Instrumentation for ac magnetic susceptibility measurements

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Magnetic susceptibility of a given sample can be measured using a mutual inductance bridge method. This is based on the principle that the voltage induced in a coil system which consists of primary and secondary coils is proportional to the magnetic susceptibility of the material introduced into the coils. A detailed description of the coil assembly, cryostat, temperature measurement, control and calibration of the apparatus developed indigenously to perform accurate measurements of magnetic susceptibility from room temperature down to 2 K has been described.

1 Introduction

Magnetic susceptibility measurement by means of an ac mutual inductance bridge is common practice in low temperature research. This is used for magnetic thermometry also. The basic form of the conventional ac mutual inductance bridge was first introduced by Hartshorn in 1925. Its application for the measurement of the ac magnetic susceptibility was first proposed and carried out by Casimir et al[1] In this method, the sample is placed inside the secondary of a mutual inductor consisting of a primary coil wound coaxially with a secondary coil. The secondary is usually divided into two identical solenoidal sections wound in opposition to each other and connected in series. With no sample in the coils, the mutual inductance of the combined coils should then be zero. With a sample in one of the secondary coils, the emf induced in the secondary circuit depends upon the magnetization, and hence upon the susceptibility, of the sample. Hence there will be an unbalance voltage at the secondary output. In order to balance the Hartshorn bridge over a range of sample susceptibilities, a compensatory variable mutual inductor is placed in series with the sample coil. If the mutual inductance of the variable mutual inductor is measured with and without the sample in the coil, the difference between these values would then be directly proportional to the ac magnetic susceptibility of the sample.

In its ideal form, the mutual inductance bridge is represented schematically as in Fig. 1. An alternating current I is passed through the primary. In the absence of a specimen, the bridge is balanced. The specimen is then introduced from outside to the midpoint of the top section of the secondary thus causing an unbalance signal to appear at the secondary output terminals. If an actual bridge could be represented in this simple manner and all the circuit constants could be accurately determined, it could serve as an absolute instrument in one of two ways:

1 The output voltage of the secondary could be taken as a measure of the magnetic moment induced in the specimen and the susceptibility calculated in terms of the exciting current I and the constants of the bridge circuit. The bridge used in this way could also serve as a dynamic or off balance instrument by following the changes in output voltage.

2 On the other hand, the bridge could also serve as an absolute null instrument, if the voltage were compensated by means of a calibrated and properly adjusted auxiliary mutual inductance.

An actual bridge, however, falls far short of being represented by such an ideally simple circuit, because of several factors: In the absence of a specimen, the bridge must be balanced to a high degree because the effective mutual inductance of a typical specimen is such an extremely small fraction of the main mutual inductances[2] + M and − M. The unbalance arises be-
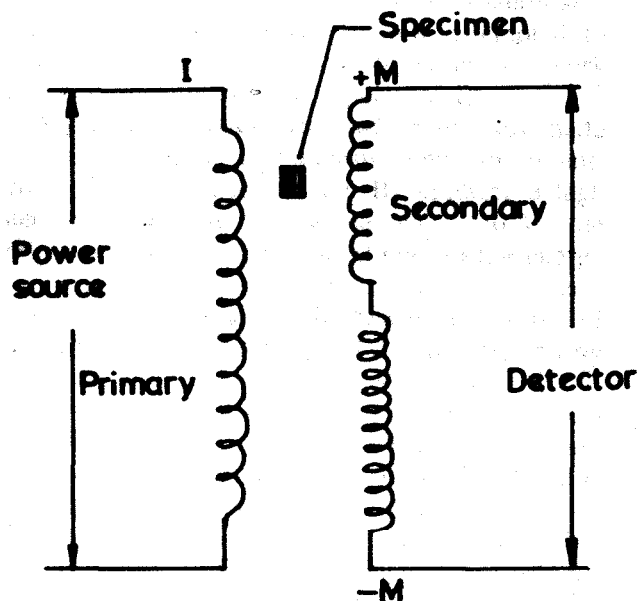

Fig. 1—Simplified schematic diagram of susceptibility bridge

cause in practice the two halves of the secondary cannot be made exactly identical in all respects. Further the finite dimensions of the coil lead to a certain degree of nonuniformity of the ac magnetic field. Hence, even when there is no sample in the coil, it is necessary to cancel this unavoidable imbalance between the two sections of the secondary coil. Further, the unbalance signal consists of a reactive as well as a resistive component, since the coil has an inductance and a distributed capacitance as well as a resistance (eddy current losses). The reactive component is in quadrature with the primary current while the resistive component is in phase with it. Besides, these components are a function of frequency of the ac primary current and the temperature of the coil.

