THE DEVELOPMENT OF A HIGH SENSITIVITY AC SUSCEPTOMETER AND ITS APPLICATION TO THE STUDY OF HIGH TEMPERATURE SUPERCONDUCTORS

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Abstract

The development of a high sensitivity ac susceptometer is outlined in this thesis. The intended use of this device is for the measurement of the magnetic field dependent penetration depth of high temperature superconductors in the Meissner state. The nature of the field depence may be useful in discerning the superconducting pairing state in these materials.

Test measurements on the completed ac susceptometer show that its field dependent background is too large to extract any meaningful results from this experiment. However, the device has been used successfully to measure the temperature dependent penetration depth and the vortex melting transition of the high temperature superconductor $YBa_2Cu_3O_{6.95}$.

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Chapter 1

Introduction

For nearly a decade now there has been intensive research into high temperature superconductors. The attraction to the study of these materials is fueled as much by their underlying physics as by their promise to be of great practical and commercial use. In terms of the physics there is a still great deal that is unknown. In fact, there is at present no theory that can satisfactorily explain the superconducting mechanism of high temperature superconductors. Many clues to solving this mystery rest in the knowledge of the pairing state, and a variety of experiments have been pursued with this in mind. One experiment that has been proposed, but not yet successfully carried out (at least in YBa₂Cu₃O_{6.95} [1, 2]), is the measurement of the magnetic field dependence of the Meissner state penetration depth.

Measurements of critical fields and the mapping of the H - T phase diagram have always been of interest in the study of superconductors. There has been little interest, however, in precision measurements of $\lambda(H)$ in the Meissner state until the work of Yip and Sauls on the nonlinear Meissner effect [3]. Their work (detailed further in Chapter 5) revealed that the nature of the field dependence of λ is sensitive to the pairing state. In particular, $\lambda(H)$ for a superconductor with a conventional isotropic gap function should vary as H^2 , while $\lambda(H)$ for a superconductor with lines of nodes should be linear in H. Furthermore, the theory suggests that the field response may be used to locate the nodal lines in the case of an unconventional gap. This may help answer the long outstanding question of what the pairing state is in the high temperature

1

superconductors. Many other experiments have provided strong evidence to suggest $d_{x^2-y^2}$ pairing, but the evidence is not conclusive [4]. Observation, then, of the nonlinear Meissner effect may supply some of the strongest evidence yet to answering this question.

The reason for building the ac susceptometer to be described in this thesis is to be able to measure the magnetic field dependence of the penetration depth of the high temperature superconductor $YBa_2Cu_3O_{6.95}$ for fields below H_{c1} . Presently, measurements using ac susceptometers have become quite popular in the field of high temperature superconductivity. Many of the interesting features of superconductors - the Meissner effect, the critical temperature, vortex nucleation and dynamics - can be viewed macroscopically through changes in the bulk magnetization of a sample. As a result an ac susceptometer lends itself very well to the study of such materials.

The ac susceptometer itself is a very simple device. Typically, it consists of a dc magnet, a solenoid used to generate an ac field, and a pair of detection coils. The mutual inductance between each of the detection coils and the solenoid is the same, so when the two are connected in series the induced voltage across the pair is zero. The sample is placed inside one of the detection coils. Its presence will alter the mutual inductance of this coil and now the net voltage across the coils is nonzero. This voltage is proportional to the sample's magnetic moment. If the sample is a superconductor in the Meissner state, its moment will be proportional to the volume of the region magnetically shielded by supercurrents. This volume, always smaller then the actual volume of the sample because of the finite penetration of the magnetic field, is proportional to $\lambda(T, H)$. This allows a measurement of $\Delta\lambda(T, H) = \lambda(T, H) - \lambda(T_o, H_o)$ where T_o and H_o are some reference temperature and field respectively.

Chapter 2

AC Susceptibility and the AC Susceptometer

The magnetic susceptibility χ of a material relates its magnetization M to an applied external magnetic field H. For linear media in a dc field this relationship is simply

$$\chi = \frac{M}{H}.\tag{2.1}$$

For nonlinear media or in the case where one applies a time dependent field a more relevant definition is

$$\chi = \frac{\mathrm{d}M}{\mathrm{d}H},\tag{2.2}$$

where χ is now the differential or ac susceptibility respectively.

2.1 The Complex Magnetic Susceptibility

Consider a sample in a periodic external magnetic field $H(t) = H_{dc} + H_1 \sin wt$. In response to the field, the sample will develop a periodic magnetization M(t), which can be expanded as the Fourier series

$$M(t) = H_1 \sum_{n=1}^{\infty} \operatorname{Im}(\chi_n e^{inwt}) = H_1 \sum_{n=1}^{\infty} (\chi'_n \sin nwt - \chi''_n \cos nwt),$$
(2.3)

where Im() denotes the imaginary part of the complex magnetization [5]. The coefficient $\chi_n = \chi'_n - i\chi''_n$ is the complex magnetic susceptibility of the *n*th harmonic, where

$$\chi'_n = \frac{w}{\pi H_1} \int_{\frac{-\pi}{w}}^{\frac{\pi}{w}} M(t) \sin nwt \, \mathrm{d}t,$$

$$\chi_n'' = \frac{-w}{\pi H_1} \int_{\frac{-\pi}{w}}^{\frac{\pi}{w}} M(t) \cos nwt \, \mathrm{d}t.$$

As defined here, the χ_n will typically be a functions of both H_{dc} and H_1 . (This definition is equivalent to that in equation 2.2. See Appendix A.)

The fundamental component of the magnetization $M(t) = H_1(\chi'_1 \sin wt + \chi''_1 \cos wt)$ has a real part in-phase with the applied field H(t) and an imaginary part that is 90° out-of-phase with H(t). This attaches very obvious physical meaning to the fundamental susceptibility χ_1 : χ'_1 corresponds to the inductive response (energy storage) of the sample, χ''_1 corresponds to losses (energy dissipation) in the sample. The higher harmonics are generally associated with hysteresis and nonlinearity of magnetization [5]. In the limit of $H_1 \rightarrow 0$, the higher harmonics will become negligible and $\chi'_1 \rightarrow \frac{dM}{dH} = \chi_{dc}$.

For example, a superconductor in the complete Meissner state will have a magnetization field that is of equal amplitude but opposite direction to that of the applied field. Equation 2.4 gives $\chi'_1 = -1$ and $\chi''_1 = 0$ reflecting the perfect diamagnetism of the material. In the normal state (neglecting core diamagnetism and the real conductivity), the external field will completely penetrate the sample, and $\chi'_1 = \chi''_1 = 0$. At intermediate temperatures χ'_1 will be a negative number (> -1), and χ''_1 will be a small positive number reflecting ac losses [6].

2.2 Experimental Technique

The general design of an ac susceptometer is shown in Figure 2.1; all coils are coaxial. An ac current is passed through the primary or drive coil to produce an alternating magnetic field. A steady state magnetic field can also be superimposed by applying a dc current to the outermost coil. The only measured quantity in this technique is the induced voltage across the series combination of the secondary coils, which of course is

(2.4)

due to the time variation of the magnetic flux through these coils. The secondary coils are made as identical as possible and are connected in opposition. In the absence of a sample, the induced voltage of the coils will be equal in magnitude but 180° out of phase resulting in an observed null signal. Specifically, from Faraday's law, $\varepsilon = -d\Phi/dt$, the induced voltage is

$$v(t) = (M_1 - M_2) \frac{\mathrm{d}I_p}{\mathrm{d}t} = 0$$
(2.5)

where M_1 and M_2 are the mutual inductances between the primary and each of the secondary coils, and I_p is the sinusoidal drive current. To the extent that the coils are identical M_1 equals M_2 and the minus sign reflects the fact that they are connected in opposition (or counterwound), and hence v(t) = 0. In practice, however, it is not a trivial matter to achieve this balance. Even if great care has been taken in winding all the coils and the secondaries are placed symmetrically in the ac field, a residual voltage is still very likely. Additional methods of compensation are usually employed to cancel this mismatch and obtain the desired level of sensitivity and stability in the susceptometer. The direct addition of a compensating voltage $v_c(t)$ of appropriate magnitude and phase is a common technique. Here

$$v(t) = (M_1 - M_2)\frac{\mathrm{d}I_p}{\mathrm{d}t} + v_c(t) = 0.$$
(2.6)

With the system nulled, the introduction of a magnetic sample into one of the coils (the pick-up coil) will now cause an imbalance in the signal. This voltage is due solely to the sample. To see this, consider the three voltage sources in the secondary circuit, which sum to

$$v(t) = \frac{d\Phi_1}{dt} + \frac{d\Phi_2}{dt} + v_c(t) = \mu_o NA \left[\frac{dH(t) + M(t)}{dt} - \frac{dH(t)}{dt} \right] + v_c(t),$$
(2.7)



Figure 2.1: General ac susceptometer design.

where M(t) and H(t) are the sample magnetization and applied field respectively, N is the number of turns in each coil, and A is their cross sectional area. The first term in this equation is the induced voltage in the pick-up coil, the second term is the induced voltage in the empty (compensation) coil, and the third term, $v_c(t)$, is again the applied compensation voltage. In light of equation 2.6, this simply reduces to

$$v(t) = \mu_o N A \frac{\mathrm{d}M(t)}{\mathrm{d}t},\tag{2.8}$$

a signal voltage proportional to the rate of change of the sample magnetization. Substituting the definition of M(t) from equation 2.3 into equation 2.8 gives

$$v(t) = \mu_{o} w NAH_{1} \sum_{n=1}^{\infty} n(\chi_{n}' \cos nwt + \chi_{n}'' \sin nwt)$$

= $v_{o} \sum_{n=1}^{\infty} n(\chi_{n}' \cos nwt + \chi_{n}'' \sin nwt),$ (2.9)

with $v_o = \mu_o w N A H_1$. The signal voltage is an infinite sum of cosine and sine wave harmonics.

