d), ...). The results of the critical state model calculation are shown in (e)-(f) of Fig. 4.1 to Fig. 4.4 when we assume no intragranular contribution. In Table 4.4 fitting parameters of the calculations are shown.

<table>
<thead>
<tr>
<th>Figure</th>
<th>Sample</th>
<th>$T_c$</th>
<th>T(K)</th>
<th>R(mm)</th>
<th>$\mu_{\text{eff}}$</th>
<th>$H_J^*(\text{Oe})$</th>
<th>$H_{sJ}(\text{Oe})$</th>
<th>$\beta_J$</th>
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<tbody>
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<td>0.491</td>
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<td>0.509</td>
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<td>2.5</td>
<td>2.22</td>
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<td>#2</td>
<td>92.5</td>
<td>85.7</td>
<td>0.85</td>
<td>0.537</td>
<td>2.3</td>
<td>2.0</td>
<td>2.20</td>
</tr>
<tr>
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<td>#3</td>
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<td>78.3</td>
<td>0.85</td>
<td>0.840</td>
<td>1.7</td>
<td>1.7</td>
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</tr>
<tr>
<td>4.5</td>
<td>#2</td>
<td>92.5</td>
<td>78.3</td>
<td>0.85</td>
<td>0.491</td>
<td>8.0</td>
<td>2.5</td>
<td>1.0</td>
</tr>
<tr>
<td>4.6</td>
<td>#2</td>
<td>92.5</td>
<td>78.3</td>
<td>0.85</td>
<td>0.491</td>
<td>8.0</td>
<td>2.5</td>
<td></td>
</tr>
</tbody>
</table>

Table 4.4. Fitting parameters of the critical state model calculations
We measured directly $T$, $T_c$, and $R$ from the experiment. $H_J^*$ was obtained from $\chi''(H)$ data. $H_{sJ}$ and $\beta_J$ were assumed for best fit of the data. We determined $\mu_{\text{eff}}$ for best fit by assuming $\lambda_g(0) = 0.8 \, \mu\text{m}$, $R_g = 10 \, \mu\text{m}$, $f_n = 0.351$, and $f_s = 1 - f_n = 0.649$ for the sample #2; $\lambda_g(0) = 1.2 \, \mu\text{m}$, $R_g = 10 \, \mu\text{m}$, $f_n = 0.643$, and $f_s = 0.357$ for the sample #3. The values of $\lambda_g(0)$ were chosen arbitrarily and seemed to be large compared to 0.14 $\mu$m and 0.42 $\mu$m reported for the field perpendicular to the $a$-$b$ and $c$ plane of YBCO crystal\textsuperscript{16} respectively. If we choose a smaller $\lambda_g(0)$, the ratio of the intergranular region increases. For example, $f_n$ increases from 0.258 to 0.470 when $\lambda_g(0)$ decreases from 1.2 $\mu$m to 0.14 $\mu$m for sample #2; we believe this to be more realistic.

For the YBCO samples used in our experiment we found $\beta_J \approx 2$ which means a steeper field dependence of the intergranular critical current density $J_{cJ}(H)$ than that of originally assumed by Bean ($\beta = 0$) or Kim-Anderson ($\beta = 1$) for low temperature type II superconductors. $H_J^*$ has a strong dependence on temperature whereas $\beta_J$ and $H_{sJ}$ decrease slowly as $T$ increases.

During fitting of measured data with calculations we found the effects of $H_J^*$, $H_{sJ}$ and $\beta_J$ on $\chi$. The effect of $H_{sJ}$ was the least important among the three parameters and $\beta_J$ produced the most significant changes in $\chi$. When Fig. 4.1(e)-(h), calculated using only an intragranular component, is compared with Fig. 4.1(a)-(d), quantitative and qualitative agreements between them are excellent. This indicates that intragranular contribution to $\chi$ is negligible at this low mag-
netic field region because a lower critical field of the intragranular component is much larger than the range of the applied field. Thus the intergranular contribution is the dominant factor of magnetic response of these new superconductors at low applied magnetic fields. As temperature is raised, agreements are gradually degraded but still fittings are quite satisfactory in general. This may be attributed to flux creep which causes a gradual relaxation of flux on a time scale comparable to the measurement.

Two things should be noted from the results of the model calculations. First, disagreements of $\chi''$ are noticeable for small magnetic field, e.g., Figs. 4.3 and 4.4. This seems to indicate that the critical current dependence on a magnetic field used here (Eq. 4.1a), is not correct for small $H$. Eq. 4.1a gives a sharper rise of $J_c$ than necessary for the data. The smoothing of $J_c$ near $H = 0$ will give a better fit. Second, the behavior $\chi'$ for a large $H_0$ (e.g., Figs. 4.3(a) and 4.4(a)): the experimental $\chi'$ increases slowly without saturation after a sharp increase at low $H_0$. The corresponding calculation of $\chi'$, on the other hand, shows saturation after the sharp increase. This suggests that the slow increase of $\chi'$ in the data comes from penetration of magnetic vortex into a new component which has a higher $H^*$, namely superconducting grains or intragranular. In the range of magnetic field shown in the figures, $H_0 \approx 40$ Oe, effect of intragranular component is minor but it can be expected for grains to contribute more and more at larger $H$, either $H_0$ or $H_1$. This will be explained in more detail in Section 4.3.
To explain magnetic properties of superconductors, many different forms of critical current have been suggested\textsuperscript{17}. Conventionally the Bean or Kim-Anderson forms of critical current have been used most for models. Our experiments are believed to be the first systematic experiment which could lead to a determination of a field dependence of the critical current density. With resolution of our experimental apparatus, $J_{cJ}(H)$ can be chosen quite uniquely from the fitting. Fig. 4.5.1 and 4.5.2 show the results of the critical state model calculation with optimum fitting parameters for two forms of $J_{cJ}$ to fit the figures. For Fig. 4.5.1, the Kim-Anderson form of the critical current, which is a special case of Eq. 4.1a ($\beta_f = 1$),

$$J_{cJ} = \alpha_f \frac{c}{(|H| + H_{sf})}$$

was used, with $R = 0.85$ mm, $H_f^* = 8.0$ Oe, $H_{sf} = 2.5$ Oe, and $\mu_{eff} = 0.491$ (from Table 4.4). The discrepancy between the experiment and the calculation is evident for small $H_1$ in $\chi$ versus $H_0$ curves and for large $H_0$ in $\chi$ versus $H_1$ curves. For Fig. 4.5.2, another critical current form given by the following equation

$$J_{cJ} = A_f \ c \ \exp(-|H|/H_{sf})$$

was used for the calculation with $R = 0.85$ mm, $H_f^* = 8.0$ Oe, $H_{sf} = 2.5$ Oe, and $\mu_{eff} = 0.491$ (from Table 4.4). Because of fast decrease of $J_c(H)$ at large $H$, the saturation of $\chi'$ is quite noticeable in the measured range of $H$, and is a poor fit to the data.

From the comparison of the result of the calculations with different critical current forms it can be deduced that $\chi$ is quite sensitive to the choice of the current
form; it can be said to show an amplification of different assumption for $J_c(H)$. All three current forms in Eqs. 4.1a, 4.5, and 4.6 with given parameters are slightly different when they are plotted on a graph as a function of $H$ in Fig. 4.6; the susceptibility, $\chi'$ and $\chi''$, however are very different as shown in the Figs. 4.1, 4.5.1, and 4.5.2.

4.2. Temperature Dependence of AC Magnetic Susceptibility

The temperature dependence of the low frequency ac magnetic susceptibility of high temperature superconductors has been reported by many authors\textsuperscript{1,3,7,8,18}. In this section the experimental results and the model calculation of temperature dependence are discussed in detail.

The temperature dependence of ac magnetic susceptibility of the sample #2 and #3 was investigated with the experimental setup shown in Fig. 3.5 and 3.6. The cylindrical bulk samples were placed in the center of the pickup coil and heater current through a heater changed their temperature. The temperature was measured with a copper-constantan thermocouple which had a 77 K reference junction. The measurements were always done by decreasing temperature from $T$ above $T_c$. The frequency of the ac magnetic field was 10 kHz.

Fig. 4.7(a)-(b) show the observed temperature dependence of $\chi$ of the sample #2 for various $H_1$ values at $H_0 = 0$. At $T = 92.5 \pm 0.5$ K the sample #2 becomes superconductive as evident from Fig. 4.7(a): $\chi'$ shows a measurable drop (\textasciitilde1%). For a very small $H_1$, $\chi'$ rapidly decreases below $T_c$ and saturates at $\chi' = -1$ at 84
As $H_1$ increases, the range of transition of $\chi'$ to $-1$ becomes larger. In the figure the change of slopes on the $\chi'$ graph are clearly observed for $H_1 = 4.03$ and 7.91 Oe. This was discussed in Ref. 1 as to suggest that there are good and bad superconducting components in high temperature superconductors; these can now be identified as inter- and intragranular components. Non-zero $\chi''$ is observed only for the temperature range where $\chi'$ is not saturated. The higher $H_1$, the larger $\chi''$. The peak value of $\chi''$ saturates around 0.18 for large $H_1$. This dissipative loss is due to magnetic vortex penetration described by the critical state model, or is called a hysteretic loss which is explained in Section 4.5.

The $H_0$ effects on the temperature dependence of $\chi$ are shown in Fig. 4.7(c), (d) for $H_1 = 0.22$ Oe. The so-called two components effect is not so clear. The peak value of $\chi''$ saturates around 0.15 as $H_0$ increases, Fig. 4.7(d).

Our experimental data show a broader transition range in $T$ in comparison with those of other previously published papers. It is believed that a thermal gradient of the sample inside our sample cell due to its compact design broadens the range in $\chi'$ and smooths the peak in $\chi''$. In the experiments we could not observe a very small narrow peak in $\chi''$ just below $T_c$. Many believe it to be an intragranular loss peak.\textsuperscript{1,3,18} It was also reported that its appearance is strongly dependent on sample preparation and treatment.
The modified critical state model described above in Section 4.1.1 was applied to attempt to explain the observed temperature dependence. Here using the parameter values of the intergranular component found for the sample #2 in Table 4.4, chosen for fitting the data in Section 4.1, we derived the empirical temperature dependence of the parameters:

\begin{align}
H_J^*(T) &= 53.33 \left[1-(T/T_c)^2\right]^{1.5} \\
H_{sJ}(T) &= 13.24 - 0.13T \\
\beta_J(T) &= 2.86 - 7.76 \times 10^{-3}T.
\end{align}

(4.7a) (4.7b) (4.7c)

The origin of this empirical temperature dependence above is not clearly understood yet.

With Eqs. 4.3, 4.4, and 4.7, a computer program chiThac presented in Appendix B calculates the temperature dependence of \( \chi \) of the intergranular component for different values of \( H_1 \) and \( H_0 = 0 \). As mentioned in Section 4.1, it should be noted that the calculation includes the contribution of field penetration into each superconducting grain. Since \( \lambda_g(T) \) increases with increasing \( T \), the intragranular contribution becomes larger as \( T \) increases. The measured susceptibility is determined by the response of both inter- and intragranular components. Since we do not have enough information about the magnetic behavior of the intragranular component at present, we focus the calculation onto the behavior of the intergranular component in the following.

Fig. 4.7(e)-(h) show the results of the modified critical state model calculation for the intergranular component only of the sample #2. The calculations are in
qualitative and semiquantitative agreement with the experimental data (the upper figures in Fig. 4.7) in general. This implies that for the bulk YBCO superconductor, the intergranular property plays an important role in the temperature dependence of $\chi$ over a wide temperature range. For very small $H_1$ and low $T$, each grain is almost diamagnetic and has no ac loss. Thus the calculated susceptibility for the intergranular component should be essentially the same as that for the two components.

There have been attempts to explain temperature dependence of the ac magnetic susceptibility by Ishida and Goldfarb,7 Müller,11 and Berg and Koziowski.19 They assumed temperature dependence of parameters somewhat different from Eqs. 4.7a, 4.7b, and 4.7c. The agreement with their data is comparable to that of Fig. 4.7. More accurate measurement of $\chi$ as a function of temperature is needed to get fuller information on inter- and intragranular behavior.

Fig. 4.8 shows measured and calculated susceptibility as a function of $T$ for the sample #3. As briefly discussed for the field dependence of $\chi$, its temperature dependence is very different from that of the sample #2 (Fig. 4.7). From the data $T_c$ is approximately $91 \pm 0.5$ K. The $\chi$ was calculated as a function of temperature by assuming the following expressions (similar to those given in Eq. 4.7):

$$H_f^*(T) = 12.55 \left[ 1 - \left( \frac{T}{T_c} \right)^2 \right]^{1.5}$$ (4.8a)
$$H_{sf}(T) = 13.24 - 0.13T$$ (4.8b)
$$\beta_f(T) = 2.11 - 7.76 \times 10^{-3} T$$ (4.8c)

Agreement of the calculation shown in Fig. 4.8(c), (d) with the data is good for
One of the interesting magnetic properties of new superconductors is a magnetic hysteresis phenomenon which can be seen clearly in the $\chi'(H_0)$ and $\chi''(H_0)$ curves of Fig. 4.3 or Fig. 4.4 as the $H_0$ is swept up and down. Depending on the sign of $H_0/dt$, $\chi$ has different values so the $\chi$ curve for $dH_0/dt > 0$ does not coincide with the curve for $dH_0/dt < 0$ when $H_0$ sweeps one cycle slowly. For example in Fig. 4.3(a), $\chi'$ displays two local minima at $H_0 = 0 \pm \Delta H$, where $\Delta H$ is a measure of the hysteresis. In Fig. 4.3(b), it can be seen that $\chi''$ similarly displays two local minima, or even two local maxima, depending on the magnitude of $H_1$. As can be seen in the figures mentioned above, the hysteresis $\Delta H$ is strongly dependent on amplitude of the ac magnetic field $H_1$ and $T$. The smaller $H_1$, the larger the hysteresis. The higher $T$, the larger the hysteresis. Similar hysteresis has been observed in the measurement of nonresonant microwave absorption in resonance experiment, harmonics generation, and ac magnetic susceptibility which will be discussed here. These observations, in general, have the common features stated above, thus they seem to be related to each other. In spite of the importance of this phenomenon, it has not been fully understood yet. Sometimes, usage of the word hysteresis is misleading. One usage is the hysteresis of a plot of the dc magnetic $M(H_0)$ around a cycle of $H_0$, eg. Fig. 1.3, which can be readily understood for hard superconductors and for high-temperature superconductors.
from the Bean-Kim-Anderson critical state model. In the measurements \( M \) is usually measured by a SQUID magnetometer. The ac hysteresis for \( \chi' \) and \( \chi'' \) which will be discussed below is novel to high-temperature superconductors and cannot be explained with the BAK model. Recently Campbell\textsuperscript{29} has applied a new model to explain the dependence on magnetic history of critical currents in multigranular high-temperature superconducting samples using ideas of Josephson-junction-like links between grains and the critical state model. This will be discussed briefly later in this section as a possible model of the hysteresis of the ac susceptibility.