For the above reasons, it is impossible to make accurate absolute calculations of the ac susceptibility of a sample. However, it may be used as an accurate comparative instrument, in one of two ways. First, it may be used as a dynamic instrument when the output voltage is calibrated by means of a standard specimen whose susceptibility has been determined absolutely. Secondly, it may be used as a null instrument when the compensator is calibrated by means of such standards.

A mutual inductance bridge suitable for magnetic relaxation measurements at low temperatures has been described recently by P.H. Muller et al[3] They used a Philips synthesizer PM 5190 as a signal generator. The primary coil is driven directly, without the use of any transformers. The output of the secondary coils is detected by a two-phase lock-in-amplifier (Ithaco dynatrac 393). The outputs of the detector are monitored with two panel meters, which are connected to the on-line computer system (LSI 11). They made no attempt to null the bridge by an additional compensation network. The signal change induced by the sample was monitored directly. They measured the frequency dependence of magnetic susceptibilities x' and x'' using this experimental set-up.

A similar system has been used in our laboratory during the past 8 years and the construction and performance of the system are reported in this paper.

2 Details of the Coil Assembly

A primary coil of 3000 turns of insulated copper wire was wound on a perspex former. The length, inner and outer diameters of the former are 60 mm, 8 mm and 10 mm respectively. Two identical secondary coils, each having 1200 turns, were wound on top of the primary coil. While winding the coils vacuum grease was applied on top of each layer so that the windings would be fixed firmly in position.

After winding the coils, the primary coil was energized by sending an alternating current at a frequency of 83 Hz using the internal oscillator of a lock-in-amplifier (PAR 5201). Thus an alternating magnetic field was produced in the primary coil and the induced voltage at the secondary coils was fed into the lock-in-amplifier in differential mode. The dc output of the lock-in-amplifier is proportional to the difference of the voltages that are induced in the two secondaries. The advantage of the differential mode is that the noise gets eliminated from the secondaries. Since the same number of turns were wound in both the secondaries, ideally the differential output of the lock-in-amplifier is expected to be zero. But since it cannot be ensured that the geometry of the windings is exactly the same, a small residual output voltage appeared across the secondary even in the absence of a sample. The output of the lock-in-amplifier was made zero by removing some of the turns from one of the secondaries, while the lock-in-amplifier was operated with a sensitivity of 10 µV.

3 Cryostat

A simplified diagram of the cryostat is shown in Fig. 2. The sample coil as described in Sec. 2 was embedded inside a copper block with a µ-metal shield separating them. It was found that in the absence of the
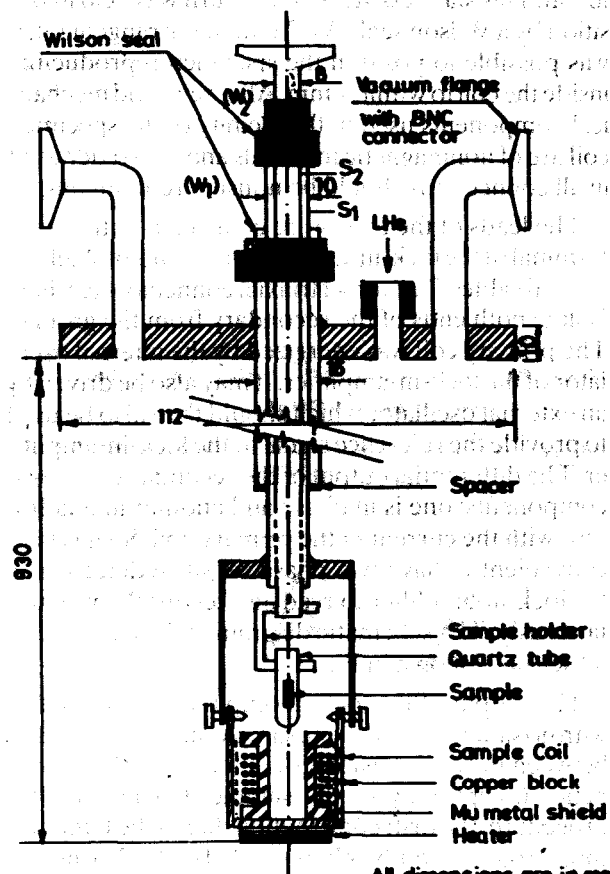