2.3 Calibration of the Susceptometer

The expression for v_o above is only strictly true for a uniformly magnetized sample that completely fills the coil. Such an arrangement is not typical. In general v(t) must depend upon the geometry and spatial orientation of the sample and the pick-up coil. This information is contained in a calibration constant, α . It is the constant of proportionality for the experimentally relevant relation $v(t) \propto dM(t)/dt = \chi wH$, and is related to the mutual inductance between the sample and the pick-up coil. α can be calculated via the following consideration.

The field due to the sample is characterized by internal currents of volume density $\nabla \times \mathbf{M}$ and surface density $\mathbf{M} \times \hat{n}$, where \mathbf{M} is the sample magnetization and \hat{n} is the unit vector normal to its surface. A cylindrical sample of uniform axial magnetization $(\nabla \times \mathbf{M} = 0)$ can be equivalently modeled as a solenoid of the same size having N_s turns each carrying current I [8]. This implies a magnetization of magnitude

$$M = N_s I/l_s, (2.10)$$

where l_s is the length of the sample.

The induced voltage in the pick-up coil is

$$v(t) = -M_{sp} \frac{\mathrm{d}I}{\mathrm{d}t},\tag{2.11}$$

where M_{sp} is the mutual inductance between the sample and the coil. Rearranging equation 2.10 and substituting gives

$$v(t) = -M_{sp} \frac{l_s}{N_s} \frac{\mathrm{d}M}{\mathrm{d}t},\tag{2.12}$$

or

$$v(t) = -\alpha \chi w H. \tag{2.13}$$

The calibration constant then is just

$$\alpha = \frac{M_{sp}}{N_s} l_s, \tag{2.14}$$

the mutual inductance per sample turn multiplied by the length of the sample.

To calculate the mutual inductance, it is almost always easiest to consider the voltage induced in an inner coil (sample) due to a current in an outer coil (pick-up.) So, for a coil much longer than the sample the mutual inductance is

$$M = M_{ps} = M_{sp} = \frac{\mu_o N_p}{l_p} N_s A_s$$

where N_p and l_p are the number of turns and length of the pick-up coil respectively and A_s is the cross sectional area of the sample. This gives

$$\alpha = \frac{\mu_o N_p}{l_p} V_s, \tag{2.15}$$

a calibration constant proportional to the sample volume V_s and independent of the parameter N_s , as it must be.

This solution for α was of course obtained using the long coil approximation for the pick-up coil, which may not be appropriate in many cases. Other particular sample/coil arrangements also lend themselves to rather simple determinations of α [7, 8], but these too might prove unsuitable approximations for many sample sizes and shapes. I therefore

present the following as a precise method for the numerical calculation of α for a uniformly magnetized sample. This method is accurate for cylindrical samples with magnetization vector parallel to both their axis and the pick-up coil axis. To the extent that one can ignore edge effects, this method will also be a very good approximation for bar or slab shaped samples that are magnetized along one its lengths parallel to the pick-up coil axis. (Most superconducting samples to be studied using the susceptometer of this thesis are slab shaped, hence the motivation for this exercise.) There are no restrictions on the length of the sample.

The fact that the sample is uniformly magnetized allows one to approximate it as a solenoid and use the definition of α from equation 2.14. It also allows one to write the mutual inductance as

$$M_{sp} = \sum_{i=1}^{N_p} \sum_{j=1}^{N_s} \int \int \mathbf{B}'(r, z_{ij}) \cdot d\mathbf{A}_j, \qquad (2.16)$$

where $\mathbf{B}'(r, z_{ij})$ is the magnetic field per unit current of the i^{th} loop of the pick-up coil calculated at the distance r from the centre axis in the plane of the j^{th} solenoid loop (see Figure 2.2.) The integration is over the cross sectional area of the solenoid loops, and can be computed numerically by dividing the area into $K \times L$ finite equally sized elements $\Delta A_s = A_s/KL$. The value $\mathbf{B}'(r_{kl}, z_{ij})$ at the centre of each ΔA_s is taken to be the value over the entire area element. This gives

$$M_{sp} = \sum_{i=1}^{N_p} \sum_{j=1}^{N_s} \sum_{k=1}^{K} \sum_{l=1}^{L} B'(r_{kl}, z_{ij}) \Delta A_s, \qquad (2.17)$$

and therefore

$$\alpha = \frac{V_s}{N_s K L} \sum_{i=1}^N \sum_{j=1}^{N_s} \sum_{k=1}^K \sum_{l=1}^L B'(r_{kl}, z_{ij}).$$
(2.18)

Notice that assuming $B'(r_{kl}, z_{ij}) = B$ is constant over the entire sample recovers the solution for α of 2.15. Of course, the idea here is to assume a more complicated form



Figure 2.2: Schematic for numerical calculation of α for a uniformly magnetized rectangular sample. It is shown approximated as a solenoid of equivalent dimension. The end view shows cross sectional area of sample split into $K \times L$ elements ΔA_s .

of the magnetic field (for greater accuracy) and use a computer program to calculate α . The summations of equation 2.18 are easily translated into four programming loops. The degree of precision in the program is determined by the size of the model parameters N_s, K , and L. The best accuracy can be had if one uses

$$B'(r, z_{ij}) = B_z/I = \frac{\mu_o}{2\pi} \frac{1}{\sqrt{(a+r)^2 + z^2}} \left[K + \frac{a^2 - r^2 - z^2}{(a-r)^2 + z^2} E \right]$$

the general expression for the field per unit current of a circular current loop [9]. K and E are complete elliptic integrals of the first and second kind respectively.¹ The degree of precision is determined by the size of the model parameters N_s , K, and L used in the program.

¹Some mathematical packages such as *Mathematica* or *Maple* have built in routines for calculating elliptic integrals.

One further refinement needs to be made, if the compensation coil is in close proximity to the sample. Under such conditions, there will be an induced voltage in this coil which is no longer negligible. The true calibration constant then is just

$$\alpha = \alpha_{pick-up} - \alpha_{compensation}, \qquad (2.19)$$

The terms are calculated separately by employing equation 2.18 for each coil.

In lieu of calculating α mathematically, calibration can also be done experimentally using a standard of known susceptibility. This is a common technique. It is an attractive method in that it allows one to make a simple determination of α without needing to know the exact geometries of the secondary coils. There is one drawback however. The resulting calibration constant, though possibly very precise, will only be strictly valid for samples having the same dimensions and the same position inside the susceptometer as the standard.

2.4 Signal Detection

The voltage v(t) induced in the secondary coils can be measured using an ac voltmeter or an oscilloscope. However, these devices can detect only the entire voltage sum of 2.9, and a great deal of information about the system is lost. Using a lock-in amplifier allows one to selectively measure both the in-phase and out-of-phase components of a given harmonic, revealing much more detail of the sample magnetization. The lock-in behaves as a voltmeter with a very high Q bandpass filter at the input. It will measure a signal of specific frequency only.

This frequency is determined by the frequency of a reference input. Usually this input is the sync out from the function generator providing the primary current. The reference phase of the lock-in must also be set so that its time base is synchronous with the drive field $H(t) = H_1 \sin wt$. This allows for the separation of the v(t) into its in-phase (χ''_n) and out-of-phase (χ'_n) components, where the n^{th} harmonic of w was used for the reference. A dual phase lock-in amplifier will display both signals. With devices having only one output, the phase must be changed by 90° to obtain the out-of-phase signal.

Clearly, the real power of ac susceptometry resides in the lock-in amplifier's ability to selectively measure both the real and imaginary parts of any χ_n .

2.5 Setting the Phase

The phase can be set by using, as the input signal to the lock-in, the voltage drop across a pure resistance that is in series with the primary circuit. This voltage will be in phase with the primary current and therefore in phase with the applied field H(t). The reference phase setting is adjusted until the signal appears in only one channel (i.e. the 0° or 90° setting.) This channel is now set to measure the signal proportional to χ''_n . The channel in quadrature will measure the χ'_n .

It is important that the voltage sampled off the resistor have the same electrical path to the lock-in as would the signal voltage from the secondary coils. For example, the signal voltage may be stepped up by a pre-amp transformer before the lock-in. Ideally the transformer would not affect the phase. In practice however, one is almost certain to pick up a phase shift as most real transformers are only nearly ideal. Having the sample voltage go through the pre-amp (at the same settings) as well will allow for an accurate phase setting.

If one is studying superconductors, another method of setting the phase employs the properties of the sample itself. This method is convenient because it can be performed without altering the experimental set up. Here one takes advantage of the fact that a superconductor cooled in zero field and kept at sufficiently low temperatures will not have any loss associated with a small drive field (ie. $H_1 \ll H_{c_1}$.) The sample temperature

is changed within this region of no loss. The resulting signal is due to the temperature dependence of the penetration depth, which is a purely inductive response. Again the reference phase is set such that the resulting change in signal appears in only one channel. This will be the out-of-phase channel.

Chapter 3

Development of the Apparatus

The real heart of an ac susceptometer is of course the trio of solenoids comprised of the primary and two secondary coils. To describe them as a single entity, which is often useful, I have coined the term *ac coil set*. Also, for the purposes of this chapter only, I will limit the use of the term (ac) susceptometer to refer only to that part of the apparatus containing the dc magnet, ac coil set, and sample holder. The cryostat and the electronics then will be described separately from the susceptometer in this chapter.

The development of this apparatus was already a work in progress of Dr. W. Hardy, when I joined the project. As a result, I will detail only those parts of the apparatus that are specific to this experimental technique, and give particular highlight to those efforts in development that were my own. It should be duly noted that the design of the apparatus was Dr. Hardy's, and that the cryostat, the susceptometer (less the dc magnet) and one version of the ac coil set had already been built. I built the magnet and a second coil set.