Fig. 4.9 shows one of the properties of the ac susceptibility hysteresis of a cylindrical bulk rod (sample #1) made by Colorado Superconductor, Inc. They were measured at \( T = 77 \text{ K} \) and \( H_1 = 0.22 \text{ Oe} \). If the magnitude of a maximum \( H_0^{\text{max}} \) is the same as that of the minimum \( H_0^{\text{min}} \) as in Fig. 4.9(a) and 4.9(b), then both \( \chi' \) and \( \chi'' \) have the property

\[
\chi(H_0) \big|_{s=1} = \chi(-H_0) \big|_{s=-1} \tag{4.9}
\]

where \( s \equiv \text{sign} \left( \frac{dH}{dt} \right) \). If \( \Delta H \to 0 \), i.e., no hysteresis, then

\[
\chi(H_0) \big|_{s=1} = \chi(H_0) \big|_{s=-1} \tag{4.10}
\]

and \( \chi(H_0) \) is a symmetric function. If \( |H_0^{\text{max}}| \neq |H_0^{\text{min}}| \), the symmetry of \( \chi \) about \( H_0 \) breaks down as shown in 4.9(c). In a \( \chi' \) versus \( H_0 \) curve, the value and position of the local minima are dependent on the \( |H_0^{\text{max}}| \) and \( |H_0^{\text{min}}| \). When the maximum magnitude of \( H_0 \) is smaller, the value of \( \chi' \) at the local minimum is more negative, which means more diamagnetic flux exclusion. The \( H_0 \) of the minimum is also nearer to zero. We believe that the local minima of \( \chi' \) occur when
penetrating magnetic flux totally cancels the remanent magnetization with opposite sign. Therefore the position of the local minimum of $\chi'$ is larger for larger $H_0^{\text{max}}$ since a larger remanent magnetization requires a larger magnetic field for cancellation to occur.

Figs. 4.10(a) and 4.10(b) show the $\chi$ dependence on magnetic history. The data were taken as follows: the sample #1 (Colorado Superconductor, Inc.) was cooled from room temperature to $T$ below $T_c$ without applying dc magnetic field $H_0$ and then $H_0$ was applied to produce the curve 1. Subsequent $H_0$ sweeps give the curves 2 and 3. The ac field ($H_1 = 0.22$ Oe) supplied from the HP impedance analyzer was continuously applied to the sample, starting at $T > T_c$. As shown in Fig. 4.10(a), (b), initial application of $H_0$ to the sample (curve 1) produces a $\chi$ curve which is quite different from the subsequent loop (curves 2 and 3), which reach a stationary loop after about 5 cycles (scan time/cycle = 600 sec). The microwave absorption curve\(^{21}\) is remarkably similar to this figure even though $T$ and $H_0$ are very different.

Fig. 4.11 plots $\chi'$ and $\chi''$ versus $H_0$ of the sample #2 for $H_0$ sweeps with various values of $|H_0^{\text{max}}|$ at $T = 78.3$ K and 85.7 K, and with $H_0^{\text{min}}$ always zero. For Fig. 4.11(a),(b), $T = 78.3$ K, $H_1 = 0.22$ Oe and $|H_0^{\text{max}}| = 40, 100, 400, 800$ Oe, respectively. When $|H_0^{\text{max}}| = 40$ Oe, there is almost no hysteresis and the position of the minimum is at $H_0 = 0$. As explained in Section 4.1, the magnetic behavior in this range is believed to be dominated by intergranular weak links which have a
small pinning force density. This also seems to indicate that the intergranular 
junction material has only a very small ac magnetic hysteresis. As $|H_0^{\text{max}}|$ is 
increased above $H_{c1g}$, the magnetic field penetrates into the grains beyond the 
penetration depth $\lambda_g$. This flux penetration into the grains increases $\chi'$ steadily 
after the rapid increase of $\chi'$ due to intergranular material.

These two regions of $\chi$ are a clear indication of two components, weak links 
and superconducting grains, in the bulk high-$T_c$ YBCO superconductor. At $H_0 \approx 
80$ Oe in Fig. 4.11(a),(b) we can observe the transition from the behavior of inter­
granular to intragranular. We will sometimes refer to the intragranular region as 
grains, although it is possible that the magnetic property arises from twinning 
dislocations in the grains themselves; in this case intergranular, or intragranular are 
more apt terms. This is also confirmed from the $\chi''$ curve that the sharp peak 
between $H_0 = 0 - 90$ Oe is the dissipation loss in the intergranular region; and the 
very broad peak at larger $H_0$ is that of the grains. Once the flux penetrates into the 
grains, the magnetic vortices are strongly pinned in the grains. As the sweep direc­
tion of $H_0$ changes, most of the penetrated vortices comes out of the grains but 
some of them are still pinned. This generates a remanent magnetization which can 
have a long lasting (dc) component as well as an ac component arising from the 
driving field $H_1$. This remanent magnetization changes the ac magnetic response 
sufficiently as to produce a large hysteresis loop. The larger the $|H_0^{\text{max}}|$, the larger 
the hysteresis. It is very interesting that the return path from $H_0^{\text{max}}$ to zero of the 
hysteresis loop does not show the sharp decrease of $\chi'$ at low $H_0$ of intergranular
material. From the figure we can conclude that the flux penetration into the intragrain and the corresponding remanent magnetization are the major source of the hysteresis of the ac susceptibility, $\chi'(H_0)$ and $\chi''(H_0)$.

The data in Fig. 4.11(c),(d), which are taken in the same fashion as Fig. 4.11(a),(b) but at a higher temperature, show a pronounced temperature dependence of the hysteresis. Since $H_{c1g}$ decreases with increasing $T$, the intragranular behavior appears above a smaller field, $H_0 \approx 20$ Oe. Furthermore, the magnetic hysteresis loop is much narrower than that at 78.3 K. High temperature increases the rate of flux creep\(^{15}\) so the flux pinning is much weaker and the remanent magnetization thus is smaller. The sense of rotation of the hysteresis loop of $\chi'$ in Fig. 4.11(c) is opposite (counterclockwise) of that of Fig. 4.11(a) (clockwise); this is not yet really understood.

Fig. 4.12 shows the dependence of hysteresis on $H_1$ of the sample #2. The data for increasing $H_0$ were taken with the first application of $H_0$ just after cooling the sample below $T_c$ without $H_0$. As $H_1$ is increased from 0.22 to 7.91 Oe, $\chi'$ clearly shows a saturation (cf. Fig. 4.12(a)) of the hysteresis loop in the intergranular region at $H_0 \geq 50$ Oe and then a steady increase at higher $H_0$, due to the grains. The sense of the hysteresis loop for $H_1 = 0.22$ Oe is opposite to that for 7.91 Oe; the behavior of $\chi''$ is more complicated (Fig. 4.12(b), (c)).

The hysteresis in ac susceptibility that we observe is believed to be related to the dynamics of trapped fluxons in the magnetic field inside the grains. As $H_0$ is
initially applied, the penetrated flux pinned at pinning centers such as defects creates a critical state and follows the critical state model. However when $H_0$ is decreased from $H_0^{\text{max}}$ (curve 2 in Fig. 4.10), if the flux is trapped, it can lose some reversibility. Thus the field pattern for increasing $H_0$ and decreasing $H_0$ are different at the same $H_0$ and consequently the hysteresis loop in the ac susceptibility appears. The dynamics of trapped flux is quite complicated since it is involved with the nature of flux-trapping centers, interaction between the flux vortices, and other factors. Here we argue in order to explain the hysteresis data that these trapped fluxons change the critical current $J_c(H)$ at given $H$ by adding an irreversible critical current $J_c^{irrev}(H)$ to an reversible critical current $J_c^{rev}(H)$ given by Eq. 4.3. Therefore after the first application of large $H_0$ the total critical current $J_c(H)$ is given as follows

$$J_c(H) = J_c^{rev}(H) + J_c^{irrev}(H). \quad (4.11)$$

A form of the total critical current could be guessed for the experimental data shown in Fig. 4.10, for example. The form can be generally written as

$$J_c(H) = \alpha \frac{c}{(|H-H_m|+H_s)^\beta} \quad (4.12)$$

where $H_m$ is the value of $H_0$ at the minimum of $\chi'$. $H_m$ and $\beta$ are parameters dependent on $H_0^{\text{max}}$ and the sweep direction of the dc field. Fig. 4.13 shows a possible critical current as a function of $H$ for fitting the data of Fig. 4.10. In the figure we used parameter values $\mu_{\text{eff}} = 0.52$, $(H_J^*, H_{J,1}, \beta, H_m)$: (2.4 Oe, 4.0 Oe, 1.8, 0 Oe) for curve 1, (2.4 Oe, 2.0 Oe, 1.2, 10 Oe) for curve 2, (2.4 Oe, 3.0 Oe,
1.7, 6 Oe) for curve 3. The results of the susceptibility calculation using the above parameters are shown in Fig. 4.10(c),(d). In general we can see a qualitative agreement between the calculation and the data. However at the turn-over region at $H_0^{\text{max}}$, $\chi$ of curve 2 is not well matched with that of curve 1 or 3. This suggests that during the change of $H_0$ sweep direction a transient state is formed and its critical current form is not characterized by Eq. 4.13. If the interpretation here is correct, it is quite interesting that trapped fluxons change the parameter $\beta$. Maybe that is why different samples have different values of the parameter $\beta$ because the samples were treated differently in preparation. Each sample may have different amounts of defects or impurities, so that $\beta$, which is closely related to the nature of pinning centers, can be variable. When we apply a larger dc field, flux penetrates more deeply and flux is more likely to be trapped inside. On the other hand, Lorentz force on fluxons becomes larger with increasing the field, so flux flow can occur. The value of $\beta$ of the sample is determined from the balance of these two counter tendencies.

A recent model proposed by Campbell has derived the result that a maximum in the critical current density can be made to appear at almost any value of the external field by either appropriate field cooling or field reversal; this corresponds to non-zero $H_m$ in Eq. 4.12. He treated high-temperature superconductor as grains that are weakly coupled by a distribution of Josephson-junction-like links which conduct current according to a phase relation between the grains. His results reach the same conclusion as ours such that intragranular field profile,
which changes in response to magnetic history, is the cause of the observed hysteresis.

4.4. AC Magnetic Susceptibility of Powdered Samples

For the bulk cylindrical samples used in the previous sections, the intergranular and intragranular behavior are clearly distinguished in a large $H_0$ field, as shown in Fig. 4.11 or 4.12. It can be imagined that the intergranular coupling between the superconducting grains might be broken when the bulk sample is ground to make a superconducting powder. If the size of the powder grain is comparable to or less than the size of the superconducting grains, the magnetic response of the powder will give information on the grains only. The sample #4 powder in Table 3.2 were made by grinding with an agate mortar a part of the YBCO disc (National Superconductor, Inc.), from which sample #2 was taken. The size of the powder grain was determined with an optical microscope and had about 10 $\mu$m of average radius. This is comparable with estimated grain size of YBCO samples. We measured the complex impedance of the pickup coil with the powder in it. The powder was sealed in a quartz tube with an inner diameter 3 mm. The height of the powder in the tube was about 10 mm so the shape of the powder container was the same as the bulk cylindrical sample used in the above experiments. The pickup coil was wound around the quartz tube.

The observed $\chi'$ of the powder shows almost perfect diamagnetism ($\chi' = -0.98$) at $H_0 = 0$ and $H_1 = 0.22$ Oe, as in Fig. 4.14. But there still is a slight trace
of intergranular effect, which is a fast rise of $\chi'$ near $H_0 = 0$, for small $H_1$. The dissipative loss of the powder was $\chi'' \leq 0.005$ which means that essentially no penetration of magnetic field occurred. Fig. 4.15 shows the $H_0$ dependence of $\chi'$ and $\chi''$ of the powder. In comparison with Fig. 4.12 of a bulk sample, $\chi'$ and $\chi''$ changes more smoothly and steadily. The dissipation loss reaches a maximum and then decreases. A large hysteresis loop is produced when $H_0$ is swept up and down. Temperature dependence of $\chi'$ of the powder was measured for various values of $H_0$ and $H_1$, Figs. 4.16(a), (b). The $H$ dependence of $\chi$ is much weaker than the bulk sample (Figs. 4.7(a), (b), (c), (d)) because the grains have much larger penetration fields than that of the intergranular material. The fall-off rate of $\chi'(T)$ of the powder just below $T_c$ is smaller than that of the bulk sample and similar to results reported previously$^{8,18}$.