Fig. 2—Sketch of the cryostat

μ-metal shield, the effect of the eddy currents due to copper gives rise to a signal, which masks the signal due to the specimen. This effect was completely eliminated by providing the μ-metal shield. However, it is possible to use nonmetallic supports to the coil assembly in order to eliminate this problem. Such a system is now under construction in our laboratory. This would also facilitate measurements of the ac magnetic susceptibility in the presence of an applied dc magnetic field. A superconducting solenoid could be used to support the coil system axially mounted inside it.

The copper block was then attached to a long thin-walled nonmagnetic stainless steel tube (S_1) of inner diameter 10 mm which is held in position by a Wilson seal (W_1) near the top flange of the glass dewar containing liquid helium. Thus the copper block could be lifted above the liquid helium level inside the dewar by about 6 to 8 cm. This facilitated the temperature of the copper block to be varied without boiling off the liquid helium. This arrangement is particularly useful in repeated monitoring of superconducting or magnetic transitions by warming up or cooling down the sample. The specimen was inserted into or removed from the sample coil by attaching the specimen holder (quartz tube) to another thin-walled stainless steel tube (S_2) of 8 mm inner diameter by means of a strip of teflon. This stainless steel tube in turn was held in position by a Wilson seal (W_2). With this arrangement, it was possible to position the specimen reproducibly inside the coil to within 1 mm. All screws and mechanical components used in the vicinity of the specimen coil are of nonmagnetic materials and the solder used in all connections should be nonsuperconducting.

The leads of the coils were connected to the input terminal of the lock-in-amplifier by means of shielded coaxial cables. The use of triaxial connectors can help isolate both ends of the secondary from the ground. The primary coil was energized by the internal oscillator of the lock-in-amplifier. It may also be driven by an external oscillator which should then also be used to provide the reference signal to the lock-in-amplifier. The differential output of the secondaries has two components: one is in phase and another in quadrature with the current in the primary coil. So it is very convenient to have two phase sensitive detectors in the lock-in-amplifier to read and record them simultaneously. The schematic diagram of the experimental set-up is shown in Fig. 3.

The output of the lock-in-amplifier was recorded with no sample and with the sample and the difference between these two readings is a measure of the magnetic susceptibility of the sample. Because of this procedure, the drift contribution due to the temperature variation will be eliminated. To check whether any contribution comes from the quartz tube, the sus-
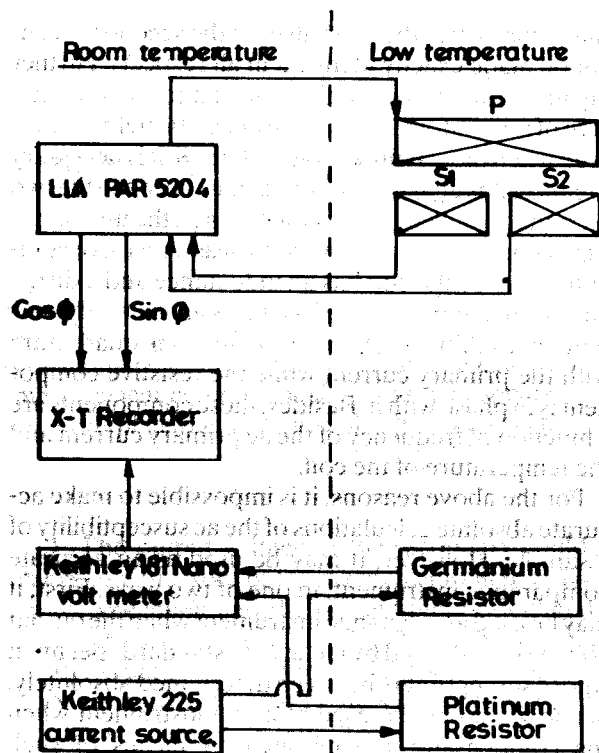

Fig. 3—Schematic diagram of ac susceptibility experimental set-up

ceptibility of the empty quartz tube was measured from room temperature down to 4.2 K and no detectable output was observed. Instead of quartz, teflon may also be used for the construction of the sample holder. The signal output at the secondary is also proportional to the filling factor which may be defined as the ratio of the sample volume to the volume of the secondary coils. In order to maximize this factor, it is advisable to wind the secondary coil inside the primary, i.e. as close to the sample as possible.