3.1 The Cryostat

The cryostat is of standard design. See Figure 3.3. It consists of a brass flange with stainless steel vertical tubes (three of them, each 3/8'' outer diameter) that connect to the susceptometer. The flange is 0.350'' thick and has an outer diameter of 5.375'' to cover the 4" inner diameter helium dewar that the susceptometer sits in. The flange also contains a 3/8'' hole and O-ring seal for a helium transfer tube (or safety blow-off valve,

after transfer is complete), and two multi-lead connectors (not shown in diagram), one for the temperature control electronics, the other for the superconducting dc magnet leads. Connected to the stainless steel tubes are five copper disks, of diameter just less then 4", that act as radiation baffles. Leads for the primary and secondary coils are run down the centre of two of the tubes. The third tube is used as a vacuum line to evacuate the susceptometer; it also contains a drive shaft allowing mechanical adjustment of a voltage compensation circuit that is housed within the cap of the susceptometer.



Figure 3.3: Diagram of the cryostat.

3.2 The AC Susceptometer

A cross section of the ac susceptometer is shown in Figure 3.4. In this diagram the susceptometer is disassembled, as it would appear when one is changing the sample. The sample holder or pot is mated with the main body of the susceptometer with the aid of two guide pins (not shown) that are permanently fixed to the dc coil form. The two pieces are held together by twelve 4-40 hex cap screws with indium wire forming the vacuum seal. The dc coil form is bolted to the susceptometer cap in a similar way,with the exception of the guide pins which are not needed here.

Because two different versions of the ac coil set were built, Figure 3.4 shows just a general template for its arrangement inside the susceptometer. Both versions are shown in greater detail in Figure 3.6. The ac coil set has a copper flange, which is fastened to the dc coil form using six 4-40 hex cap screws. The sample is mounted on a very thin, very pure sapphire plate with a small amount of vacuum grease. The sample plate is held to the sapphire mounting block of the pot also with a small amount of vacuum grease. When the ac coil set and the pot are fully assembled, the sample sits at the centre of the first coil of the secondary pair, ie. the pick-up coil.

Other pertinent aspects of the susceptometer are discussed in more detail below. The voltage compensation circuit, which is also shown in Figure 3.4, will be described in the next section of this chapter.

3.2.1 Thermometry

It is essential to be able to change the temperature of the sample during an experiment without interferring with the ac measurements on the sample. For this reason the sample heater and thermometer are connected to a sapphire mounting block situated well away from the sample and the ac coil set. Sapphire has a very high thermal conductivity and



Figure 3.4: Diagram of the ac susceptometer. The sample holder or pot is disassembled from the dc coil form. The orientation of the primary and secondary coils is shown only schematically.

can be made very pure, so it is also used for the sample mounting plate. Quartz on the other hand has much lower thermal conductivity and is therefore used to thermally isolate yet mechanically connect the mounting block to the rest of the pot. Provided then that the layer of vacuum grease between the block and the plate, and the plate and the sample is very thin, this system will allow for very quick, effective and non-intrusive regulation of the sample's temperature.

It is often desirable to keep the temperature of the helium bath surrounding the susceptometer accurately constant. To do this requires a separate bath heater, thermometer and temperature controller. The total thermal power delivered to the bath is the sum of the power from the sample heater P_s and bath heater P_b . If P_b is initially set (with $P_s = 0$) to a value just greater than the anticipated maximum power P_{max} to be used by the sample heater over the course of an experiment, then P_b can always be adjusted later such that $P_s + P_b = P_{max}$, a constant, for any setting of P_s .

The sample thermometer is a piece of a Lakeshore carbon glass resistor potted in Stycast 2850FT epoxy to a strip of copper foil. The foil is held to the mounting block with GE varnish. The sample heater is a 200Ω chip resistor attached to the underside of the sapphire block, and cannot be seen in the view of Figure 3.4. Manual or automated control of the sample temperature is done with a Conductus LTC-20 temperature controller.

The bath heater, a 200 Ω metal glaze resistor, is mounted to the base of the pot with Stycast 2850FT epoxy. The bath thermometer, an Allan Bradley carbon composition resistor (200 Ω nominal room temperature resistance) is located at the side of the susceptometer cap. The temperature controller, built in-house, allows only manual adjustment of the set point.

3.2.2 The Superconducting DC Magnet

The magnetic field along the central axis of a thin solenoid of finite length l and radius a is given by the equation

$$B_{z} = \frac{\mu_{o}NI}{2l} \left\{ \frac{1 - 2z/l}{\left[(2a/l)^{2} + (2z/l - 1)^{2}\right]^{1/2}} + \frac{1 + 2z/l}{\left[(2a/l)^{2} + (2z/l + 1)^{2}\right]^{1/2}} \right\},$$
(3.20)

where N is the number of turns, I is the current, and z is the distance from the centre of the solenoid [12]. Using this equation a computer program was written to help select an appropriate design for the dc magnet. The program returned only those winding configurations that would produce a symmetric magnet with a field homogeneity of better than 1 part per thousand over the length ($\sim 1-2mm$) of a typical sample positioned at the centre of the pick-up coil. The secondary coils, separated by 0.25", are symmetrically placed about the geometrical centre of the magnet. If the magnet is wound accurately, the field will be the same at each coil.

It was necessary to also include in the program the following design constraints:

- The size of the superconducting (Nb-Ti) wire 0.014 inches in diameter.
- The number of layers of windings in the magnet 30 layers each being separated by 0.002 inches of mylar. This limit set by the amount of available space.
- The size and shape of the magnet cylindrical, 2.0 inches in length, with a central notch. The notched design is simplest configuration to wind that can give a reasonably homogeneous field.

The final design can be seen as a part of Figure 3.4. The notch is formed from a copper collar epoxied (with Stycast 1266) to the magnet coil form. There are 14 layers of windings to the top of the notch, each of layer having 40 turns (20 turns on either side of the copper collar.) Each of the 16 complete layers above the notch has 136 turns on it.

The magnet calibration was determined theoretically using equation 3.20 and the average values of its turns density and layer thickness. The axial field profile of the magnet is shown in Figure 3.5. The magnitude of the field in the region of the susceptometer is calculated to be 468 gauss/amp.



Figure 3.5: The calculated axial field profile for the superconducting dc magnet. The inset highlights the region of interest containing the ac coil set. The positions of the secondary coils are shown centred at ± 0.225 inches.

3.2.3 The AC Coil Set

The two versions of the ac coil set are shown in Figure 3.6. Only the top half of the cross section of each piece is shown; the axial field strength of the respective primary coil is superimposed on this view. The reason a second, slightly different version of the ac coil set had to built was that the first design proved to be completely unsuitable for the type of dc field scan measurements to be done. (This will be discussed further in Chapter 3.)

Common to both designs, however, is the copper flange used to mount the ac coil set to the main body of the dc coil form. Also, in both cases, phenolic linen washers are used as a lateral form for the primary coil, and a copper faraday shield is epoxied (with Stycast 1266) to the outer diameter of the primary coil's radial form. The shield is electrically and thermally grounded to the copper flange.

The faraday shield is composed of a single layer of closely packed insulated copper wires that are parallel to the coil axis. This provides effective shielding of static electric fields, but doesn't support induced tangential currents that would otherwise shield the desirable magnetic drive field. To build the shield, the copper wire is first wound on a teflon form and potted in 1266 epoxy. Before the epoxy has fully cured this rather malleable *coil* is removed from the teflon and cut to form a sheet of parallel wires. This is then wrapped in place around the radial coil form and the epoxy is allowed to completely cure. In the version I coil set, the bare ends of the faraday shield are folded against the back of the phenolic linen washers and held in pressure contact with the copper flange; the washer is bolted to the flange. In version II, these ends fit into a groove in the flange that is filled with silver epoxy. Here the washer is simply butted to the flange and held in place with 1266 epoxy.

The coil form of version I was machined out of phenolic paper rod. It has an outer diameter of 0.250", an inner diameter of 0.125", and an overall length of 1.250". Two square grooves of length 0.200" and inner diameter 0.167" act as forms for the secondary coils. These grooves, 0.250" apart, are positioned symmetrically about the centre of the form. The secondary coils are counter wound out of a single length of #44 copper wire (insulation unknown) and potted in Stycast 1266 epoxy. There are 1359 turns in each of the coils. The primary coil is wound with #34 formvar insulated copper wire. The faraday shield was made from this same size wire. There are four layers of windings in the primary. The first and third layers are complete. The second and fourth layers have



Figure 3.6: Diagram of the ac coil sets. The faraday shielding is butted against the flange in version I, in version II it is siver epoxied into a groove in the flange. The field profiles of each primary coil is superimposed on the figure.

only ten turns at either end; these *compensation* turns homogenize the field over the length of the coil. A spacer made of mylar tape fills the remainder of the second layer. The primary coil has a constant field strength of 127 gauss/amp over the length of the secondary coils, as calculated numerically using equation 3.20.

As mentioned before a second coil set had to be built, because this first one was discovered to be unsuitable for magnetic measurements. Explicitly, the problem with this version of the ac coil set was that it had a huge background signal dependent upon the applied dc magnet field. To make matters worse, this signal was hysteretic suggesting the presence of ferromagnetic impurities in the apparatus itself. Obviously, if one wants to make measurements as a function of the field, this can not be tolerated. The magnetism of the various construction materials of the coil set were tested with a dc SQUID magnetometer. These tests did not reveal a definite source for the signal. The goal then in building a second version was to limit the amount of known magnetic impurities in the vicinity of the secondary coils. This was to be achieved in two ways: (1) using high purity construction materials and (2) by reducing the amount of material in the vicinity of the secondary coils.

A custom ground sapphire tube [13], with an outer diameter 0.250", inner diameter of 0.198" and length 1.250", was used for the primary coil form in this design. Apart from its high purity, which should ensure negligible contribution to the field dependent background, the sapphire also has another advantage. Its high thermal conductivity, will quickly remove any heat (generated by the primary, radiated by the sample, etc.) from the ac coil set to the helium bath.