In order to check the validity of the critical state model, the fitting parameters for the grains, $H_g^*, H_{sg}, \beta_g$, and $R_g$, should be estimated. As discussed in Section 4.1, $H_g^*$ can be given as the position of maximum of $\chi''$ in a $\chi''$ versus $H_1$ graph. Our experimental setup has a limit of $H_1$ (approximately 8 Oe) so it is impossible to find a peak corresponding to $H_g^*$ which is much larger than $H_J^*$. $H_J^*$ was found to be about 10 Oe at 77 K in our measurement. Fortunately $\chi''$ versus $H_1$ was measured up to $H_1 \approx 400$ Oe by Lam$^{24}$ with a two-coil method using the same sample. From the measurements at 77 K for a cylindrical specimen with a similar geometry to the sample #2, $\chi''$ clearly showed two maxima, at $H_1 \approx 10$ Oe and at $\approx 250$ Oe. The maximum of $\chi''$ at lower $H_1$ which comes from the intergranular
region has disappeared for the powdered sample. This confirms further that fine enough powder shows only the property of grains as expected. According to Eq. 4.2, the pinning strength \( \alpha \) is inversely proportional to \( R \):

\[
\alpha_f = \frac{(H_f^* + H_{sf})^{1+\beta_f} - H_{sf}^{1+\beta_f}}{4\pi(1+\beta_f)R}, \tag{4.13a}
\]

\[
\alpha_g = \frac{(H_g^* + H_{sg})^{1+\beta_g} - H_{sg}^{1+\beta_g}}{4\pi(1+\beta_g)R_g}. \tag{4.13b}
\]

In the following calculation we assume that \( H_f^* \) is 250 Oe. When we assume \( H_f^* = 250 \text{ Oe}, R_f \approx 10 \mu\text{m}, H_{sg} \approx H_{sf}, \) and \( \beta_g \approx \beta_f \), the pinning strength of the grains is found to be about \( 10^6 \) times larger than that of the intergranular region.

Fig. 4.15(c), (d) shows a result of the modified critical state model calculation with parameter values of \( H_g^* = 250 \text{ Oe}, H_{sg} = 30 \text{ Oe}, \) and \( \beta_g = 2.3 \) for the superconducting grains. The results shown in Fig. 4.15(c), (d) are not in quantitative agreement with the data in Fig. 4.15(a), (b), but are qualitatively correct. For \( H_1 = 7.91 \text{ Oe} \) the model predicts that \( \chi' \) increases monotonically and that \( \chi'' \) has a very broad peak, as the experimental data does. The calculated \( \chi' \) and \( \chi'' \) for \( H_1 = 0.22 \text{ Oe} \) are found to be too small to compare to the data. Some possible explanations are the distribution of \( H_g^* \) because of different quality of grains, distribution of \( H_{sg} \), demagnetization effect due to rough spherical shape of grains, and the anisotropy of critical current densities. The temperature dependence of \( \chi' \) and \( \chi'' \) of the powder was calculated by following the relation, similar to Eqs. 4.7 and 4.8:

\[
H_g^*(T) = 1469.3 \left[ 1-(T/T_c)^2 \right]^{1.5}, \tag{4.14a}
\]
\[ H_{sg}(T) = 40.22 - 0.13T, \quad (4.14b) \]
\[ \beta_g(T) = 2.47 - 7.76 \times 10^{-3}T. \quad (4.14c) \]

The results are shown in Fig. 4.16(c), (d) for the values of \( H_0 = 0, 300, \) and 800 Oe. This figure shows a very sharp transition for \( \chi' \) in zero dc field. The data in Fig. 4.16(a), (b) shows a very broad decrease of \( \chi' \). The calculated \( \chi'' \) shows peaks very similar to that of the intergranular region while we found essentially no ac loss experimentally. At present it is not certain whether the critical state model is still useful to explain grain behavior. More systematic studies are necessary to understand the property of the powder, and hopefully the superconducting grains.

### 4.5. Frequency Dependence of AC Magnetic Susceptibility

The dc resistivity of the superconductors is essentially zero unless the pinning force on flux lines is smaller than the Lorentz force acting on them. This is no longer true for ac resistivity. Superconductors were originally considered to have two electron fluids; the normal electrons and the superconducting electrons. At dc the superconducting electrons can completely short out the normal electrons so we have zero dc resistance. At ac the normal electrons carry some current because the inertial properties of the electrons cannot do short circuiting completely. In consequence, there appears ac loss.

It is well known that there are two types of ac loss in superconductors,\(^{25,26}\) hysteretic loss and flux-flow loss. When an ac magnetic field is applied, flux sweeps in and out of each cycle from the specimen and a layer of bulk current is induced analogous to the skin effect in a normal conductor. This is the hysteretic
loss. This hysteretic loss does not show any frequency dependence of $\chi$.

The flux-flow loss appears due to flux-flow resistivity. In an ideal type II superconductor, an electric field appears by a flow of the flux lines when the flux lines are no longer pinned. The vortex structure is approximated by an inner core that is normal surrounded by superconducting material. The ac loss then occurs as Joule heating in this essentially normal core region. Compared with the hysteretic loss, the flux-flow loss has a very strong frequency dependence.

Fig. 4.17 shows the measured complex susceptibility $\chi$ of sample #1 as a function of frequency $f$ of $H_1$ at $T = 77$ K. Here $H_1 = 0.3$ Oe and $H_0 = 0$. In order to cover the full frequency range (1 kHz – 13 MHz) three different coils listed in Table 3.3 were used to maximize the resolution of the HP impedance analyzer. Crossover frequencies of the pickup coils are 100 kHz and 1 MHz. Up to about 5 MHz $\chi'$ in particular is almost independent of frequency as shown in Fig. 4.17(a). Starting from ~ 5 MHz, $\chi'$ increases monotonically with increasing frequency which strongly suggests that the ac loss mechanism changes from hysteretic loss to flux-flow loss. In Fig. 4.17(b) $\chi''$ was found to rise slowly as the frequency increases up to 9 MHz. The data show above 9 MHz a decrease in $\chi''$ due to a decrease of $H_1$ supplied by the HP impedance analyzer; this region of Fig. 4.17(b) should be ignored. The dashed line in the figure is the extrapolated behavior at high frequency.

Experiments with superconducting Pb-In and Nb-Ta alloy foils\textsuperscript{27,28} have earlier shown that their flux-flow resistivity increases to that at their normal state at
radio-frequency $f \approx 10$ MHz even for the transport currents several orders of magnitude below the critical value. Fig. 4.17 may indicate the similar behavior. More accurate measurement on the frequency dependence of $\chi'$ and $\chi''$ over a wide range of frequency is necessary to get information on the dynamical behavior of flux motion.
REFERENCES. CHAPTER 4


FIGURE CAPTIONS

Fig. 4.1. (a)-(d): Measured complex ac magnetic susceptibility $\chi'$ and $\chi''$ (in MKS units) of a sintered polycrystalline cylindrical rod (sample #2) as a function of applied ac magnetic field $H_1$ or dc magnetic field $H_0$ at $T = 78.3$ K. Here $f = 10$ kHz and value of $H_1$ is the amplitude of the applied ac field. Resolution of the HP impedance analyzer for $H_1 = 0.03$ Oe is ten times lower than that for higher $H_1$. All values of $\chi'$ and $\chi''$ are discrete, giving the small steps in $\chi'$ and $\chi''$ at small values. (e)-(h): Calculated complex ac magnetic susceptibility by using the modified critical state model in Section 4.1.1 for sample #2 rod as a function of applied ac magnetic field $H_1$ or dc magnetic field $H_0$ at $T = 78.3$ K. See the text for detail of the calculations; parameters are given in Table 4.4.

Fig. 4.2. (a)-(d): Measured complex ac magnetic susceptibility of the sample #2 at $T = 82.3$ K. (e)-(h): Calculated susceptibility from the model; parameters from Table 4.4.

Fig. 4.3. (a)-(d): Measured complex ac magnetic susceptibility of the sample #2 at $T = 85.7$ K. (e)-(h): Calculated susceptibility from the model; parameters from Table 4.4.

Fig. 4.4. (a)-(d): Measured complex ac magnetic susceptibility of a cylindrical rod of sample #3 as a function of $H_0$ and $H_1$ at $T = 78.3$ K. Other parameters are same as in Fig. 4.1. (e)-(h): Calculated susceptibility from the model; parameters from Table 4.4. See the text for discussion of the fitting parameters of the model.
**Fig. 4.5.1.** Calculated complex ac magnetic susceptibility with a Kim-Anderson critical current form (Eq. 4.4) with $\beta_f = 1$. The value of fitting parameters are $R = 0.85$ mm, $H_f^* = 8.0$ Oe, $H_{sJ} = 2.5$ Oe, and $\mu_{eff} = 0.491$. Notice that large deviation between this calculation and the data (Figs. 4.1(a), (b), (c), (d)) for small $H_1$; A calculation using $\beta_f = 2.25$, Figs. 4.1(e), (f), (g), (h) fit the data much better.

**Fig. 4.5.2.** Calculated complex ac magnetic susceptibility with the exponential critical current form in Eq. 4.5. $R = 0.85$ mm, $H_f^* = 8.0$ Oe, $H_{sJ} = 2.5$ Oe, and $\mu_{eff} = 0.491$ are used for the calculation. Because of fast decrease of $J_c$ at large $H$, the saturation of $\chi'$ is quite noticeable in the measured range of $H$. Comparison to data (Figs. 4.1(a), (b), (c), (d)) shows poor agreement.

**Fig. 4.6.** Three different forms of the critical current density $J_{cJ}(H)$ are plotted as a function of $H$. (a) Kim-Anderson form in Eq. 4.5. For the fit of the data, $R = 0.85$ mm, $H_f^* = 8.0$ Oe, and $H_{sJ} = 2.5$ Oe are used. (b) Modified Kim-Anderson form in Eq. 4.1a. $R = 0.85$ mm, $H_f^* = 8.0$ Oe, $H_{sJ} = 3.0$ Oe, and $\beta_f = 2.25$ are used. (c) Exponential form. $R = 0.85$ mm, $H_f^* = 8.0$ Oe, and $H_{sJ} = 2.5$ Oe are used.

**Fig. 4.7.** (a)-(d): Measured complex ac magnetic susceptibility of the sample #2 as a function of temperature in the presence of applied dc and ac magnetic fields. The frequency of the ac magnetic field was 10 kHz. The sample began superconducting at 92.5 K ($T_c$). (e)-(h): Calculated temperature dependence of ac magnetic susceptibilities of the sample #2 with parameters given in Eq. 4.7. The details of the
calculation are given in Section 4.2.

**Fig. 4.8.** (a), (b): Measured temperature dependence of complex ac magnetic susceptibility of the sample #3. Because the sample has a low intergranular critical current density, $\chi'$ decreases very slowly for large $H_1$. Compare it with Fig. 4.7. (c), (d): Calculated temperature dependence of ac magnetic susceptibility of the sample #3 with parameters given in Eq. 4.8.

**Fig. 4.9** Hysteresis loop of ac magnetic susceptibility of a bulk cylindrical sample #1 (Colorado Superconductor, Inc.). $T = 77$ K and $H_1 = 0.22$ Oe, (a), (b): $|H_0^{\text{max}}| = |H_0^{\text{min}}|$; (c), (d): $|H_0^{\text{max}}| \neq |H_0^{\text{min}}|$.

**Fig. 4.10.** (a), (b): Change of $\chi$ after an initial application of $H_0$. Curve 1 is taken just after the cooling in zero dc field, starts at $H_0 = 0$, increasing to $H_0 = 100$ Oe. In curve 2, $H_0$ is then reduced from $H_0 = 100$ Oe to $H_0 = 0$, etc. The sample #1 was used at $T = 77$ K. $H_1 = 0.22$ Oe. (c), (d): Hysteresis curves calculated with the field dependence of the critical current density $J_c(H)$ in Fig. 4.13. The parameters used in the figure are given in Section 4.3.

**Fig. 4.11.** For $H_1 = 0.22$ Oe, magnetic hysteresis loops of $\chi'$ and $\chi''$ for the sample #2. (a), (b): $T = 78.3$ K; (c), (d): $T = 85.7$ K.

**Fig. 4.12.** Dependence of magnetic hysteresis loops on $H_1$. The sample #2 was used. Notice the difference of change of sense of rotation of $\chi'$ during $H_0$ sweep for $H_1 = 7.91$ and 0.22 Oe.
Fig. 4.13. Possible critical currents for fitting Fig. 4.12. $J_c$ is the total critical current, $J_c^{rev}$ the reversible critical current, $J_c^{irrev}$ the irreversible critical current. $J_c^{irrev}$ is the subtraction of $J_c$ for curve 1, which is also a $J_c^{rev}$, from $J_c$ for curve 2 in this figure. We used parameter values for Eq. 4.12 $\mu_{eff} = 0.52$, $(H_J^x, H_{sj}, \beta, H_m)$: (2.4 Oe, 4.0 Oe, 1.8, 0 Oe) for curve 1; (2.4 Oe, 2.0 Oe, 1.2, 10 Oe) for curve 2; and (2.4 Oe, 3.0 Oe, 1.7, 6 Oe) for curve 3.

Fig. 4.14. Measured complex ac magnetic susceptibility versus $H_0$ and $H_1$ of powder sample #4 (made by grinding sample #2). Because of grinding, most of the intergranular coupling between superconducting grains seems to be broken so the powder shows almost perfect diamagnetism of the grains. Still there is a slight trace of intergranular effect, which is a fast rise of $\chi'$ near $H_0 = 0$, for small $H_1$.

Fig. 4.15. (a), (b): Measured susceptibility of sample #4 as a function of $H_0$. This powdered sample shows a magnetic hysteresis loop. (c), (d): Calculated susceptibility of the sample. $H_g^* = 250$ Oe, $H_{sg} = 30$ Oe, and $\beta = 2.3$ are used for the calculation. The bottom curve is for $H_1 = 0.22$ Oe, the upper one for $H_1 = 7.91$ Oe.