There is no permanent coil form for the secondary coils of this coil set. They were wound separately on teflon forms, potted in 1266 epoxy, and then removed from the form once the epoxy had cured. This process, which had an ~ 80% success rate of producing an intact coil, is described further in Figure 3.7. The completed coils have 512 turns and nominal dimensions: OD= 0.188" and length= 0.23". (The OD of the two coils differ by ~ 0.002 - 0.003". Inadvertently, some of the coil forms were made of a softer teflon, which resulted in a slightly smaller coil.) The coils are then epoxied (with 1266) in place inside the sapphire tube. Two slits ground into the sapphire helped facilitate the application of the epoxy and allowed for the passage of the pick-up coil leads around the outside of the compensation coil. (It should be noted that the layer of epoxy between the coils and the sapphire appeared to go cloudy after being cooled in liquid nitrogen. This apparent cloudiness is presumed to be due to the coils contracting away from the sapphire.)

The secondary coils were wound out of 0.003" diameter pure Nb wire insulated with formvar [15]. Niobium was chosen for its superconducting properties. It has a $T_c = 9.2K$, so at liquid helium temperatures it would shield any possible ferromagnetic impurities inside its bulk from the magnetic fields. Niobium was chosen over other superconducting metals such as Pb or PbSn, because it has a high critical field $H_{c1} = 1980$ gauss. This is much larger than fields to be used in the experiment, which ensures that the coil windings should always remain in the Meissner state and not give rise to a signal due to a transition into the mixed state. Also, there will be no losses and therefore no heating associated with a type II superconductor in an oscillating field above H_{c1} .

The faraday shield and the primary coil were made from 0.003" diameter 99.99% pure copper wire insulated with formvar [14]. The primary was wound using the same scheme as the one described above. The wire is of a smaller diameter so a greater number of turns, 18 on either side, is required in the compensation layers (ie. the second and fourth layers.) This primary coil has a constant field strength of 254 gauss/amp over the length of the secondary coils, as calculated using equation 3.20.

To protect the very fine wire leads used in this ac coil set, all connections were made on fixed terminal posts. The five posts, 2-56 threaded brass rod, are held in a lucite ring that is fastened around the copper flange of the coil set inside the susceptometer cap. There is a pair of posts for the primary coil connections with the power leads from the top of the cryostat, a second set for the secondary leads to the preamp, and a fifth post where the secondary coils are connected in series opposition. All lead ends from the ac coil set are placed inside tubes of 99.95% pure copper foil, which are then crushed between washers on the 2-56 bolt. The niobium leads are first cleaned with HF acid. Solder connections are easily made to the copper foil or to the brass posts themselves.



Figure 3.7: Construction of niobium secondary coils: The teflon form fits the 8mm collet of coil winding machine. The wound coil is potted in epoxy, which is allowed to fully cure. At this point the pilot hole at the end of the form is extended as the dashed line, and the extremity of teflon is pulled away. The entire piece is then immersed in liquid N_2 after which the solid coil is easily removed. Because the teflon does stretch during winding, the final length of the coil is ~ 0.23".

3.3 The Electronics

The electronic set up for this apparatus resembles that shown if Figure 2.1, with the inclusion of a preamplification stage before the lock-in detector. The preamp is a Princeton Applied Research (PAR) Model 119 Differential Preamplifier input to a PAR Model 114 Signal Conditioning Amplifier. The preamp is operated in the differential input mode (and almost exclusively in the 100:1 transformer mode) and has a single output to the Stanford Research Systems Model SR850 DSP Lock-In Amplifier. The function generator, a Hewlett Packard 3325A, is coupled to the primary circuit via an isolation transformer to break its ground loop.

The schematic of the susceptometer circuitry, including the voltage compensation, is shown in Figure 3.8. The 100 Ω dropping resistor, in the primary circuit, is much larger than the impedance (at f = 10 kHz) of the rest of the circuit when the primary coil is at liquid helium temperatures. As a result, under the conditions of operation, the combination of the dropping resistor and function generator behaves as an ac current source. The voltage drop across the 1 Ω resistor can be used to set the phase of the lock-in amplifier. The capacitor C_{res} in the secondary circuit is used to achieve series resonance with the inductive component (largely dominated by the secondary coils) of the circuit. This is done to optimize the source impedance for the PAR Model 119 preamp operating in the transformer mode at 10 kHz. At resonance the source impedance is the purely resistive 3.5 Ω of the secondaries, which means the Model 119 will be operating within the 0.5dB noise figure contour at this frequency [16]. It should be noted, that the second version of the ac coil set has a much smaller impedance (due to the smaller self inductance of the secondary coils) and the capacitor C_{res} was not needed.



Figure 3.8: The susceptometer circuit. Coarse compensation of the out-of-phase voltage is done at low temperatures with the variable mutual inductance. All fine compensation is done at room temperature via decade transformers.

The voltage compensation circuitry has two distinct parts: (1) the coarse compensation done at low temperatures by a homemade variable mutual inductance and (2) the fine compensation done at room temperature by a set of Electro Scientific Industries Dekatran DT72A decade transformers capable of supplying six decades of voltage resolution. The low temperature compensation is used exclusively to null out-of-phase signals, while the room temperature circuit is used for both in-phase and out-of-phase compensation. Coarse compensation is not needed for any in-phase imbalance in the signal. It is quite small compared to the out-of-phase imbalance, which is due to the mismatch of the secondary coils and the inductive shielding of the sample (if present.) The advantage to the low temperature nulling is the lower thermal noise.

The variable inductor of (1) is a set of equal turn (but counter wound) coils in series with the primary circuit that straddle a pick-up coil in series with secondary circuit, see Figure 3.9. These coils are wound on a 5/16" diameter phenolic paper tube. A 1/4" diameter copper slug sits inside the tube; its position can be varied from outside the cryostat by the drive shaft described in Section 3.1 and shown in Figure 3.4 of this chapter. The skin depth of copper at 4.2K and 10 kHz is $\sim 0.002 - 0.003$ ", so essentially the entire volume of the slug is shielded under these conditions, and it can therefore be used to effectively set the magnitude and sign of the magnetic flux cutting the pick-up coil. This means the magnitude and sign of this induced compensating voltage can be set by the position of the copper slug. The phase of this induced voltage is $\simeq 90^{\circ}$ out of phase with the drive current. Certainly losses from the copper and the phenolic paper are present, but they appear to be negligible. This can be seen in the plot of induced voltage vs. position of copper slug, displayed in Figure 3.9. Only the out-of-phase component of the signal voltage imbalance is affected by this circuit.

For fine tuning, two reference voltages (one in-phase, the other out-of-phase) are picked off the primary circuit, their magnitudes are appropriately transformed by the


Figure 3.9: The low temperature compensation circuit. (a) Diagram of the circuit itself. (b) Typical response of the circuit. The position of the tuning slug is plotted as turns of the key from the extreme counter clockwise setting. In practice, one would null the imbalanced signal with this circuit to near one of the minima indicated by the arrows. Fine adjustment of the compensation is then done with the decade transformers.

decade transformers, and the resulting voltages are then injected into the secondary circuit. All transformer ratios given in Figure 3.8 are with respect to the top inductor, and represent the actual number of turns in each case. The phase difference between the primary current and output voltage from the transformer across the 1.3Ω resistor was experimentally determined to be 0.97° . The phase of the output voltage from the transformer in series with the primary circuit was measured to be 89.95° . The orthogonality of these two voltages allows one to quickly set the decade transformers to give a null signal. Their performance over several decades of voltage transformation is shown in Figue 3.10.



Figure 3.10: The performance of the decade transformers. There is negligible phase shift in the output from the decade transformers. The orthogonality of the input voltages is preserved.

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Chapter 4

Performance of the Apparatus

This apparatus was designed such that one could "see" a 1Å change in the penetration depth, λ , of a typical sized YBCO single crystal. What is the sensitivity required for such resolution? To answer this consider equation 2.13,

$$v = \frac{\mu_o N_p}{l_p} V_s \chi w H, \tag{4.21}$$

the induced voltage in a pick-up coil due to a sample of volume V_s and susceptibility χ . For a superconductor in the Meissner state, $\chi = \chi'_1 = -1$. (The signal is purely inductive.) The area of a typical high quality YBCO crystal is $\sim 1 \times 1 \text{ mm}^2$, to estimate its change in apparent volume due to a 1Å change in λ one multiplies the area by 2λ . This gives $\Delta V = 2 \times 10^{-16} \text{ m}^2$. For the first version of the pick-up coil $N_p = 1360$, and $l_p = 0.2^{"} = 5.08 \times 10^{-3} \text{ m}$. Taking $w = 2\pi \times 10^4 \text{ radHz}$ and $\mu_o H = 10^{-3} \text{ T}$ (ie. 10 gauss), gives a signal of about

$$v = 3.4 \times 10^{-9}$$
 volts.

The intrinsic noise from the secondary coils at 1.2K is

$$v = \sqrt{4kTR\Delta\nu}$$

 $\approx \sqrt{4(1.38 \times 10^{-23})(1.2)(3.5)(1)}$
 $\approx 1.5 \times 10^{-11}$ volts.

The amplifier noise when operating at 10 kHz within the 0.5 dB noise contour is \sim 0.09 nV for a 3.5 Ω input. Theoretically, measuring a signal of 3.4 nV with a reasonable signal to noise ratio does seem quite possible.

The actual behavior of the ac susceptometer is the subject of this chapter. The resolution along with measurements on the temperature and magnetic field response of the device will be presented here. The two ac coils sets essentially represent two different susceptometers. They are discussed separately and in chronological order to highlight how the unacceptable field dependence of the first coil set became the impetus for construction of the second.