Fig. 4.16. (a), (b): Measured susceptibility of the sample #4 as a function of $T$. (c), (d): The results of $T$ dependence of $\chi$ evaluated with the critical state model. Temperature dependence of $H_g^*$, $H_{sg}$, and $\beta_g$ is assumed as follows: $H_g^*(T) = 1469.3 \left[1-(T/T_c)^2\right]^{1.5}$, $H_{sg}(T) = 40.22 - 0.13T$, and $\beta(T) = 2.47 - 7.76 \times 10^{-3}T$.

Fig. 4.17. Measured complex ac magnetic susceptibilities of a cylindrical rod (sample #1) as a function of frequency $f$ of $H_1$. Here $H_1 = 0.3$ Oe and $H_0 = 0$. In
order to cover the full frequency range (1 kHz ~ 13 MHz) three different coils listed in Table 3.3 were used to maximize a resolution of the HP impedance analyzer. The changes in $\chi'$ and $\chi''$ at $f \approx 10^6$ Hz may mark the transition from hysteretic to flux-flow ac loss.
Figure 4.1
Figure 4.3
Figure 4.6

\( J_c (H) \) (A/cm\(^2\))

\( H \) (Oe)

35

800
Figure 4.7
Figure 4.7
Figure 4.8
Figure 4.11

(a) $T = 78.3$ K
$H_1 = 0.22$ Oe

(b) $T = 78.3$ K

(c) $T = 85.7$ K
$H_1 = 0.22$ Oe

(d) $T = 85.7$ K
Figure 4.12

\( T = 77 \, \text{K} \)

\( H_1 \, (\text{Oe}) = 7.91 \)

\( \chi' \)

\( H_0 \, (\text{Oe}) \)
Figure 4.12
Figure 4.13
Figure 4.14
Figure 4.15
Figure 4.16
Figure 4.17

(a) $H_0 = 0.0 \text{ Oe}$
$H_1 = 0.30 \text{ Oe}$

(b) $H_0 = 0.0 \text{ Oe}$
$H_1 = 0.30 \text{ Oe}$
CHAPTER 5. CONCLUSIONS

In this Chapter we will summarize the results found for the complex ac magnetic susceptibility of the granular YBCO superconductor in experiments and in calculations using a modified critical state model. Conclusions will be derived from the results, and suggestions for future experiments will be discussed in this chapter.

In our experiments, a complete and systematic study of the complex ac susceptibility, $\chi = \chi' - i\chi''$ is undertaken. We have measured the dependence on temperature $T$, ac magnetic field $H_1$, frequency $f$, and dc magnetic field $H_0$. The measurements of the two susceptibility components are performed with a computer-controlled impedance analyzer and are described fully in Chapter 2.

It is experimentally found for sintered cylindrical rods of YBCO that:

(1) Both $\chi'$ and $\chi''$ depend strongly on $H_0$ and $H_1$. $\chi'(H_0)$ increases monotonically as $H_0$ increases for small $H_1$. For larger $H_1$, $\chi'(H_0)$ has a minimum value at non-zero $H_0$. The ac loss $\chi''(H_0)$ for small $H_1$ is essentially zero at $H_0 = 0$ and has a maximum as $H_0$ increases. But for larger $H_1 \chi''(H_0)$ has a maximum at $H_0 = 0$. This loss is related to flux penetration into the sample. $\chi'(H_1)$ increases monotonically as $H_1$ increases, to a saturated value. $\chi''(H_1)$ for $H_0 = 0$ shows monotonic increase with $H_1$ and reaches a maximum at a value which can be shown to be the field $H^*$ for penetration to the center of the cylindrical sample.
(2) The ac susceptibility $\chi'$ and $\chi''$ shows strong temperature dependence. For small $H_1$, $\chi'$ rapidly decreases as $T$ goes below the transition temperature $T_c$ and approaches a perfect diamagnetic state as $T \to 77$ K. For larger $H_1$, $\chi'(T)$ shows clearly two different slopes. When the applied field is small, the loss $\chi''(T)$ is negligible except for a region near $T_c$, i.e., $\chi''$ has a sharp peak near $T_c$. When larger field, $H_0$ or $H_1$, is applied, the $\chi''$ peak is broad and has a maximum at a value which is dependent on the magnitude of $H_0$ and $H_1$.

(3) Both $\chi'(H_0)$ and $\chi''(H_0)$ show hysteresis when $H_0$ is swept cyclically up and down. When the maximum values of $H_0$ sweep is small enough, the hysteresis in $\chi'(H_0)$ and $\chi''(H_0)$ is negligibly small. If the maximum $H_0$ is increased further, a pronounced hysteresis loop is developed in both $\chi'$ and $\chi''$. We observe two behaviors in $\chi'(H_0)$: rapid increase at low field region ($H_0 \leq 40$ Oe), and very slow monotonic increase at higher field region ($H_0 \geq 100$ Oe). This behavior is shown to be related respectively to intergranular and intragranular region of the sample. Furthermore it is believed that irreversible flux pinning in the intragranular region is the cause of the large hysteresis observed in the ac susceptibility. The hysteresis is also dependent on temperature and sample preparation. The ac susceptibility is found to have dependence on magnetic history; for example, the position and the value of the minimum of $\chi'$ are dependent on the maximum value of scanning $H_0$.

(4) The ac susceptibility, $\chi'$ and $\chi''$, shows a very small dependence ($\leq 3\%$) on
frequency below $f = 10^6$ Hz. Above this frequency both $\chi'$ and $\chi''$ increase monotonically; this may indicate that the ac loss mechanism changes from hysteretic loss to flux-flow loss.

(5) The powder made by grinding the bulk sintered sample used for the above measurements shows a much simpler dependence of $\chi'$ and $\chi''$ on magnetic fields and temperature. The losses $\chi''(H_0)$ and $\chi''(H_1)$ are unmeasurably small and $\chi'$ increases very slowly with $H_0$ or $H_1$. This simplicity is believed to come from lack of the intergranular weak links because the powder size is comparable to the grain size.

From calculations using the modified critical state model described in Section 4.1.1 and Appendix A in detail, it is found that:

(1) Much of the data, complex $\chi(H_0)$ and $\chi(H_1)$, can be understood by the modified critical state model. The critical current density $J_c(H)$ in the original critical state model\textsuperscript{1-3} is modified by the expression in Eq. 4.1. At low temperatures (eg., 77 K) the model calculations using only an intergranular component (which has a small critical current density $J_c \approx 600 \text{ A/cm}^2$), are quantitatively and qualitatively in excellent agreement with the data. $J_c$ of the intergranular region is found to have a steeper field dependence, $\beta \approx 2$, than that originally assumed by Bean ($\beta = 0$) and by Kim-Anderson ($\beta = 1$) for low-temperature type II superconductors.

(2) Our calculations using empirical expressions for the temperature dependence
of relevant fitting parameters of the modified critical state model can explain qualitatively the measured temperature dependence of the observed ac susceptibility \( \chi' \) and \( \chi'' \). It is shown that two regions in \( \chi'(T) \) are due to flux motion in the intragranular region and in the outer surface of each grain of thickness \( \lambda_g(T) \), the temperature-dependent penetration depth of the grain.

(3) The dependence of \( \chi' \) and \( \chi'' \) on magnetic history can be qualitatively understood by introducing an irreversible current given by Eq. 4.12 when a maximum value of scanning \( H_0 \) is sufficiently large. The total critical current density has a maximum at a nonzero \( H_0 \) which is dependent on the magnetic history. It is believed that a recent model proposed by Campbell provides a microscopic picture of this phenomena.

(4) The hysteresis of \( \chi' \) and \( \chi'' \) shown for large \( H_0 \) sweep can be understood by a transition from the intergranular to intragranular behavior. Two regions are characterized by different magnitudes of the critical current density. It is estimated from the data that the critical current density of the intergranular region is approximately \( 10^6 \) times smaller than that of the intragranular region. This huge difference in \( J_c \) explains the rapid increase of \( \chi'(H_0) \) in the intergranular region at small \( H_0 \) up to a saturated value, and then a very slow monotonic increase of \( \chi' \) up to about \( 10^3 \) Oe. The small value of \( J_c \) of the intergranular region is the cause of the strong ac and dc field dependence of both \( \chi' \) and \( \chi'' \).

(5) Although the modified critical state model can explain correctly the observed weak dependence of the ac susceptibility of the powder if we assume that each
powder is a single superconducting grain, or an ensemble of a few grains with very weak coupling, details from the calculation are not in good agreement with the data; The calculation for \( \chi' \) or \( \chi'' \) shows a sharp transition for small \( H_1 \) but the data show a much broader transition.

It is a very complicated problem to understand the electrodynamics of high-temperature granular superconductors. The above study should be extended to other high-temperature superconductors which are believed to have also two different components: intergranular and intragranular. Similarities and differences of their electromagnetic behaviors should be clarified. The data presented in this thesis strongly suggest that bulk YBCO samples have these two components. It is found that the intergranular weak links are well described by the modified critical state model. Poor agreement of the ac susceptibility of the powder between the data and the calculation is not still understood. Anisotropy of the grains makes the modelling difficult; the demagnetizing field which is sensitive to the geometry of the grain can be another factor of difficulty. The intragranular behavior should be studied further at very high dc \( H_0 \) or ac field \( H_1 (\geq 10^3 \text{ Oe}) \) where the intragranular behavior becomes dominant.

Finally it should be mentioned that the systematic measurement of the ac susceptibility, \( \chi'(H_0) \), \( \chi''(H_0) \), \( \chi'(H_1) \), and \( \chi''(H_1) \), of the sample with well-defined geometry can determine the field dependence of the induced critical current density which is very important to the understanding of the electrodynamics of the superconductor. The \( H_0 \) and \( H_1 \) field dependence of the ac susceptibility is
directly predicted from the critical state model \textit{when} the field dependence of the critical current density is known. Since calculated susceptibility is quite sensitive to the form of the critical current density, detailed ac susceptibility measurements may offer one of best empirical methods for determining the magnetic field dependence of the critical current density for a number of high temperature superconductors.
REFERENCES. CHAPTER 5

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APPENDIX A. SUMMARY OF STEPS USED IN MODIFIED CRITICAL STATE MODEL

In this appendix steps of calculation of the modified critical state model in Section 4.1.1 is summarized.

\textit{Step 1}: Choose parameter values: $R$, $H^*$, $H_s$, and $\beta$ for intergranular and intragranular region. $R$ is the radius of a cylindrical sample for the intergranular region, which can be measured, and a radius $R_g$ of the grain for the intragranular region, which should be assumed. $H^*$ can be estimated from the experimental $\chi''(H_1)$ curve because it is approximately the same as the value of $H_1$ at the peak at $H_0 = 0$. If $H_1$ is scanned up to $10^3$ Oe, there are two $\chi''$ peaks; the one at small $H_1$ is the intergranular $H^*_f$, the other is the intragranular $H^*_g$. The other two parameters, $H_s$ and $\beta$, are arbitrarily chosen initially in the calculation, and later adjusted for best fit to the data. Typical values are: $H_s \approx 2$ to 10 Oe; $\beta \approx 1$ to 3.

\textit{Step 2}: With the parameter values chosen for the calculation, the pinning force density $\alpha$ given by Eq. 4.13 is then calculated.

\textit{Step 3}: Following the procedure in Appendix B the local field $H_l(r,t)$ is numerically calculated at given applied field $H(t) = H_0 + H_1 \cos \omega t$. This is a main step in applying the modified critical state model. $H_l(r,t)$ is calculated separately for the inter- and the intragranular region because of the difference of the parameters.

\textit{Step 4}: The average magnetic induction $B(t)$ for each region is obtained by spatial integration over the whole sample (the intergranular region) or a single grain (the
intragranular region). In this step the effective permeability $\mu_{\text{eff}}$ defined by Eq. 4.3a for the intergranular region is used for the calculation of $B(t)$. For the intragranular region, the similar definition of the effective permeability $\mu_g$ can be used and is discussed in Ref. 11 in Section 4.

**Step 5:** The ac magnetic susceptibility $\chi'$ and $\chi''$ is calculated using definitions given by Eq. 2.17a, 2.17b. In this step the contribution from both components, inter- and intragranular, is naturally included. But at small fields we can reasonably assume the property of the intragranular region: $\chi' = -1$ and $\chi'' = 0$; so in low fields the calculation can be done only for the intergranular region. This is what is usually done in this thesis.

**Step 6:** For temperature dependence of the ac susceptibility, the temperature dependence of the relevant parameters should be assumed. There is no systematic way to choose this; one can be guided roughly by the standard assumptions for low temperature superconductors. In this thesis we derive the dependence empirically from the parameter values obtained from the fitting of $\chi'(H)$ and $\chi''(H)$ for several temperatures. Using assumption of the temperature dependence of the parameters the steps from 3 to 5 are repeated at each temperature and it produces the temperature dependence of the calculated ac susceptibility.
APPENDIX B. COMPUTER PROGRAMS FOR THE CRITICAL STATE MODEL

The critical state model (Eq. 2.15 and 2.16) is solved analytically for a long cylindrical specimen with a radius $R$ in the presence of an applied magnetic field $H(t) = H_0 + H_1 \cos \omega t$ given as Eq. 2.14. The magnetic field is parallel to the sample axis. The solutions of the model are local magnetic fields inside the sample and are dependent on values of $H_0$, $H_1$, radius $R$, and time $t$, and whether the field is increasing and decreasing with time.

(1) $H_0 \leq H_1$

In this case, a cycle is divided into five intervals; (a), ..., (e).

(a) $0 \leq t \leq t_1$, where $H(t_1) = 0$

Fig. A.1 shows the local field $H_t(r)$ inside the sample, i.e. the applied field plus the field generated by the shielding current.

$$H_t(r) = \left[ (H_0+H_1+H_s)^{1+\beta} - (1+\beta)4\pi\alpha(R-r) \right]^{\frac{1}{1+\beta}} - H_s \ (r \leq r_t),$$  \hspace{1cm} (A.1)

$$H_t(r) = \left[ (H_0+H_1\cos(\omega t)+H_s)^{1+\beta} + (1+\beta)4\pi\alpha(R-r) \right]^{\frac{1}{1+\beta}} - H_s \ (r > r_t),$$  \hspace{1cm} (A.2)

where $\omega = 2\pi f$, and $f$ is the frequency of the ac magnetic field and

$$r_t = R - \frac{1}{8\pi\alpha(1+\beta)} \left[ (H_0+H_1+H_s)^{1+\beta} - (H_0+H_1\cos(\omega t)+H_s)^{1+\beta} \right]$$  \hspace{1cm} (A.3)

The maximum penetration depth $r_0$ is given by

$$r_0 = R - \frac{1}{4\pi\alpha(1+\beta)} \left[ (H_0+H_1+H_s)^{1+\beta} - H_s^{1+\beta} \right].$$  \hspace{1cm} (A.4)
(b) \( t_1 \leq t \leq T/2 \), where \( T = 1/f \)

Fig. A.2 shows the local field \( H_I(r) \) inside the sample. \( H_I(r) \) for \( r \leq r_t \) is the same as in (a).

\[
H_I(r) = \left[ H_s^{1+\beta} - (1+\beta)4\pi\alpha (r-r_t) \right] \frac{1}{1+\beta} - H_s \quad (r_t < r \leq r_r) \tag{A.5}
\]

\[
H_I(r) = H_s - \left[ (-H_0-H_1 \cos(\omega t)+H_s)^{1+\beta} - (1+\beta)4\pi\alpha (R-r) \right] \frac{1}{1+\beta} \quad (r > r_r) \tag{A.6}
\]

where

\[
r_r = R - \frac{1}{4\pi\alpha(1+\beta)} \left[ (-H_0-H_1 \cos(\omega t)+H_s)^{1+\beta} - H_s^{1+\beta} \right] \tag{A.7}
\]

and

\[
r_t = \frac{1}{2}(R+r_r) - \frac{1}{8\pi\alpha(1+\beta)} \left[ (H_0+H_1+H_s)^{1+\beta} - H_s^{1+\beta} \right]. \tag{A.8}
\]

(c) \( T/2 \leq t \leq t_2 \), where \( H(t_2) = 0 \)

Fig. A.3 shows the local field \( H_I(r) \) inside the sample. \( H_I(r) \) for \( r \leq r_1 \) is the same as in (a).

\[
H_I(r) = \left[ H_s^{1+\beta} + (1+\beta)4\pi\alpha (r_r-r) \right] \frac{1}{1+\beta} - H_s \quad (r_1 < r \leq r_r) \tag{A.9}
\]

\[
H_I(r) = H_s - \left[ H_s^{1+\beta} + (1+\beta)4\pi\alpha (r-r_r) \right] \frac{1}{1+\beta} \quad (r_r < r \leq r_t) \tag{A.10}
\]

\[
H_I(r) = H_s - \left[ (-H_0-H_1 \cos(\omega t)+H_s)^{1+\beta} + (1+\beta)4\pi\alpha (R-r) \right] \frac{1}{1+\beta} \quad (r > r_t) \tag{A.11}
\]

where

\[
r_r = R - \frac{1}{4\pi\alpha(1+\beta)} \left[ (-H_0+H_1+H_s)^{1+\beta} - H_s^{1+\beta} \right] , \tag{A.12}
\]

\[
r_t = \frac{1}{2}(R+r_r) + \frac{1}{8\pi\alpha(1+\beta)} \left[ (-H_0-H_1 \cos(\omega t)+H_s)^{1+\beta} - H_s^{1+\beta} \right]. \tag{A.13}
\]

and
\[ r_1 = \frac{1}{2}(R + r_r) - \frac{1}{8\pi\alpha(1+\beta)} \left[ H_0 + H_1 + H_s \right]^{1+\beta} - H_s^{1+\beta}. \] 

(A.14)

(d) \( t_2 \leq t \leq t_3 \), where \( t_3 = 1 - \cos^{-1}(1-2H_0/H_1)/\omega \)

Fig. A.4 shows the local field \( H_l(r) \) inside the sample. \( H_l(r) \) for \( r \leq r_1 \) is the same as in (c). \( r_1 \) and \( r_r \) are the same as in (c).

\[ H_l(r) = H_s - \left[ H_s^{1+\beta} + (1+\beta)4\pi\alpha(r - r_r) \right]^{1+\beta} \quad (r_r < r \leq r_{r1}) \] 

(A.15)

\[ H_l(r) = \left[ (H_0 + H_1 \cos(\omega \tau) + H_s)^{1+\beta} - (1+\beta)4\pi\alpha(R - r) \right]^{1+\beta} - H_s(r > r_{r1}) \] 

(A.16)

where

\[ r_r = \frac{1}{2}(R + r_{r1}) + \frac{1}{8\pi\alpha(1+\beta)} \left[ H_s^{1+\beta} - (H_0 + H_1 \cos(\omega \tau) + H_s)^{1+\beta} \right]. \]

(A.17)

and

\[ r_{r1} = R - \frac{1}{4\pi\alpha(1+\beta)} \left[ (H_0 + H_1 \cos(\omega \tau) + H_s)^{1+\beta} - H_s^{1+\beta} \right]. \]

(A.18)

(e) \( t_3 \leq t \leq T \)

Fig. A.5 shows the local field \( H_l(r) \) inside the sample. \( H_l(r) \) for \( r \leq r_t \) is the same as in (d). \( r_1 \) and \( r_r \) are the same as in (c).

\[ H_l(r) = \left[ (H_0 + H_1 \cos(\omega \tau) + H_s)^{1+\beta} - (1+\beta)4\pi\alpha(R - r) \right]^{1+\beta} - H_s(r > r_t) \] 

(A.19)

where

\[ r_t = \frac{1}{2}(R + r_r) + \frac{1}{8\pi\alpha(1+\beta)} \left[ H_s^{1+\beta} - (H_0 + H_1 \cos(\omega \tau) + H_s)^{1+\beta} \right]. \]

(A.20)

(2) \( H_0 > H_1 \)

In this case, the cycle is divided into two intervals; (a), (b).

(a) \( 0 \leq t \leq T/2 \)
The local field \( H_t(r) \) looks exactly the same as Fig. A.1 and Eqs. A.1 to A.4 describe the local field \( H_t(r) \).

(b) \( T/2 \leq t \leq T \)

The local field \( H_t(r) \) looks exactly the same as Fig. A.5 and equations in (e) describe the local fields.

The following FORTRAN programs calculate ac magnetic susceptibility as a function of an applied magnetic field by using a time series of the local field \( H_t(r,t) \) described above. These programs were written in FORTRAN 77 and compiled with a SUN 3.0 FORTRAN compiler.

**Program 1. chihdc**

This program gives values of ac magnetic susceptibility coming from intergranular or intragranular material as a function of \( H_0 \). \( H_1 \) is given for the calculation. There are four parameters for fitting experimentally measured data: \( H_s, H^*, \mu, \) and \( \beta. \)

c
dimension fldh(550)
dimension depth(550),t(401),phi(401)
dimension rv(105),cv(105),h(105)
character filename*20,sample*10
    ,pi=2.*asin(1.)
c
    c initial setting of parameters
c
    write(6,1)
1  format(1x,'data filename ?')
read*,filename
write(6,2)
2  format(1x,'max Hdc,Hac,Ha*,H0,beta ')
read*,dchmx,acfldh,ptfldh,spfldh,beta
dchmin=0.
dc=100
delh=dchmx/float(ndc)

c    R is a radius of a cylindrical sample in cm.
R=0.085
write(6,3)

3 format(1x, 'fraction of intergrain : ', $)
read*, vmu
write(6,4)

4 format(1x, 'sample : ', $)
read*, sample
acftdh=acftdh
ptftdh=ptftdh
spftdh=spftdh
f=1.

H field profiles when H1 is at a fldhmax (maximum field value)

find alpha (pinning force density)
alpha=((ptftdh+spftdh)**(1.+beta)-spftdh**(1.+beta)) /
6format(1x, 'Hac=", f6.2, '0e, Ha*=", f6.2, '0e, H0=", f6.2, 
'0e, mhu=" , f4.2, 
/, lx', R=", f6.3, ' cm, pinning density=", e8.2, ' G0e/cm2', 
', cv=", f6.3,f4.1)

smni=le30
smxi=-le30
smnr=le30
smxr=-le30
do 11 j=1,ndc+1
dcfldh=dchmin+delh*float(j-1)
h(j)=dcfldh
fldhmax=acftdh+dcfldh

maximum penetration depth
r0=R-((fldhmax+spftdh)**(1.+beta)-spftdh**(1.+beta)) /
/a*(1.+beta)
if(r0.lt.0.) r0=0.
if(dcfldh.ge.acftdh) then
r2=R-((dcfldh-acftdh+spftdh)**(1.+beta)-(fldhmax+spftdh) **(1.+beta)) /(2.*a*(1.+beta))
else
r2=R-((dcfldh+acftdh+spftdh)**(1.+beta)+(fldhmax+spftdh) **(1.+beta)-2. *spftdh**(1.+beta)) /
endif
if(r2.lt.0.) r2=0.
write(6,7)j,h(j),r0/R,r2/R

7 format(1x,i3, ': Hdc = ', f6.2,'G, r0 = ',f6.4,' cm, pinning density=’, e8.2, ' G0e/cm2', 
r2 = ',f6.4)
nd=100
delr=(R-r2)/float(nd)
time=50

do 10 k=1,ntime
t(k)=(float(k-1)+0.5)/float(ntime)
apfl=(dcpf+acpf*cos(2.*pi*t(k))
phi(k)=0.
if((dcpf.ge.acpf)) go to 12

t1=acos(-1.*dcpf/acpf)/(2.*pi)
t2=1.-t1
t3=1.-acos(1.-2.*dcpf/acpf)/(2.*pi)
if((t(k).ge.0.0)) go to 13
if((t(k).le.0.5)) go to 14
if((t(k).le.0.5)) go to 15
if((t(k).le.0.5)) go to 16
if((t(k).le.0.5)) go to 17
if((t(k).le.0.5)) go to 18

rt=R+((apfl+spfl)**(1.+beta)-(dcpf+spfl)**(1.+beta))
os(1.+beta))/(2.*a*(1.+beta))
if(rt.lt.0.) rt=0.
do 20 i=1,nd+1
depth(i)=(r2+float(i-1)*delr)/R
if(depth(i).lt.t/R) then
fdh(i)=((1.+beta)*a*R*(depth(i)-1.)+(dcpf+spfl)**(1.+beta)**(1./(1.+beta))-spfl
else
fdh(i)=(-(1.+beta)*a*R*(depth(i)-1.)+(apfl+spfl)**(1.+beta)**(1./(1.+beta))-spfl
endif
phi(k)=phi(k)+2.*pi*delr*fdh(i)*depth(i)*R
20 continue
go to 10

c

rr=R+((dcpf-dacf+spfl)**(1.+beta)-(apfl+spfl)**(1.+beta))
os(1.+beta))/(2.*a*(1.+beta))
if(rr.lt.0.) rr=0.
do 21 i=1,nd+1
depth(i)=(r2+float(i-1)*delr)/R
if(depth(i).lt.t/R) then
fdh(i)=((1.+beta)*a*R*(depth(i)-1.)+(dcpf-dacf+spfl)**(1.+beta)**(1./(1.+beta))-spfl
else
fdh(i)=((1.+beta)*a*R*(depth(i)-1.)+(apfl+spfl)**(1.+beta)**(1./(1.+beta))-spfl
endif
phi(k)=phi(k)+2.*pi*delr*fdh(i)*depth(i)*R
21 continue
go to 10

c

rr=R-((-apfl+spfl)**(1.+beta)-(spfl)**(1.+beta))
\[ \begin{align*}
rt &= 0.5 \times (R + rr) - ((\text{fldhmax} + \text{spfldh})^2) - \text{spfldh}^2 \\
\text{spfldh} &= \left( \frac{1}{2 \times (1 + \beta) \times a} \right) \\
& \text{if } (rt < 0.5) \text{ then } rt = 0.5 \\
& \text{if } (rr < 0.5) \text{ then } rt = 0.5 \\
& \text{do } 22 \text{ i=1,nd+1} \\
& \text{depth}(i) = (r2 + \text{float}(i - 1) \times \text{delr}) / R \\
& \text{if } (\text{depth}(i) \leq rt/R) \text{ then } \\
& \text{fldh}(i) = \left( (1 + \beta) \times a \times R \times \frac{\text{depth}(i) - 1}{1 + \beta} \right) \\
& \text{else} \\
& \text{if } (\text{depth}(i) > rt/R) \text{ then } \\
& \text{fldh}(i) = \left( (1 + \beta) \times a \times (rr - R \times \text{depth}(i)) + \text{spfldh} \right) \\
& \text{else} \\
& \text{phi}(k) = \text{phi}(k) + 2 \times \pi \times \text{delr} \times \text{fldh}(i) \times \text{depth}(i) \times R \\
& \text{22 continue} \\
& \text{go to 10} \\
\end{align*} \]
if(rt.lt.0.) rt=0.
if(rr1.lt.0.) rr1=0.
do 24 i=1,nd+1
  depth(i)=(r2+float(i-1)*dclr)/R
  if(depth(i).lt.rt/R) then
    fldh(i)=((1.+beta)*a*(rr-R*depth(i))+spfldh**(1.+beta))**((1./(1.+beta)) - spfldh)
  else
    if(depth(i).lt.rt/R) then
      fldh(i)=spfldh-((1.+beta)*a*R*(depth(i)-1.)+(dclfdh+acclfdh)*
                       +spfldh)**((1.+beta))**(1./(1.+beta))
    else
      if(depth(i).lt.r1/R) then
        fldh(i)=spfldh-((1.+beta)*a*R*(depth(i)-1.)+(apfld+spfldh)*
                         *(1.+beta))**(1./(1.+beta))
      endif
      endif
    endif
  else
    phi(k)=phi(k)+2.*pi*delr*fldh(i)*depth(i)*R
  endif
  phi(k)=phi(k)+2.*pi*delr*fldh(i)*depth(i)*R
  do 25 i=1,nd+1
    depth(i)=(r2+float(i-1)*dclr)/R
    if(depth(i).lt.rt/R) then
      fldh(i)=((1.+beta)*a*(rr-R*depth(i))+spfldh**(1.+beta))**((1./(1.+beta)) - spfldh)
    else
      if(depth(i).lt.rt/R) then
        fldh(i)=spfldh-((1.+beta)*a*R*(depth(i)-1.)+(dclfdh+acclfdh)*
                         +spfldh)**((1.+beta))**(1./(1.+beta))
      else
        if(depth(i).lt.r1/R) then
          fldh(i)=spfldh-((1.+beta)*a*R*(depth(i)-1.)+(apfld+spfldh)*
                           *(1.+beta))**(1./(1.+beta))
        endif
        endif
      endif
    else
      phi(k)=phi(k)+2.*pi*delr*fldh(i)*depth(i)*R
    endif
  25 continue
  go to 10