4.1 AC Coil Set - Version I

Here the secondary circuit includes the capacitor C_{res} shown in Figure 3.8. The true resonant frequency was determined experimentally to be 10.7 kHz. This frequency for the drive field was used for all experiments in this thesis except where explicitly stated.

4.1.1 Sample Holder Temperature Dependence

The dependence of the apparatus on the sample holder temperature T_s was measured with the helium bath in regulation at 1.2K, which is the lowest temperature reachable with our pumping system. Only the sapphire sample plate was present inside the pick-up coil. The results for the out-of-phase signal are shown in Figure 4.11. The signal is very small, and it rapidly becomes constant with increasing temperature. Although there are very few data points, the susceptibility roughly fits a 1/T dependence suggestive of a Curie term due to paramagnetic impurities[17].

This sample plate was later replaced with a much higher purity sapphire plate, which when tested gave no detectable temperature dependent signal. It appears then that impurities in the former plate were responsible for the paramagnetic observations in Figure 4.11.



Figure 4.11: The temperature dependent backgound signal of the sapphire sample plate. f = 10.7 kHz. $H_1 = 8$ gauss. $\chi \propto v \propto 1/T$ appears to indicate paramagnetic behavior.

4.1.2 DC Magnetic Field Dependence

The field dependence of the background is shown in Figure 4.12. Again only the sapphire sample plate was present inside the pick-up coil. The data plotted is for the out-of-phase signal. The size of the signal is very large. Even at an applied current of 0.5 amps (ie. 0.5 amps \times 468 gauss/amp = 234 gauss $\approx H_1$ of YBCO) the signal is in excess of 100X the desired resolution. To make matters worse, the signal is also hysteretic. The dc magnet current was supplied by a HP 6632A programable power supply. This power supply is of single polarity only, so one had to switch the leads to the magnet to measure the reverse field. There was sufficient interruption in data taking, during this process, to make uncertain the true pattern of the hysteresis loop around zero current. The presence of hysteresis in general, however, is absolutely unmistakable.

A systematic study was undertaken to determine the magnetic characteristics of the



Figure 4.12: The dc field dependent backgound signal. f = 10.7 kHz. $H_1 = 8$ gauss. Inset shows more clearly the hysteresis.

construction materials that are in the vicinity of the secondary coils. This, it was hoped, would reveal which of the substances was responsible for the large background signal. The measurements were done with a Quantum Design dc SQUID magnetometer. Samples of phenolic paper, 1266 epoxy, #44 copper wire (secondary coils), and #36 heavy formvar copper wire (similar to the wire in the primary coil though of a smaller gauge) were studied. Hysteresis loops over ± 2000 gauss at 5K for all materials are shown in Figure 4.13. The data is normalized with respect to mass by plotting the magnetic moment in units of emu per mass of sample in grams. The results show that all the materials are magnetic, each with a susceptibility $|\chi| = |M/H| \sim 5 \times 10^{-7}$ emu g⁻¹ gauss⁻¹. Only the 1266 epoxy does not show signs of hysteresis.

It is impossible to tell from this data which of the materials are responsible for the



Figure 4.13: Hysteresis loops at 5K for ac coil set construction materials measured with a dc SQUID magnetometer.

background signal. (We do know that the 1266 is not responsible for the hysteresis.) It is also impossible to say whether the signal arises from a mass imbalance, or a winding imbalance. By mass imbalance, meant an inhomogeneity in the spatial distribution of a given material with respect to the secondary coils. If a magnetic material is evenly distributed, then the voltage it would induce in each coil would be the same and no signal would be observed. Of course, if imperfections exist between the windings of the coils (and this is quite likely over 1360 turns) then there will be a signal regardless of balanced mass. These two effects cannot be distinguished from each other. To remedy the problem, it was decided at this point to build another ac coil set. Great effort and expense was put into procuring the high purity materials for its construction. The design was altered to limit the mass in close proximity to the secondary coils.

4.2 AC Coil Set - Version II

The inductance of each of the new secondary coils was ~ 10^{-3} H giving a combined reactance for the pair of just 125 Ω at 10 kHz, so it was decided to forgo the use of the capacitor C_{res} . The effect on the noise was negligible.

4.2.1 Sample Holder Temperature Dependence

The dependence of the apparatus on the sample holder temperature T_s was again measured with the helium bath in regulation at 1.2K, and with only the high purity sapphire sample plate present inside the pick-up coil. As expected, there was no detectable signal at low temperatures. However, as the sample plate was taken to higher temperatures a signal was observed. The onset of this high temperature response was at about 30K. This experiment was repeated on a separate cool down of the apparatus, the sample plate having been removed, cleaned and replaced. The same behavior was observed again; the magnitude of the effect was almost twice as large this time. It is believed that the plate was positioned slightly farther inside the pick-up coil in the second experiment. This strongly suggests that the effect may be due to differential heating of the secondary coils caused by thermal radiation from the sample plate. It is also known that the coils are not in good contact with the sapphire tube (the coils pulled away from the tube at low temperature as mentioned in Chapter 2), which makes this idea seem reasonable. Figure 4.14 shows the raw data of the induced out-of-phase voltage as a function of T_s along with a plot of $\ln(v)$ vs. $\ln(T)$ for both trials. These show the signal voltage varying with temperature as $T^{5.8}$. While not strictly consistent with the Stefan-Boltzman law (radiative power $\propto T^4$), any mechanism with such a temperature dependence almost certainly involves a radiative process. The temperature dependence of the emissivity of the sapphire plate may account for the deviation from T^4 behavior.



The experimental implications of this thermal effect will be discussed later.

Figure 4.14: Dependence of the susceptometer signal as a function of the sample holder temperature for the version II ac coil set. Taking the natural log of both variables gives a near linear relationship. At high temperature the respective slopes for each trial are 5.6 and 6.0, an average of 5.8.

4.2.2 DC Magnetic Field Dependence

To facilitate proper hysteresis measurements, two power supplies were used here. One, an Anatek 6007, was used to supply a constant current $-I_{max}$ to the dc magnet. The other power supply, the HP 6632A, connected in parallel, was programmed to step through current values from 0 to $2I_{max}$. The total dc magnet current as measured by an HP 3478A multimeter and the signal voltage from the SR850 lock-in was logged by the control computer via the IEEE 488 bus. With this method, many continuous hysteresis loops can be measured in succession.

Field-dependent-background measurements over several loops are shown in Figure 4.15. The data is plotted as a function of the dc magnet current (shown on the left) and as a function of real time (shown on the right). Trial 1 is from a separate cool down from Trials 2 and 3. In Trial 3 the sample holder temperature was set at 100K, it was 1.2K for the other two.



Figure 4.15: The dc field dependent backgound signal of the version II ac coil set for f = 10.7 kHz and $H_1 = 8$ gauss. The out-of-phase voltage signal is plotted in real time and as function of the magnet current. The drift (in Trial 1) was eliminated by thermally isolating the electronics. The background signal is much larger at $T_{sample} = 100$ K

There are many notable features in this data. Most notable, of course, is the fact that the background of the version II coil set is also field dependent. Despite a huge effort to eliminate the problem, the field dependence still persists and it is hysteretic. Compared to the first coil set at an applied current of 0.5 amps (see Figure 4.12) the signal for the $T_{sample} = 1.2$ K data is roughly 10X less. To give a true comparison of the backgrounds however, one needs to multiply this figure by the ratio of turns of the two pick-up coils (this is essentially the ratio of the sensitivities of the two coil sets.) This means that the field dependent background of the second coil set was reduced by a factor of $10 \times 511/1360 \sim 3.8$ over the first design. A reasonable improvement, but not quite the 100X reduction required to bring this background down to the level of the resolution.

Also present in the Trial 1 data is a fluctuation in the voltage signal baseline. This is easily seen in the time plot. It affects the hysteresis plots by smearing them in the voltage axis direction. The source of this drift was discovered to be changes in the air temperature of the laboratory. In particular the fluctuations in an otherwise constant background signal were found to be syncronized with the automated room air-conditioner. To fix this problem, the room temperature electronics of Figure 3.8 (less the decade transformers) were thermally anchored to an aluminum plate and isolated inside a styrofoam box. Water circulation from a temperature controlled bath kept the temperature of the aluminum plate constant. The improvements in performance were significant. The hysteresis loops in Trials 2 and 3 lie almost directly on top of one another. This repeatability, coupled with the use of the computer (for data collection and current control), allows one to make a very large number of continuous traces over the hysteresis loop. This will be useful as a form of signal averaging when it comes to measuring the nonlinear Meissner effect in $YBa_2Cu_3O_{6.95}$. This background is still too large to allow for direct measurement of the effect, but it may be possible to subtract background data from sample data to give a reliable measure of the sample's field response. Averaging over many subsequent current loops for both sets of data will allow improvement in the signal to noise.

In the normal state, the thickness of a YBCO crystals is much smaller than its skin

depth at 10 kHz, so it appears transparent to the magnetic fields. In theory then, it may be possible to measure the background by taking the sample above $T_c = 93$ K. In practice however, the results of Trial 3 rule this out. The field dependent background of the apparatus with sample plate temperature at 100K is significantly larger than at 1.2K. The two would have to be the same for this technique to work.

4.3 Determining the Resolution of the Apparatus

4.3.1 Measuring $\Delta\lambda(T)$ of YBa₂Cu₃O_{6.95}

Precision measurements of the temperature dependence of the penetration depth of YBCO have been done at UBC using microwave cavity perturbation. The linear temperature dependence of $\lambda(T)$ at low T, a signature of superconductors with nodes in the gap function, was first observed here using this technique [18]. Repeat measurements of this experiment were done with the ac susceptometer. Both versions of the ac coil set were used to measure $\Delta\lambda(T) = \lambda(T) - \lambda(1.2K)$ for the sample Ylq, a high purity, twinned YBa₂Cu₃O_{6.95} single crystal. The crystal is a thin slab of thickness $t \sim 27 \ \mu m$. The broad face of the crystal, the *ab* plane, is $\sim 1.5 \times 1.5 \ mm^2$. As in the microwave measurements, H_1 is applied parallel to the *ab* plane. Measurements were taken over the range 0 - 20K, well below temperatures where the thermal effects in the version II coil set become noticeable. Figure 4.16 shows $\Delta\lambda(T)$ for both susceptometer measurements as well as the microwave measurements for the same crystal. All three sets of data are in good agreement, which establishes the ability of the ac susceptometer to do precision, state-of-the-art measurements.