! calculate real or imaginary component of ac susceptibility
rv(j)=0.
cv(j)=0.
do 27 l=1,ntime
  rv(j)=rv(j)+vmu*phi(l)*sin(2.*pi*f*t(l))/float(ntime)
  cv(j)=cv(j)+vmu*phi(l)*cos(2.*pi*f*t(l))/float(ntime)
27 continue
rv(j)=2.*rv(j)/acclfdh/R**2/pi
cv(j)=2.*cv(j)/acclfdh/R**2/pi
cv(j)=cv(j)-1.
write(2,*) h(j),cv(j),rv(j)
if(rv(j).gt.smxi) smxi=rv(j)
if(rv(j).lt.smrni) smrn=rv(j)
if(cv(j).gt.smxr) smxr=cv(j)
if(cv(j).lt.smnri) smnri=cv(j)
print*, 'Xr = ',cv(j),', Xi = ',rv(j)
11 continue
  close(unit=2)
print*, 'real signal range : ',smnri,' - ',smxr
print*, 'imag signal range : ',smnri,' - ',smxi
end
Program 2. chihac

This program gives values of ac magnetic susceptibility coming either from intergranular or intragranular material, depending on input parameters, as a function of $H_1$. $H_0$ is given for the calculation.

c
dimension fldh(550)
dimension depth(550),t(401),phi(401)
dimension rv(105),cv(105),h(105)
character filename*20,sample*10
data pi/3.141592/
c
c initial setting of parameters
c
write(6,1)
1 format(1x,'data filename ?')
read*,filename
c
c input parameters
c
write(6,2)
2 format(1x,'Hdc, max Hac, Ha*,H0,beta')
read*,dcfldh,achmx,ptfldh,spfldh,beta
acv=1.
nac=40
R=0.085
write(6,3)
3 format(1x,'fraction of intergrain : ','$)
read*, vmu
write(6,4)
4 format(1x,'sample : ','$)
read*,sample
achmx=achmx*acv
ptfldh=ptfldh*acv
spfldh=spfldh*acv
delh=achmx/float(nac)
f=1.
c
H field profiles when H1 is at a fldhmax (maximum field value)
c
find alpha (pinning force density)
alpha=((ptfldh+spfldh)**(1.+beta)-spfldh**(1.+beta))/
 x    (4.*(1.+beta)*pi*R)
print*, 'pinning force density = ',alpha
a=4.*pi*alpha
c
H field profiles
open(unit=2,file=filename,status='new')
write(2,*) sample
write(2,6) dcfldh,pftfldh,spfldh,ymu,R,alpha,acv,beta
6 format(1x,'Hdc=' ,f6.2,' Oe, Ha*=' ,f6.2,' Oe, H0= ' ,f6.2,
x ' Oe, mhu= ' ,f4.2,
 x ,'/,x,' R= ',f6.3,' cm, pinning density= ',e8.2,' GOe/cm2'
x ,'; cv= ',f6.3,f4.1)
c
smni=1e30
smax=1e30
smnr=1e30
smax=1e30
do 11 j=1,nac
acfldh=delh*float(j)
h(j)=acfldh
fldhmax=acfldh+dcfldh
c maximum penetration depth
r0=R-((fldhmax+spfldh)**(1.+beta)-spfldh**(1.+beta))
x /a*(1.+beta)
if(r0.lt.0.) r0=0.
if(dcfldh.ge.acfldh) then
r2=R+((dcfldh-acfldh+spfldh)**(1.+beta)-(fldhmax+spfldh)
x **(1.+beta))/(2.*a*(1.+beta))
else
r2=R-((-dcfldh-acfldh+spfldh)**(1.+beta)+(fldhmax+spfldh)
x **(1.+beta)-2.*spfldh**(1.+beta))/(2.*a*(1.+beta))
endif
if(r2.lt.0.) r2=0.
write(6,12) j,h(j),r0/R,r2/R
12 format(1x,i3,': Ha = ',f6.2,' G, r0 = ',f6.4,', r2 = ',f6.4)
end

nd=100
delr=(R-r2)/float(nd)
time=50
do 10 k=1,ntime
t(k)=(float(k-1)+0.5)/float(ntime)
apfld=dcfldh+acfldh*cos(2.*pi*t(k))
phi(k)=0.
if(dcfldh.ge.acfldh) go to 13
if((t(k).ge.O. and.t(k).lt.t1) go to 14
if((t(k).ge.t1.and.t(k).le.0.5) go to 15
if((t(k).gt.0.5.and.t(k).le.1.5) go to 16
if((t(k).gt.1.5 and.t(k).le.2.5) go to 17
if((t(k).gt.2.5) go to 18
13 if((t(k).ge.O. and.t(k).lt.0.5) go to 14
if((t(k).ge.0.5) go to 19

Ha is decreasing from Ha,max to Ha,min

c
rt=R+((apfld+spfldh)**(1.+beta)-(fldhmax+spfldh)**
x (1.+beta))/(2.*a*(1.+beta))
if(rt.lt.0.) rt=0.
do 21 i=1,nd+1
    depth(i)=(r2+float(i-1)*delt)/R
    if(depth(i)>R) then
        fldh(i)=((1.+beta)*a*R*(depth(i)-1.)+(fldhmax+spfldh)**(1.+beta))**(1./(1.+beta)))-spfldh
    else
        fldh(i)=(-(1.+beta)*a*R*(depth(i)-1.)+(apfld+spfldh)**(1.+beta))**(1./(1.+beta)))-spfldh
    endif
    phi(k)=phi(k)+2.*pi*vmu*delt*fldh(i)*depth(i)*R
21 continue
    go to 10

19 rr=R+((dcfldh-acfldh+spfldh)**(1.+beta)-(apfld+spfldh)**(1.+beta))**(1./(1.+beta))-spfldh
    x**(1.+beta))**(1.+beta)-spfldh
    if(rr>0.) then
        rr=rr
        do 22 i=1,nd+1
            depth(i)=(r2+float(i-1)*delt)/R
            if(depth(i)>R) then
                fldh(i)=((1.+beta)*a*R*(depth(i)-1.)+(dcfldh-acfldh+spfldh)**(1.+beta))**(1./(1.+beta)))-spfldh
                x**(1.+beta))**(1./(1.+beta))-spfldh
            else
                fldh(i)=(-(1.+beta)*a*R*(depth(i)-1.)+(apfld+spfldh)**(1.+beta))**(1./(1.+beta)))-spfldh
            endif
        phi(k)=phi(k)+2.*pi*vmu*delt*fldh(i)*depth(i)*R
22 continue
    go to 10

15 rr=R-((-apfld+spfldh)**(1.+beta)-spfldh)**(1.+beta)*a
    x**(1.+beta))/2.*(1.+beta)*a)
    rt=0.5*(R+rr)-((fldhmax+spfldh)**(1.+beta)-spfldh)**(1.+beta))**(1./(1.+beta))-spfldh
    x**(1.+beta))**(1./(1.+beta))-spfldh
    if(rt>0.) then
        rt=rt
        do 23 i=1,nd+1
            depth(i)=(r2+float(i-1)*delt)/R
            if(depth(i)>R) then
                fldh(i)=((1.+beta)*a*R*(depth(i)-1.)+(fldhmax+spfldh)**(1.+beta))**(1./(1.+beta)))-spfldh
                x**(1.+beta))**(1./(1.+beta))-spfldh
            else
                fldh(i)=spfldh-((1.+beta)*a*(rr-R*depth(i))+spfldh)**(1.+beta))**(1./(1.+beta)))-spfldh
                x**(1.+beta))**(1./(1.+beta))-spfldh
            endif
            endif
        phi(k)=phi(k)+2.*pi*vmu*delt*fldh(i)*depth(i)*R
23 continue
    go to 10

16 rr=R-((-dcfldh-acfldh+spfldh)**(1.+beta)-spfldh)**(1.+beta))
x 1/(1.+beta)*a
rt=R+((-apfld+spfldh)**(1.+beta)-(-dcfldh+acfldh+spfldh)**(1.+beta))/(2.*(1.+beta)*a)

if(rr.lt.0.) rr=0.
if(rt.lt.0.) rt=0.
do 24 i=1,nl+1
depth(i)=(r2+float(i-1)*delr)/R
if(depth(i).lt.r/R) then
fldh(i)=((1.+beta)*a*(rr-R*depth(i))+spfldh**(1.+beta))
else
fldh(i)=spfldh**((1.+beta))**(1./(1.+beta))
endif
endif
phi(k)=phi(k)+2.*pi*vmu*delr*fldh(i)*depth(i)*R
24 continue
go to 10

17 rr=R-((-apfld+spfldh)**(1.+beta)-(-dcfldh+acfldh+spfldh)**(1.+beta))/((1.+beta)*a)
rrl=0.5*(rr+R)+(spfldh**(1.+beta)-(-dcfldh+acfldh+spfldh)**(1.+beta))/((1.+beta)*a)
rt=0.5*(rrl+R)+(spfldh**(1.+beta)-(-dcfldh+acfldh+spfldh)**(1.+beta))/((1.+beta)*2.*a)
if(rr.lt.0.) rr=0.
if(rt.lt.0.) rt=0.
if(rr1.lt.0.) rr1=0.
do 25 i=1,nl+1
depth(i)=(r2+float(i-1)*delr)/R
if(depth(i).lt.r/R) then
fldh(i)=spfldh**((1.+beta))**(1./(1.+beta))
else
fldh(i)=spfldh*((1.+beta)*a*R*(depth(i)-1.)+(-dcfldh+acfldh+spfldh)**(1.+beta))
endif
endif
phi(k)=phi(k)+2.*pi*vmu*delr*fldh(i)*depth(i)*R
25 continue
go to 10
\[
rr = R - \frac{((dcfldh + acfldh + spfldh)**(1 + \beta) - spfldh**(1 + \beta))}{(1 + \beta)^a}
\]

if (rr < 0.0) \( rr = 0.0 \)

\[
rt = 0.5 * (rr + R) + (spfldh**(1 + \beta) - (apfld + spfldh)**(1 + \beta))
\]

if (rt < 0.0) \( rt = 0.0 \)

\[
do 26 i = 1, nd + 1
depth(i) = (r2 + float(i - 1) * delr) / R
if (depth(i) > rt / R) then
fldh(i) = \( (1 + \beta) * a * (rr - R * depth(i)) + spfldh**(1 + \beta) \)
else
fldh(i) = -spfldh + \( (1 + \beta) * a * R * depth(i) - 1.0 + (apfld + spfldh)**(1 + \beta) \)**(1 / (1 + \beta))
endif
phi(k) = phi(k) + 2. * pi * vmu * delr * fldh(i) * depth(i) * R
26 continue
10 continue

calculate real or imaginary component of ac susceptibility

c rv(j) = 0.
c cv(j) = 0.
do 27 l = 1, ntime
rv(j) = rv(j) + phi(l) * sin(2 * pi * f * t(l)) / float(ntime)
cv(j) = cv(j) + phi(l) * cos(2 * pi * f * t(l)) / float(ntime)
27 continue
rv(j) = 2 * rv(j) / (pi * R**2)
cv(j) = 2 * cv(j) / (pi * R**2)
cv(j) = cv(j) - 1.
write(2,*) h(j), cv(j), rv(j)
if (rv(j) > smxi) smxi = rv(j)
if (rv(j) < smni) smni = rv(j)
if (cv(j) > smxr) smxr = cv(j)
if (cv(j) < smnr) smnr = cv(j)
print*, 'Xr (j)' = cv(j), 'Xr (j)' = rv(j)
11 continue
close(unit=2)
print*, 'real signal range : ', smnr, ' - ', smxr
print*, 'imaginary signal range : ', smni, ' - ', smxi
end
Program 3. chiThac

This program calculates ac magnetic susceptibility of intergranular materials with assumption that a cylindrical sample consists of two components; intergranular and intragranular materials. The details of the geometry of the sample is explained in Chapter 4.

c It calculates real and imaginary part of ac magnetic susceptibility of intergranular materials numerically as function of T at Hac. Jc has a form of \( \frac{\alpha}{(H+H_0)^\beta} \).

dimension fldh(550),temp(121)
dimension depth(550),t(401),phi(401)
dimension rv(125),cv(125)
character filename*20,sample*10

initial setting of parameters

\( \pi = 2. * \arcsin(1.) \)

temperature dependence of parameters

\( H_a(T) = a(1-(T/T_c)^2)^{1.5} \)
\( H_0(T) = c + d(T-78), \ d < 0 \)
\( \beta(T) = e + ff(T-78), \ ff < 0 \)

b-401 sample
\( a_a = 53.3 \)
\( c = 3.02 \)
\( d = -0.131 \)
\( e = 2.25 \)
\( ff = -7.76e-3 \)
\( T_c = 92.5 \)

fn is a fraction of intergranular material
gr(nm) is a radius of superconducting grains
penr0(nm) is a penetration depth of grains at T=0

\( \text{rg} = 10 \)
\( \text{penr0} = 1.2 \)
\( \text{fn} = 0.258 \)

write(6,1)
1 format(1x,'data filename ?')
read*,filename
write(6,2)
2 format(1x,'Hac : ',S)
read*, acfldh
R=0.085
acv=1.
nt=120
delt=(tc-77.)/float(nt)
sample='b-401'
f=1.
c
H field profiles
c
open(unit=2, file=filename, status='new')
write(2,*) sample
write(2,6) acfldh, ptfldh, spfldh, vmu, R, alpha, acv, beta
6 format(1x,'Hdc=',f6.2,'Oe, Ha*=',f6.2,'Oe, H0=',f6.2,
x 'Oe, mhu=',f4.2,x /
, lx,'R=',f6.3,' cm, pinning density=' ,e8.2,' G0e/cm2'
, 'cv=',f6.3,f4.1)
c
smni=1e30
smxi=-1e30
smnr=1e30
smxr=-1e30
do 11 j=1, nt+1
temp(j)=77. + float(j-1)*delt
if(temp(j).ge.tc) go to 11
tnorm=temp(j)/tc
ptfldh=aa*1.-(tnorm)**2)**1.5)
spftdh=c+d*(temp(j)-78.)
beta=e+ff*(temp(j)-78.)
penetr=penr0/sqrt(1.-tnorm**4)
rmnorm=rg!/penetr
vmu=fn+(1.-fn)**2.*BESS11(rnorm)/rnorm/BESS10(rnorm)
c
find alpha (pinning force density)
alpha=((ptfldh+spfldh)**(1.+beta)-spfldh**(1.+beta)/
, (4.* (1.+beta)**pi*R)
print*, 'T = ',temp(j),', pinning force = ',alpha
a=4.*pi*alpha
c
dcfldh=0.
flmax=acfldh+dcfldh
c
maximum penetration depth
r0=R-((flmax+spfldh)**(1.+beta)-spfldh**(1.+beta))
/(a**(1.+beta))
if(r0.lt.0.) r0=0.
if(dcfldh.ge.acfdh) then
r2=R-(-(-dcfldh+acfldh+spfldh)**(1.+beta)+(flmax+spfldh)
***(1.+beta))/2.*a**(1.+beta))
else
r2=R-((-dcfldh+acfldh+spfldh)**(1.+beta)+(flmax+spfldh)
***(1.+beta)-2.*spfldh**(1.+beta))/(2.*a**(1.+beta))
endif
if(r2.lt.0.) r2=0.
c
nd=100
\[ \text{dclr} = \frac{(R-r2) \text{float(nd)}}{ \text{ntime} = 50} \]

\[ \text{do 10 k=1,ntime} \]

\[ t(k) = (\text{float(k-1)}+0.5) / \text{float(ntime)} \]

\[ \text{apfld} = \text{dcfldh} + \text{acfldh} \times \cos(2. * \pi * t(k)) \]

\[ \phi(k) = 0. \]

\[ \text{if} (\text{dcfldh} \geq \text{acfldh}) \text{ go to 13} \]

\[ t1 = \text{acos}(-1.* \text{dcfldh} / \text{acfldh}) / (2.*\pi) \]

\[ t2 = 1. - t1 \]

\[ t3 = 1. - \text{acos}(1.-2.* \text{dcfldh} / \text{acfldh}) / (2.*\pi) \]

\[ \text{if} (t(k), \text{ge}.0. \text{and.} .(t(k), \text{lt}.t1) \text{ go to 14} \]

\[ \text{if} (t(k), \text{gt}.0.5 \text{and.} .(t(k), \text{le}.t2) \text{ go to 15} \]

\[ \text{if} (t(k), \text{gt}.t2 \text{and.} .(t(k), \text{le}.t3) \text{ go to 17} \]

\[ \text{if} (t(k), \text{gt}.t3) \text{ go to 18} \]

\[ 13 \quad \text{if} (t(k), \text{ge}.0. \text{and.} .(t(k), \text{lt}.0.5) \text{ go to 14} \]

\[ \text{if} (t(k), \text{ge}.0.5) \text{ go to 19} \]

\[ 14 \quad \text{rt} = R + ((\text{apfld} + \text{spfldh})**(1.+\beta) - (\text{fldhmax} + \text{spfldh})**x (1.+\beta)) / (2.*a*(1.+\beta)) \]

\[ \text{if} (\text{rt}.l0.) \text{ rt} = 0. \]

\[ \text{do 21 i=1,nd+1} \]

\[ \text{depth}(i) = (r2 + \text{float(i-1)}* \text{dclr}) / R \]

\[ \text{if} (\text{depth}(i), \text{lt}.R) \text{ then} \]

\[ \text{fldh}(i) = ((1.+\beta)*a*R*(\text{depth}(i)-1.) + (\text{fldhmax} + \text{spfldh}) **x (1.+\beta))**(1.)/(1.+\beta)) - \text{spfldh} \]

\[ \text{else} \]

\[ \text{fldh}(i) = (- (1.+\beta)*a*R*(\text{depth}(i)-1.) + (\text{apfld} + \text{spfldh})**x (1.+\beta))**(1.)/(1.+\beta)) - \text{spfldh} \]

\[ \text{endif} \]

\[ \phi(k) = \phi(k) + 2.*\pi*\text{vmu} * \text{dclr} * \text{fldh}(i) * \text{depth}(i) * R \]

\[ 21 \quad \text{continue} \]

\[ \text{go to 10} \]

\[ 19 \quad \text{rr} = R + ((\text{dcfldh} - \text{acfldh} + \text{spfldh})**(1.+\beta) - (\text{apfld} + \text{spfldh})**x (1.+\beta))/ (2.*(1.+\beta)*a) \]

\[ \text{if} (\text{rr}.l0.) \text{ rr} = 0. \]

\[ \text{do 22 i=1,nd+1} \]

\[ \text{depth}(i) = (r2 + \text{float(i-1)}* \text{dclr}) / R \]

\[ \text{if} (\text{depth}(i), \text{lt}.R) \text{ then} \]

\[ \text{fldh}(i) = (- (1.+\beta)*a*R*(\text{depth}(i)-1.) + (\text{dcfldh} - \text{acfldh} + \text{spfldh})**x (1.+\beta))**(1.)/(1.+\beta)) - \text{spfldh} \]

\[ \text{else} \]

\[ \text{fldh}(i) = ((1.+\beta)*a*R*(\text{depth}(i)-1.) + (\text{apfld} + \text{spfldh})**x (1.+\beta))**(1.)/(1.+\beta)) - \text{spfldh} \]

\[ \text{endif} \]

\[ \phi(k) = \phi(k) + 2.*\pi*\text{vmu} * \text{dclr} * \text{fldh}(i) * \text{depth}(i) * R \]

\[ 22 \quad \text{continue} \]

\[ \text{go to 10} \]

\[ 15 \quad \text{rr} = R - ((\text{apfld} + \text{spfldh})**(1.+\beta) - \text{spfldh}**x (1.+\beta))/ (1.+\beta)*a) \]
rt=0.5*(R+rr)-((fldhmax+spfldh)**(1.+beta)-spfldh**
(1.+beta))/(2.*((1.+beta)*a)

if(rt.lt.0.) rt=0.
if(rr.lt.0.) rr=0.
do 23 i=1,nd+l
depth(i)=(r2+float(i-1)*delr)/R
if(depth(i).lt.rt/R) then
fldh(i)=((1.+beta)*a*R*(depth(i)-1.)+(fldhmax+spfldh)**
(1.+beta))**(1./(1.+beta))-spfldh
else
if(depth(i).lt.rr/R) then
fldh(i)=((1.+beta)*a*(rr-R*depth(i))+spfldh***(1.+beta))
**((1./(1.+beta))-spfldh
else
fldh(i)=spfldh-((1.+beta)*a*R*(depth(i)-1.)+(-apfld+spfldh)
**((1.+beta))**(1./(1.+beta))
endif
endif
phi(k)=phi(k)+2.*pi*vmu*delr*fldh(i)*depth(i)*R
continue
go to 10
rr=R-((-dcfldh+acfldh+spfldh)**(1.+beta)-spfldh**(1.+beta))
/(1.+beta)*a)
rr=R+((-apfld+spfldh)**(1.+beta)-(-dcfldh+acfldh+spfldh)**(1.+beta)*a)
if(rr.lt.0.) rr=0.
if(rt.lt.0.) rt=0.
do 24 i=1,nd+l
depth(i)=(r2+float(i-1)*delr)/R
if(depth(i).lt.rt/R) then
fldh(i)=spfldh-((1.+beta)*a*R*(depth(i)-1.)+(dcfldh+acfldh)
**spfldh)***(1.)/(1.+beta))
else
fldh(i)=spfldh-((-apfld+spfldh)**(1.+beta)-(1.+beta)*a*R*
(depth(i)-1.))**(1./(1.+beta))
endif
endif
phi(k)=phi(k)+2.*pi*vmu*delr*fldh(i)*depth(i)*R
continue
go to 10
rr=R-((-dcfldh+acfldh+spfldh)**(1.+beta)-spfldh**(1.+beta))
/(1.+beta)*a)
rr=R+(spfldh***(1.+beta)-(apfld+spfldh)***(1.+beta))/
(1.+beta)*a)
rt=0.5*(rr+R)+(spfldh***(1.+beta)-(apfld+spfldh)***(1.+beta))/
***(1.+beta))/((1.+beta)*2.*a)
if(rr.lt.0.) rr=0.
if(rt.lt.0.) rt=0.
if(rrl.lt.0.) rrl=0.
do 25 i=1,nd+1
   depth(i)=(r2+float(i-1)*delr)/R
   if(depth(i).lt.R/R) then
      fldh(i)=((1.+beta)*a*(rr-R*depth(i))+spfldh**(1.+beta))
      x**(1./(1.+beta)) - spfldh
   else
      if(depth(i).lt.t/R) then
         fldh(i)=spfldh-((1.+beta)*a*R*(depth(i)-1.)+(dfldh+acfldh)
         x +spfldh)**(1.+beta))**(1./(1.+beta))
      else
         if(depth(i).lt.rl/R) then
            fldh(i)=spfldh-(spfldh**(1.+beta)+a*(rrl-depth(i)*R)
            x )**(1./(1.+beta))
         else
            fldh(i)=-spfldh+( (1.+beta)*a*R *( depth(i)-1.)+(apfld+spfldh)*
            x )**(1./(1.+beta))
         endif
      endif
   endif
   phi(k)=phi(k)+2.*pi*vmu*delr*fldh(i)*depth(i)*R
25 continue
go to 10
c18 rr=R-((-dfldh+acfldh+spfldh)**(1.+beta)-spfldh**(1.+beta))
   x /((1.+beta)*a)
   if(rr.lt.0.) rr=0.
   rt=0.5*(rr+R)+(spfldh**%(1.+beta)-3*(spfldh)**(1.+beta))
   x /(2.*(1.+beta)*a)
   if(rr.lt.0.) rt=0.
do 26 i=1,nd+1
   depth(i)=(r2+float(i-1)*delr)/R
   if(depth(i).lt.R/R) then
      fldh(i)=((1.+beta)*a*(rr-R*depth(i))+spfldh**(1.+beta))
      x**(1./(1.+beta))-spfldh
   else
      fldh(i)=spfldh-((1.+beta)*a*R*(depth(i)-1.)+(apfld+spfldh)*
      x**(1.+beta))**(1./(1.+beta))
   endif
   phi(k)=phi(k)+2.*pi*vmu*delr*fldh(i)*depth(i)*R
26 continue
10 continue
c calculate real or imaginary component of ac susceptibility
c rv(j)=0.
cv(j)=0.
do 27 l=1,ntime
   rv(j)=rv(j)+phi(l)*sin(2.*pi*f*t(l))/float(netime)
   cv(j)=cv(j)+phi(l)*cos(2.*pi*f*t(l))/float(netime)
27 continue
rv(j)=2.*rv(j)/acfldh/pi*R**2
rv(j)=2.*cv(j)/acfldh/pi*R**2
cv(j)=cv(j)-1.
write(2,*), temp(j), cv(j), rv(j)
if(rv(j).gt.smxi) smxi=rv(j)
if(rv(j).lt.smni) smni=rv(j)
if(cv(j).gt.smxr) smxr=cv(j)
if(cv(j).lt.smnr) smnr=cv(j)
print*, ' Xr = ', cv(j), ', Xi = ', rv(j)
          continue
          close(unit=2)
print*, 'real signal range: ', smnr, '-', smxr
print*, 'imaginary signal range: ', smni, '-', smxi
end
FIGURE CAPTIONS

Fig. A.1. Profile of local field $H_I(r)$ inside a long cylindrical superconducting sample when $H_0 \leq H_1$ and $0 \leq t \leq t_1$ where an applied magnetic field $H(t_1) = 0$. The parameters $H_0 = 1$ Oe, $H_1 = 4$ Oe, $R = 0.085$ cm, $H^* = 8$ Oe, $H_s = 3$ Oe, $\beta = 1$, and $t = T/4$ were used to generate this figure. When $H_0 > H_1$ and $0 \leq t \leq T/2$, $H(r)$ profile looks exactly the same. $T$ is the period of an ac magnetic field.

Fig. A.2. Profile of local field $H_I(r)$ when $H_0 \leq H_1$ and $t_1 \leq t \leq T/2$. $H(t_1) = 0$. This field profile is at $t = 0.45 \, T$ with the same parameter values given in Fig. A.1.