In the cavity perturbation method, the change in penetration depth is related to a shift in the resonant frequency of the cavity [18]. For the ac susceptometer, $\Delta\lambda(T)$ is proportional to real part of the susceptibility χ'_1 . The calibration for penetration depth

measurements is exceptionally simple; one does not need to resort to any of the techniques described in Chapter 2. Here the susceptometer is calibrated *in situ* for each sample by measuring the out-of-phase signal, $V_{s,n}$, that results from taking the sample from 1.2K to 100K. In the normal state, the crystal is in the thin limit so there is essentially complete penetration of the drive field. At 1.2K the penetration depth is negligible compared to the crystal thickness t. To a very good approximation then the transition from 1.2K to 100K corresponds to a $\Delta\lambda(T) = t/2$, and the calibration constant is just $k = 2V_{s,n}/t$.

For version I, $k_I = 2(27 \times 10^4)/(1.63^{-4}) = 0.83 \text{ Å/nV}.$

For version II, $k_{II} = 2(27 \times 10^4)/(0.4898^{-4}) = 2.8 \text{ Å/nV}.$

The value $k_{II}/k_I = 3.4$ is a precise measure of the sensitivity ratio between the two ac coil sets. In the section above, this was simply estimated by the turns ratio of the pick-up coils ie. $N_I/N_{II} = 2.7$. This new value means that the improvement in the field dependent background in version II was actually just a factor of 3, not 3.8.

4.3.2 Calculating the Resolution from $\Delta\lambda(\mathbf{T})$

The resolution of the apparatus in angstroms is calculated by multiplying half the peakto-peak voltage noise (measured off a strip chart recorder that is output from the lock-in) by the calibration constant. Typical values for the noise during the $\lambda(T)$ experiments were 12×10^{-10} and 5×10^{-10} volts peak-to-peak for version I and II respectively. This gives a resolution of 0.5 Å for version I and 0.7 Å for version II. Both coil sets exceed the desired 1 Å resolution set out as the design minimum at the beginning of this chapter.



Figure 4.16: $\Delta\lambda(T)$ for the YBa₂Cu₃O_{6.95} sample Ylq.

Chapter 5

The Nonlinear Meissner Effect: $\lambda(H)$ of YBa₂Cu₃O_{6.95}

5.1 Theory

The electrodynamics of a superconductor in the Meissner state $(H < H_{c1})$ obey the London equations

$$egin{array}{rcl}
abla^2ec{B}&=&rac{1}{\lambda^2}ec{B}\
abla^2ec{j}_s&=&rac{1}{\lambda^2}ec{j}_s. \end{array}$$

It is a well known result from these equations that a magnetic field \vec{B} decays inside a semi-infinite superconducting slab as

$$B(z) = Be^{-z/\lambda},$$

(satisfying the boundary condition B(0) = B) where z is the distance into the superconductor. For high temperature superconductors, which are Type II superconductors, the characteristic length of this decay is the London penetration depth, $\lambda = \sqrt{m/(\mu_o \rho_s(T)e^2)}$. Here, $\rho_s(T)$ is the superfluid density; m and e are the electron mass and charge respectively.

Under most conditions, as it was above, the supercurrent density is sufficiently defined by the linear relation

$$\vec{j}_s = -e\rho_s(T)\vec{v}_s,\tag{5.22}$$

where \vec{v}_s is the supercurrent velocity. Yip and Sauls derive a nonlinear velocity term in

 \vec{j}_s . (In their model they consider the supercurrent to be largely confined to two dimensions.) They then re-solve the London equation, now nonlinear with the inclusion of the higher order term in \vec{j}_s , and from this derive an effective penetration depth that depends explicitly on H. They do this for both conventional and unconventional superconductors. Their results are summarized below.

5.1.1 Conventional Gap

Here the supercurrent is given by

$$\vec{j}_s = -e\rho_s(T)\vec{v}_s \left[1 - \alpha(T)\left(\frac{v_s}{v_c}\right)^2\right].$$
(5.23)

The second term is a correction to the supercurrent of 5.22, which is reduced due to pair breaking in finite fields at nonzero temperatures. Here $v_s \ll v_c$, the critical velocity, defined as $\Delta(T)/p_f$ (the energy gap divided by the Fermi momentum.) The coefficient $\alpha(T)$ is always positive and tends to zero as $T \to 0$, as a consequence of the existence of the gap.

Substituting this \vec{j}_s into the London equation gives

$$\nabla^2 \vec{v}_s - \frac{1}{\lambda^2} \vec{v}_s \left[1 - \alpha(T) \left(\frac{v_s}{v_c} \right)^2 \right] = 0, \qquad (5.24)$$

where the term $\alpha(T) \left(\frac{v_s}{v_c}\right)^2 \ll 1$ has been ignored in the Laplacian term. This nonlinear London equation is solved¹ in closed form for *H*, satisfying the boundary condition

$$\frac{\mathrm{d}v_s}{\mathrm{d}z}\mid_{z=0} = \frac{e}{c}H.$$

An effective penetration depth is defined from the initial decay rate of H inside the superconductor. It is given as

$$\lambda_{eff}(T,H) = \lambda(T) \left[1 - \frac{1}{3}\alpha(T) \left(\frac{H}{H_o(T)} \right)^2 \right]^{-1}$$

¹Details of this calculation are given by Xu, Yip and Sauls [2]. They do not appear in the original paper on the nonlinear Meissner effect [3].

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$$\simeq \lambda(T) \left[1 + \frac{1}{3} \alpha(T) \left(\frac{H}{H_o(T)} \right)^2 \right], \qquad (5.25)$$

where $\lambda(T)$ is the zero field London penetration depth and $H_o(T) = \frac{3}{4}cv_c(T)/e\lambda(T)$ is a constant of the order of the thermodynamic critical field. The change in λ_{eff} is proportional to H^2 and thus as expected λ_{eff} is larger than $\lambda(T)$. The effect of the field therefore is to increase the effective penetration depth. By pair breaking, the supercurrent density is reduced, and so too is the superconductor's ability to screen the field.

Experimentally, one can probably expect a smaller correction to $\lambda(T)$ than what is predicted above. Here λ_{eff} was defined from the initial decay rate, and so it is only truly applicable in describing the magnetic field near the surface of the superconductor. However, any measurement will be sensitive to the entire shielding range and not just the interface. We have estimated that this definition of λ_{eff} leads to a correction in $\lambda(T)$ that is ~ 3X too large. Although it is not explicitly stated in [2], this same definition for λ_{eff} is most likely used in the case of unconventional gap as well. As a result, the corrections to $\lambda(T)$ given below sould also be considered as too large.

5.1.2 Unconventional $(d_{x^2-y^2})$ Gap

A cross section of the Fermi tube for the $d_{x^2-y^2}$ pairing state is shown in Figure 5.17. There are four nodes in the gap occurring at the positions $|\hat{k}_x| = |\hat{k}_y|$. At T = 0, quasiparticle occupation at the nodes will remain nonzero. This gives rise to a quadratic \vec{v}_s term in the supercurrent \vec{j}_s that is independent of temperature. Because of the anisotropy of the gap, however, no single form for the $d_{x^2-y^2}$ supercurrent can be written. For the case where \vec{v}_s is along a node (see Figure 5.17), the supercurrent is given by the equation

$$\vec{j}_s = -e\rho_s(T)\vec{v}_s \left[1 - \frac{|\vec{v}_s|}{v_o}\right],\tag{5.26}$$

where $v_o = 2\Delta_o/v_f$ is a characteristic scale and Δ_o is the gap maximum at an antinode. The second term can be viewed as the quasiparticle backflow from the node opposite \vec{v}_s , that reduces the supercurrent. This backflow is calculated over the wedge of occupied states about the node.



Figure 5.17: The $d_{x^2-y^2}$ gap function. The supercurrent velocity is along a node. Opposite this node is the wedge of occupied quasiparticle states that constitute the backflow current.

For the case where \vec{v}_s is along an antinode (ie. 45° to a node, along the direction of the gap maximum), the supercurrent is

$$\vec{j}_s = -e\rho_s(T)\vec{v}_s \left[1 - \frac{1}{\sqrt{2}}\frac{|\vec{v}_s|}{v_o}\right].$$
(5.27)

The only difference with equation 5.26 is the factor $1/\sqrt{2}$ that appears in the backflow term. This factor is just the $\cos(45^\circ)$ for the new projection of \vec{j}_s along the nodal lines. This anisotropy will appear in the resulting nonlinear London equation and in its solution as well.² The factor of $1/\sqrt{2}$ of course shows up in the equations for the

²Again the details of the calculation are given in [2].

effective penetration depth, where hopefully it can be exploited experimentally to probe the anisotropy of the gap in high T_c superconductors.