Fig. A.3. Profile of local field $H_I(r)$ when $H_0 \leq H_1$ and $T/2 \leq t \leq t_2$ where $H(t_2) = 0$ and $t_2 \geq T/2$. This field profile is at $t = 0.6 \, T$ with the same parameter values given in Fig. A.1.

Fig. A.4. Profile of local field $H_I(r)$ when $H_0 \leq H_1$ and $t_2 \leq t \leq t_3$ where $t_3 = 1 - \cos^{-1}(1-2H_0/H_1)/\omega$. This field profile is at $t = 0.8 \, T$ with the same parameter values given in Fig. A.1.

Fig. A.5. Profile of local field $H_I(r)$ when $H_0 \leq H_1$ and $t_3 \leq t \leq T$. This field profile is at $t = 0.85 \, T$ with the same parameter values given in Fig. A.1. When $H_0 > H_1$ and $T/2 \leq t \leq T$, $H(r)$ profile looks exactly the same.
Figure A.1

Figure A.2
Figure A.5
APPENDIX C. COMPUTER PROGRAMS FOR THE DATA ACQUISITION

The FORTRAN computer programs in this appendix are for the data acquisition from various electronic devices connected through IEEE 488 bus to a LSI-11 computer. See the overall circuit, Fig. 3.7. The programs are linked with subroutines for graphics (graph.obj) on a HP plotter (HP 7470A) and for a GPIB controller GPIB11V-1 (gpib.obj) made by National Instruments, Inc. Each device has its own device number for the GPIB controller. The devices are called when program lines such as j=ibup(...) are executed. In the programs the following device numbers are used; 2: HP plotter for graphics, 4: HP 3325 Synthesizer/Function Generator for $H_0$ or a heater power supply, 6: Keithley 197 Microvolt Digital Multimeter for reading temperature, 7: HP 4192A Impedance Analyzer for ac magnetic susceptibility, 8: Keithley 197 Microvolt Digital Multimeter for reading $H_0$.

Program 1. LRTEMP

This program reads inductances $L$ and serial resistances $R$ of a pickup coil wound around a superconducting sample and connected as DUT to the HP 4192A impedance analyzer in order to get its ac magnetic susceptibility as functions of $H_0$ and $T$. In the program $H_0$ or $T$ can be varied while running it, but $H_1$ and $f$ are fixed.

c byte ans,an.fm,cm(4),dname(11),sm(60),osc(5),freq(9)
byte out(50),outt(13),outh(13),dy
common /bl1/tp(200),vt(200),nn,vref
read a conversion table of temperature

open(unit=2,name='cucost.dat',type='old')
n=0
read(2,1)
1 format(/)
do 2 i=1,200
   read(2,3,end=4) tp(i),vt(i)
nn=nn+l
2 continue
4 close(unit=2)
vref=-5539.0

c
write(7,11)
11 format(1x,'oscillation freq (kHz) : ',$)
   accept*, frq
   write(7,12)
12 format(1x,'OSC level (V) : ',$)
   accept*, vosc
   write(7,13)
13 format(1x,'dc bias voltage (V) : ',$)
   accept*, vbias
   write(7,14)
14 format(1x,'Circuit mode (serial,parl,auto) : ',$)
   read(5,15) (cm(i),i=1,4)
15 format(4a1)
   write(7,16)
16 format(1x,'data filename (6 characters) : ',$)
   read(5,17) (dname(i),i=1,6)
17 format(6a1)
   write(7,18)
18 format(1x,'sample : ',$)
   read(5,19) (sm(i),i=1,60)
19 format(60a1)
   write(7,20)
20 format(1x,'conversion of Iac to Hac (Oe/mA) : ',$)
   accept*, convac
   write(7,21)
21 format(1x,'conversion of Idc to Hdc (Oe/mA) : ',$)
   accept*, convdc
   type*,' '  
c
   j=gpiib(4)
   j=gpiib(12,0)
   encode(9,22,freq) frq
   format(9.3)
   j=ibup(0,7,'fr',2)
   j=ibup(0,7,freq,9)
   j=ibup(0,7,'en',2)
   j=ibup(5,7)
   encode(5,23,osc) vosc
23 format(5.3)
c
write(7,24)
format(1x,'put a paper on a plotter. ready ? ',$,)
read(5,25) an
format(a1)
write(7,27)
format(1x,'x-axis (T, D, A) : ',$,)
read(5,25) dy
format(1x,'y-axis (L(H), R(OHM), Hac(G), Hdc(G)),
T(K) : ',$)
accept*, tmin,tmax,ttic,tymin,ymax,ytic

24 open a data file
25
write(7,30) (dname(i),i=1,10)
format(1x,'** filename = ',IOal,' **')
open(unit=2,name=dname,type='new',form='formatted')
write(2,31) (sm(i),i=1,60)
format(1x,'sample : ',60al)
write(7,32) frq,vbias
format(1x,'temperature sweep mode' J,1x,'frequency : ',f9.3,
'kHz',/1x,'bias voltage : ',f6.3,' V')
write(2,33) (cm(i),i=1,4)
format(1x,'circuit mode : ',4al)
write(2,34)
format(1x,'L(H), R(OHM), Hac(G), Hdc(G),
T(K) : ',$)
type*, ' 

30 data acquisition
31
write(7,35) format(1x,'number of data : ',$,)
accept*, nsw
write(7,36) int(1.6*nsw)
format(1x,'sweep time(sec), minimum ',j4,': ',$)
accept*, swt
idelay=int(1e3*(swt/nsw-1.6)/2.125)+1
j=ibup(0,7,'t3fl',4)
j=ibup(0,6,'0g1x',5)
j=ibup(0,8,'0g1x',5)
write(7,37)
format(1x,'ready to take data? ',$)
read(5,25) an
type*, *
write(7,34)
do 40 i=1,nsw
j=ibup(0,7,'ta',2)
j=ibup(0,7,'ex',2)
j=ibup(1,7,out,50)
decode(j-2,41,out) a,b,c
41 format(4x,e11.5,5x,e11.5,2x,f10.3)
j=ibup(1,6,out,13)
j=ibup(1,8,outh,13)
decode(11,42,outh) temp
decode(11,42,outh) hdc
42 format(e11.9)
v=1000.*temp
call tcalib(v,tmp)
hdc=convdc*hdc
hac=convac*c*sqrt(2.)
write(7,43) a,b,hac,hdc,tmp,i
write(2,43) a,b,hac,hdc,tmp,i
43 format(1x,'e13.5,', ',e13.5,', ',f10.3,', ',f10.3,', ',f6.2,',
',',',j5)
if(dy.eq.'T') call dotat(tmp,a)
if(dy.eq.'D') call dotat(hdc,a)
if(dy.eq.'A') call dotat(hac,a)
call delay(idelay)
40 continue .
call penup
j=ibup(0,7,'t1',2)
j=gpib(4)
j=ibup(5,6)
j=ibup(5,7)
j=ibup(5,8)
close(unit=2)
stop
end

Program 2. LRTHAC

This program is almost same as the program 1 LRTEMP except that in this
program $H_1$ can be varied during running it but $H_0$, $T$, and $f$ are fixed; it is used
to take data for $\chi'(H_1)$ and $\chi''(H_1)$. 
byte an, cm(4), dname(11), sm(60), osc(5), freq(9)
byte out(50), outt(13), outh(13), dy
common /bll/tP(200), vt(200), nn, vref

read a conversion table of temperature

open(unit=2, name='cucost.dat', type='old')
nn=0
read(2,1)
1 format(/)
do 2 i=1,200
read(2,3, end=4) tp(i), vt(i)
3 format(5x, f7.1, 11x, f8.2)
nn=nn+1
2 continue
4 close(unit=2)
vref=-5539.0

write(7, 11)
11 format(1x, 'oscillation freq (kHz) : ', $)
accept*, frq
write(7, 13)
13 format(1x, 'dc bias voltage (V) : ', $)
accept*, vbias
write(7, 14)
14 format(1x, 'Circuit mode (serial, parl, auto) : ', $)
read(5, 15) (cm(i), i=1, 4)
15 format(4a1)
write(7, 5)
5 format(1x, 'increase step of Vosc (mV) : ', $)
accept*, vs
vs=le-3*vs
write(7, 16)
16 format(1x, 'data filename (6 characters) : ', $)
read(5, 17) (dname(i), i=1, 6)
17 format(6a1)
write(7, 18)
18 format(1x, 'sample : ', $)
read(5, 19) (sm(i), i=1, 60)
19 format(60a1)
write(7, 20)
20 format(1x, 'conversion of Iac to Hac (Oe/mA) : ', $)
accept*, convac
write(7, 21)
21 format(1x, 'conversion of Idc to Hdc (Oe/mA) : ', $)
accept*, convdc
type* 

j=gpinb(4)
j=gpinb(12,0)
encode(9, 22, freq) frq
22 format(f9.3)
j=ibup(0, 7, 'fr', 2)
j=ibup(0,7,freq,9)
j=ibup(0,7,'en',2)
j=ibup(5,7)
c
write(7,24)
24 format(1x,'put a paper on a plotter. ready? ',S)
read(5,25) an
25 format(a1)
write(7,28)
28 format(1x,'min, max, tic Hac (Oe) : ',S)
accept*, tmin,tmax,ttic
write(7,26)
26 format(1x,'min, max, tic L(H) : ',S)
accept*, ymin,ymax,ytic
c
c graphics
c
call initgr

callspen(1)
call window(0.15,0.8,0.15,0.8)
call scale(tmin,tmax,ymin,ymax)
call axis(tmin,ymin,ttic,ytic)
type*,

dname(7)='.
dname(8)='d'
dname(9)='a'
dname(10)=''
c
c open a data file
c
c write(7,30) (dname(i),i=1,10)
30 format(1x,'** filename = ',10a1,' **')
onopen(unit=2,name=dname,type='new',form='formated')
write(2,31) (sm(i),i=1,60)
31 format(1x,'sample: ',60a1)
write(7,32) frq,vbias
32 format(1x,'Hac sweep mode' /,1x,'frequency : ',f9.3,
  ' kHz' /,1x,'bias voltage : ',f6.3,' V')
write(2,33) (cm(i),i=1,4)
33 format(1x,'circuit mode : ',4a1)
write(2,34)
34 format(1x,'/7x, 'L(H)' /9x,'R(OHM)' /8x,'Hac(G)' /5x,'Hdc(G)' ,
  5x,'T(K)')
type*,

c data acquisition
c
c j=ibup(0,6,'t0g1x',5)
j=ibup(0,8,'t0g1x',5)
write(7,37)
37 format(1x,'one way or round trip (O/R)? ',S)
read(5,25) an
39 write(7,34)
vosc=0.
do 51 i=1,500
vosc=vosc+vs
if(vosc.gt.1.1) go to 52
encode(5,53,osc) vosc
format(f5.3)
j=ibup(0,7,'ol',2)
j=ibup(0,7,osc,5)
j=ibup(0,7,'en',2)
j=ibup(0,7,'ta',2)
j=ibup(0,7,'ex',2)
j=ibup(1,7,out,50)
decode(j-2,41,out) a,b,c
41 format(4x,e11.5,5x,e11.5,2x,f10.3)
j=ibup(1,6,out,13)
j=ibup(1,8,outh,13)
decode(11,42,outh) temp
decode(11,42,outh) hdc
42 format(e11.9)
v=1000.*temp
call tcalib(v,tmp)
hdc=convdc*hdc
hac=convac*c*sqrt(2.)
write(7,43) a,b,hac,hdc,tmp,i
write(2,43) a,b,hac,hdc,tmp,i
43 format(1x,'e13.5',',e13.5',',f10.3',',f10.3',',f6.2',',i5)
call dotat(hac,a)
51 continue
52 if(an.eq.'O') go to 60
c
do 53 i=1,500
vosc=vosc-vs
if(vosc.lt.5e-3) go to 60
encode(5,53,osc) vosc
j=ibup(0,7,'ol',2)
j=ibup(0,7,osc,5)
j=ibup(0,7,'en',2)
j=ibup(0,7,'ta',2)
j=ibup(0,7,'ex',2)
j=ibup(1,7,out,50)
decode(j-2,41,out) a,b,c
j=ibup(1,6,out,13)
j=ibup(1,8,outh,13)
decode(11,42,outh) temp
decode(11,42,outh) hdc
v=1000.*temp
call tcalib(v,tmp)
hdc=convdc*hdc
hac=convac*c*sqrt(2.)
write(7,43) a,b,hac,hdc,tmp,i
write(2,43) a,b,hac,hdc,tmp,i
call dotat(hac,a)
53 continue
60 call penup
List of Plotting Programs

HPLRTP: This program reads a data file which contains the values of \( L, R, H_0, H_1, T \) and plots \( L \) and \( R \) as a function of one of the parameters \( H_0, H_1, \) and \( T \).

HPRLKI: This program reads and compares two data files: a file of the empty coil and a file of the coil with the superconducting sample. \( \Delta L = L \) (with the sample) \(- L \) (without the sample) at exactly the same values of parameters, \( H_0, H_1, f, \) and \( T \), gives \( \chi' \), the real component of the ac magnetic susceptibility.

HPIMKI: This program is exactly the same as HPRLKI but this calculates \( \chi'' \), the imaginary component of the ac magnetic susceptibility, from the values of \( \Delta R \).

GRAPH: This program contains several subroutines which control HP plotters to draw graphs or figures.

Initgr: this sets HP plotters to default conditions.

Window: this sets a plotting area inside which plotting can occur.

Scale: this changes plotter units of the plotting area into user units.
Axis: this draws the box around the plotting area and tic marks along the boundary of the box.

Spen: this select a plotting pen.

Penup: this raises the pen.

Pendwn: this lowers the pen.

Move: this moves the pen at a given position.

Dotat: this marks a dot at a given position.
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