The pertinent equations are

0

0

$$\lambda_{eff}(T,H) = \lambda(T) \left[1 - \frac{2}{3} \frac{H}{H_o} \right]^{-1}, \quad \vec{H} \parallel \text{node},$$

$$r \quad \lambda_{eff}(T,H) \simeq \lambda(T) \left[1 + \frac{2}{3} \frac{H}{H_o} \right],$$
(5.28)

and

$$\lambda_{eff}(T,H) = \lambda(T) \left[1 - \frac{1}{\sqrt{2}} \frac{2}{3} \frac{H}{H_o} \right]^{-1}, \quad \vec{H} \parallel \text{antinode},$$

$$r \quad \lambda_{eff}(T,H) \simeq \lambda(T) \left[1 + \frac{1}{\sqrt{2}} \frac{2}{3} \frac{H}{H_o} \right],$$
(5.29)

where $H_o = (v_o/\lambda(T))(c/e)$ is also of order the H_c , thermodynamic critical field. The observation of this linear field dependence in the penetration depth would strongly support the existence of unconventional pairing. The subsequent observation of an anisotropy of $1/\sqrt{2}$ in λ_{eff} , for field orientations along a node and antinode, would be very strong evidence for $d_{x^2-y^2}$.

One should keep in mind that the thermal excitations, responsible for the H^2 dependence of λ_{eff} in the conventional superconductor, must necessarily be present in unconventional superconductors as well. However, for sufficiently small T and large H, the linear term will dominate. Experimentally, the magnitude of H is limited by the critical field H_{c1} , the superconductor must be in the Meissner state for meaningful results, so one must work at low temperatures. At any finite T however, there will remain a cross-over field below which the quadratic contribution will become significant. If care is not taken, this could obscure detection of the linear behavior.

5.1.3 Some Estimates

How low must the temperature be to allow for the observation of the linear behavior in the field range $0 \rightarrow H_{c1}$? Yip and Sauls argue that the cross-over field and temperature will be related by $\frac{H}{H_o} \simeq \frac{T}{T_c}$. Assuming $H_o \simeq 10^4$ gauss, $T_c \simeq 100$ K, and $H_{c1} \simeq 250$ gauss for a typical cuprate superconductor [3] and taking $H = H_{c1}/10$ implies a temperature of 0.25K. This is roughly 1/4 of the lowest temperature presently achievable with our apparatus. At 1K then, the cross-over field is $\simeq 100$ gauss. This a very large fraction of H_{c1} , but possibly the linear term will still be observable over the remainder of the field range.

How large is the nonlinear Meissner effect expected to be? Xu, Yip and Sauls [2] suggest the change in $\lambda_{\parallel}(\vec{H})$ is roughly $2\lambda_{\parallel}H_{c1}/H_o \sim 30$ Å for an ac measurement. For a crystal of the size of Ylq ($\sim 1.5 \times 1.5 \times 0.027 \text{ mm}^3$) and using the version II ac coil set, this translates into a $\sim 11 \text{ nV}$ signal. This is $\sim 40 \text{X}$ larger than the voltage noise of the version II ac coil set. However, in terms of the field dependent signal of the apparatus itself, 30 Å is only $\sim \frac{1}{6}$ of the background at 250 gauss. This is certainly measurable, if an accurate background signal can be subtracted from the total signal.

5.2 Experiment

Measurements of the induced voltage as a function of applied dc current were made with the version II ac coil set on the YBa₂Cu₃O_{6.95} sample Ylq, a large crystal which will improve resolution. Only the field dependence of λ will be tested for now.

The experimental set up was the same as that described in section 4.2.2. The probe was cooled with magnetic shielding around the dewar with the shield left in place for all measurements. Field sweeps were made at different sample temperatures (1 and 10K in that order) for three different maximum dc current levels, corresponding to fields of 100, 200, and 240 gauss. The sample was cooled in zero field before each set of trials for the three maximum current settings; this was done by heating the sample to 100 K, turning off the dc magnet and the drive solenoid, then allowing the sample to cool. Bath temperature was held constant at 1.2 K, and the drive field was kept at the same magnitude (8 gauss) and frequency (10.7 kHZ) for all trials. The number of consecutive sweeps through the current loop was typically 50 for the sample trials, while for the background trials it was 100. Background measurements were made at 1.2 K only, for each of the three dc field levels.

Because of the temperature dependent signal associated with this coil set, the background response could not be measured by heating the sample above T_c , which requires a sample temperature ~ 100 K. It is known (see section 4.2.2) that the field dependent background at this temperature is much larger than it is at 1.2 K, making this approach completely unacceptable. The only alternative was to measure the background, before and after the sample measurement, by actually removing the sample from the susceptometer. This requires the susceptometer to be brought to room temperature and opened up between the subsequent runs.

5.3 Results and Discussion

In the end only one set of background measurements (the one taken prior to the sample measurements) could be used in the data analysis. The background measured after the sample was removed was markedly different, both in magnitude and shape of its hysteresis loop, from any field response previously observed for this coil set. See Figure 5.21. Fearing contamination, the sample plate was removed and cleaned, and N_2 gas was blown through the susceptometer. The background measurement was repeated, but this *anomalous* response persisted. The source of this problem has not yet been

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identified. Figure 5.20 shows three typical backgrounds (including the first one from this experiment) for the apparatus. They are very similar, but as they were not taken under identical conditions the repeatability of the background for this method is still unknown.

The results from the 240 gauss trials are shown below. The data for each trial has been averaged over all of its consecutive hysteresis loops. The averaging is not sensitive to the direction in which the current is stepped through the loop. This was done deliberately to remove the effects of any slow monotonic drift in the baseline. The hysteresis is also averaged when using this approach, so the signal appears as a unique function of each current setting. Figure 5.18 displays the averaged data for all measurements. Figure 5.19 shows the sample data corrected for the background. The results are very unclear, and without proper knowledge of the background very little can be concluded about $\lambda(H)$ for YBa₂Cu₃O_{6.95}.

There is a large asymmetry associated with the direction of the magnet current. The sample data for 1.2 and 10.0 K is very nearly the same for negative current values, but is substantially different for positive current. There is also an asymmetry in the magnitude of the corrected data. This is also seen in the background measurements, but had it been an effect of the background only then it should have disappeared from the corrected data. It did not. Possibly, this is an indication that the magnetic field might be distorted by some permanent magnetism in the proximity of the pick-up coils.

The fact that all background measurements (see Figure 5.20) differ from the sample data by the same sign does suggest that some field effect of the YBa₂Cu₃O_{6.95} crystal was truly detected. A signal voltage of 6 nV was measured at -0.51 amps (ie. -240 gauss), and if it is to be believed then this constitutes a $\Delta\lambda(H) = 16$ Å for $H \simeq H_{c1}$. If one considers the 4 nV spread between the background measurements at this field level to represent the uncertainty in the background, then $\Delta\lambda(H) = 16 \pm 11$ Å at $H \simeq H_{c1}$. This

would strongly suggest that the $\Delta\lambda(H_{c1}) \sim 30$ Å predict by Xu, Yip and Sauls represents an upper limit for the nonlinear Meissner effect in YBa₂Cu₃O_{6.95}.

The only real conclusion that can be drawn is that more work on the background will have to be done before a proper measurement of the $\lambda(H)$ can be made. It would be best if the background could be reduced further. This would certainly entail building another ac coil set. However, reliable measurements might still be possible with the present apparatus if a better method for determining the background can be found. One solution might be to place a thermal shield inside the secondary coils. This would eliminate the problem of the field dependent background being a function of the sample temperature as well. This in turn would allow for the in situ determination of the background by heating the YBa₂Cu₃O_{6.95} sample above Tc.



Figure 5.18: Averaged data for magnetic field response of $YBa_2Cu_3O_{6.95}$ sample Ylq and background.



Figure 5.19: Corrected data for $YBa_2Cu_3O_{6.95}$ sample Ylq.



Figure 5.20: Background measurements from separated cool downs, not done under identical conditions. This data is consistent with an observable magnetic effect in $\rm YBa_2Cu_3O_{6.95}$.



Figure 5.21: Anomalous background measurement for the version II coil set. The origins of the problem are not yet known.

Chapter 6

The Vortex Melting Transition in YBa₂Cu₃O_{6.95}

In this chapter I will present some preliminary results from studies of the vortex melting transition in $YBa_2Cu_3O_{6.95}$ done using this ac susceptometer with the version I coil set.

This transition was believed to be of first order, and so should be accompanied by a discontinuity in the first derivatives of the free energy G. Recent observations at the University of British Columbia by Liang, Bonn and Hardy [19] of a discontinuity in the magnetization ($M = -\partial G/\partial H$) of a YBa₂Cu₃O_{6.95} single crystal provided the first direct proof that the vortex melting transition was indeed first order. The sample, of superior quality, had exceptionally low pinning, as characterized by an irreversibility line well below its vortex melting transition. This allowed for the very clear measurements. In the case where there is large amounts of pinning, the transition from a vortex liquid to a vortex solid would be much more broad and therefore escape detection. This is precisely why there had never been a previous observation of this effect in YBa₂Cu₃O_{6.95}.

The measurements of Liang et al. were done using dc SQUID magnetometer, but certainly similar tests on the same crystal using the ac susceptometer should also reveal the existence the vortex melting transition. What will the transition look like for an ac measurement? If one takes the real part of the complex susceptibility to closely approximate $\frac{dM}{dH}$ in the limit that $H_1 \ll H_{dc}$, then χ'_1 would be expected to diverge at the discontinuity in M. See Figure 6.22. This hypothesis is the same whether the experiment is done as a function of dc field or temperature.



Figure 6.22: Predicted ac response of the vortex melting transition. The real part of the complex susceptibility (dashed line) is expected to follow the derivative of the dc magnetization. The measurement can be made as a function of temperature of magnetic field.

6.1 Experiment

Using the same YBa₂Cu₃O_{6.95} sample as above (labeled Up2), measurements were done in a fixed field ($H_{dc} = 10, 20 \text{ kOe}$) as a function of temperature ($T = 100 \rightarrow 60 \text{ K}$), a region of operation suitable for the version I coil set. The dc magnet current was supplied by an HP6260B power supply. A custom computer program was written to control the sample temperature via the Conductus LTC-20, and collect data from the SR850 lock-in. The sample was held perpendicular to the magnetic field on the face of a sapphire block that was epoxied to the end of a standard sapphire sample plate. This orientation is favorable for vortex formation in the sample. There was no background signal (within the resolution $\pm 10^{-8}$ volts) due to the sample holder over the temperature range 4 K \rightarrow 100 K.

6.2 Results and Discussion

Figure 6.23 shows the voltage signal as a function of temperature for two field settings, 10 and 20 kOe. The drive voltage was $1.0V_{p-p}$; the drive field $H_1 = 0.8$ gauss. There is clearly a step in both sets of data marking the possible presence of the vortex lattice melting transition. The steps are present in both the in and out-of-phase data. On either side of the step the out-of-phase components do behave as predicted, however there is no *spike* marking the transition as would also be expected if χ'_1 is truly the derivative of the dc magnetization. Is this the vortex melting transition then?

Comparison with the dc data [19] strongly suggests that it is. Transitions occur at 87.90 K and 88.25 K at 10 kOe, and at 89.80 K and 90.20 K at 20 kOe for the ac susceptometer and dc magnetometer measurements respectively. One would expect the two measurements to be identical, since $H_1 \ll H_{dc}$ and so only represents a very small perturbation to the dc condition. At both field values however, the difference between the two types of measurement is 0.4 K. The sample thermometer in the ac susceptometer was shown to change by only 0.06 K over an applied field of 20 kOe at 85 K, so this is not the problem. The T_c of this crystal was measured with the susceptometer in zero field ($H_1 = 0.2$ gauss) and found to be 92.7 K. It is given in [19] as 93.1 K. Clearly, the temperature discrepancy between the two devices is systematic. Possibly the sample thermometer in the susceptometer was not properly calibrated in this temperature range.

It has been suggested [20] that due to hysteresis about the transition, the crystal, under the low drive fields present here, cannot respond quickly enough to the changes in the field. As a result, one does not measure a sharp transition. The crystal is in a mixture of the two states at the expected transition point and as a result one just measures an average magnetization. Unfortunately, no drive fields larger than 0.8 gauss $(1.0V_{p-p})$ were studied at the time of the experiment, so this hypothesis has not yet been tested. However, the data in Figure 6.24 does show a small peak on the $1.0V_{p-p}$ data (just at the low temperature end of the transition) that is not present for the lower drive fields. Possibly this is remnant of the much larger peak that was expected at the transition.

Data for the in-phase (loss) signal is shown in Figures 6.23 and 6.25. There is a step at the transition, much like the step in the out-of-phase signal. It is obviously associated with the latent heat of the first order transition. Also present is a peak in the data occurring at temperatures below the transition point. This peak has been associated with the irreversibility line [21]. It is clear from Figure 6.25 that the data is consistent with this idea. Here the peak is seen to shift to lower temperature as the frequency drive field reduced. The dc measurement of the irreversibility line at 20 kOe is 75 K [19].



Figure 6.23: The vortex melting transition in the $YBa_2Cu_3O_{6.95}$ sample Up2 at various dc fields.



Figure 6.24: The vortex melting transition in the $YBa_2Cu_3O_{6.95}$ sample Up2 at various drive fields.



Figure 6.25: The vortex melting transition in the $YBa_2Cu_3O_{6.95}$ sample Up2 at various drive frequencies.

Chapter 7

Conclusion

The primary goal of this thesis was to develop a high sensitivity ac susceptometer to measure the nonlinear Meissner effect in the high temperature superconductor $YBa_2Cu_3O_{6.95}$. In this regard the work to date must be considered unsuccessful and the goal presently unfulfilled. The susceptometer has a field dependent background that is significantly larger than the effect to be measured. It should be possible to subtract the background signal from the sample data, but this requires a reliable determination of the background which was never obtained.

Measuring the background with the sample in situ requires heating the sample above $T_c = 93$ K. At this temperature the response of the pick-up coil is significantly altered by some heating effect that makes this approach unsuitable. The only option was to measure the background by removing the sample, a very poor technique that was not reproducible to the desired level of resolution. So, not having sufficiently accurate knowledge of the background rendered the ac susceptometer useless for measuring $\lambda(H)$ with any degree of precision. In fact, the only quantitative result that could be gathered about the nonlinear Meissner effect in YBa₂Cu₃O_{6.95} is that $\Delta\lambda(H_{c1})$ is less than the 30 Å estimated by Xu, Yip and Sauls.

Experiments on the vortex melting transition and the temperature dependence of the penetration depth of $YBa_2Cu_3O_{6.95}$, however, present quite a different view of the susceptometer. At a fixed field, very precise, state-of-the-art measurements can be made with this device. Ideally an ac susceptometer can be used to make measurements along any path in the H-T plane. It can be said then that a susceptometer was developed having a far greater sensitivity when measuring along field contours in the H-T plane. This is a more balanced conclusion to this thesis. It acknowledges the merits of the device, but does not hide its failures.

It should be noted, that an experiment to measure the nonlinear Meissner effect is still pertinent, and so the motivation to make further improvements to the design of the ac susceptometer still exists. For the present ac coil set it may be possible to thermally shield the secondary coils from the sample holder, thereby allowing for the in situ determination of the background. If the coil set is to be rebuilt, the design should include a sapphire coil form that replaces the phenolic paper coil form of the version I coil set. Here the secondary coils would be wound onto the form and good thermal contact at low temperatures would be assured. Also the inaccuracies that arose from winding the secondaries on teflon forms would be greatly reduced using a rigid form again.
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Appendix A

Complex Susceptibility

1. It can be shown that the definition of χ from equation 2.4 is equivalent to the definition $\chi = dM/dH$.

Assume a general expression for the sample magnetization

$$M(t) = \sum_{n=0}^{\infty} M_n \, e^{inwt},$$

where the M_n are independent of t. For an applied field $H(t) = H_{dc} + H_1 e^{iwt}$ evaluate the expression

$$\frac{\mathrm{d}M}{\mathrm{d}H} = \frac{\mathrm{d}M(t)}{\mathrm{d}t} \frac{\mathrm{d}t}{\mathrm{d}H(t)}$$
$$= \sum_{n=1}^{\infty} n M_n \, e^{i(n-1)wt} / H_1. \tag{A.1}$$

The left hand side of this equation must necessarily be a periodic function, call it $\tilde{\chi}(t)$ and write it as the expansion

$$\tilde{\chi}(t) = \sum_{k=0}^{\infty} \tilde{\chi}_k \, e^{ikwt},$$

or by relabelling the terms as the equivalent series

$$\tilde{\chi}(t) = \sum_{l=1}^{\infty} \tilde{\chi}_l \, e^{i(l-1)wt}. \tag{A.2}$$

Comparing terms of A.2 and A.1 gives

$$\tilde{\chi}_n = \frac{nM_n}{H_1},\tag{A.3}$$

for $n = 1, 2, 3 \cdots \infty$.

Redefining $\chi_n = \tilde{\chi}_n/n$ (which seems to be convention [1-2]) returns the result Chapter 1. The dc field was explicitly included here.

2. Throughout this thesis the applied magnetic field was defined as

$$H(t) = H_1 Im(e^{iwt}) = H_1 \sin wt.$$

There is, of course, no reason why it could not be defined as

$$H(t) = H_1 Re(e^{iwt}) = H_1 \cos wt$$

where Re() denotes the real part. Redefining H(t) only shifts the time origin and does not change the physical reality of the sample's magnetization.

Here the magnetization would be expressed as

$$M(t) = H_1 \sum_{n=1}^{\infty} \operatorname{Re}(\kappa_n e^{inwt})$$

= $H_1 \sum_{n=1}^{\infty} (\kappa'_n \cos nwt + \kappa''_n \sin nwt),$ (A.4)

with the complex susceptibility $\kappa_n = \kappa_n' - i \kappa_n''$ given by

$$\kappa'_{n} = \frac{w}{\pi H_{1}} \int_{\frac{-\pi}{w}}^{\frac{\pi}{w}} M(t) \cos nwt \, \mathrm{d}t,$$

$$\kappa''_{n} = \frac{w}{\pi H_{1}} \int_{\frac{-\pi}{w}}^{\frac{\pi}{w}} M(t) \sin nwt \, \mathrm{d}t.$$
(A.5)

To compare this definition of the susceptibility with that of equation 2.4, it is easiest to rewrite A.4 as

$$M(t) = H_1 \sum_{n=1}^{\infty} \operatorname{Im}(i\kappa_n e^{inwt}), \qquad (A.6)$$

and then rewrite 2.3 in a new temporal variable au

$$M(\tau) = H_1 \sum_{n=1}^{\infty} \operatorname{Im}(\chi_n e^{inw\tau}),$$

which is equivalent to

$$M(t) = H_1 \sum_{n=1}^{\infty} \operatorname{Im}(\chi_n e^{in(wt + \frac{\pi}{2})}),$$
(A.7)

as $w\tau = wt + \frac{\pi}{2}$ is the transformation between the two time scales. From A.6 and A.7 one gets the relation $\chi_n = i\kappa_n e^{-in\pi/2}$, for all n = 1, 2, 3, Writing $i = (-1)^{\frac{1}{2}}$ and recognizing that for this sequence the quantity $e^{-in\pi/2} = (-1)^n (i)^n = (-1)^{3n/2}$, this can be transposed into the more appealing form

$$\chi_n = (-1)^{(3n+1)/2} \kappa_n. \tag{A.8}$$

The definition of the fundamental susceptibility remains unchanged. This not true in general for the higher harmonics, though for all $n, |\chi_n| = |\kappa_n|$. The third harmonic (which is often studied [10, 11]) changes sign under the alternate definition of H(t), and the real and imaginary parts of even harmonics are interchanged. Heeding Ishida and Goldfarb [5], one must keep these relations in mind for interlaboratory comparisons of the harmonic susceptibilities as well as for theoretical calculations.