4 Temperature Measurement and Control

A constantan heater wire (20 ohms), connected to a relay and a dc power source and wound around the bottom of copper block, enabled the block to be heated. In the case of a helium bath cryostat, pumping the bath with a sealed rotary pump enables the temperature to be reduced down to about 1.2 K. Alternatively, the measurements may also be carried out inside a liquid helium dewar itself. A Leybold temperature controller (model ER 3) with a calibrated thermistor or a carbon resistor as the temperature sensor activates the relay and regulates the temperature of the block to better than 0.1 K. The temperature of the sample was measured using a calibrated germanium resistance thermometer (GRT) below 35 K and a calibrated platinum resistance thermometer (PRT) above 35 K. Carbon glass thermometers have to be

used when a large dc magnetic field is also present. These thermometers were mounted in the vicinity of the sample. A Keithley model 225 current source and a Keithley model 181 nanovoltmeter were employed to measure the resistance of the thermometers. Recently a PREMA model 6000 digital multimeter has been used for the direct temperature readout with a standard PT 100 platinum sensor.

5 Calibration of the Set-up

The difference between the output voltages with and without the sample is directly proportional to the susceptibility of the sample. To find out the constant of proportionality a material whose absolute susceptibility is well known was employed. For this purpose a powder sample of Gd_2O_3 was chosen whose susceptibility variation with temperature is known[4-6]. The inverse of the measured output voltage at a frequency of 83 Hz due to Gd_2O_3 is shown as a function of temperature in Fig. 4. It can be seen from the figure that the graph is a straight line indicating that the constant of proportionality is the same throughout the temperature range from room temperature down to 4.2 K. Using this calibration constant, the susceptibility of $Fe(NH_4SO_4)_2 6H_2O$ (Mohr salt) was measured and is shown in Fig. 5. The measured susceptibility of the Mohr salt agrees well with the value reported in the literature[7].

6 Detection of Superconducting Transitions

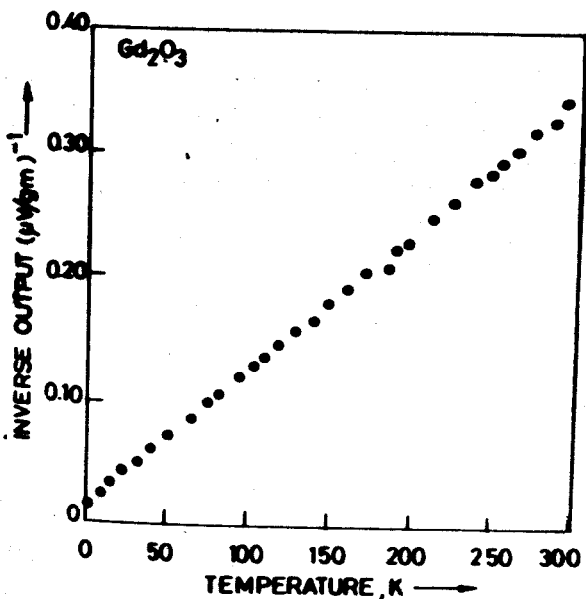The ac magnetic susceptibility provides a contactless method of detecting superconducting phase transitions. In contrast to a resistive method, this method does not suffer from the disadvantage of not being able to distinguish between bulk superconductivity and a superconducting percolation path across an otherwise normal material. It is based on the establishment of diamagnetic screening currents within the surface layer of the superconductor of thickness equal to penetration depth. However, it must be borne in mind that this is not a measurement of the Meissner effect which alone is a truly bulk effect arising from the expulsion of magnetic flux from within the bulk of the material.

A typical superconducting susceptibility transition is shown in Fig. 6 for the high temperature oxide superconductor $GdBa_2Cu_3O_{7-y}$. This corresponds to the dispersive component (i.e. real part) of the complex ac susceptibility. The volume fraction of superconducting material in the specimen may be estimat-
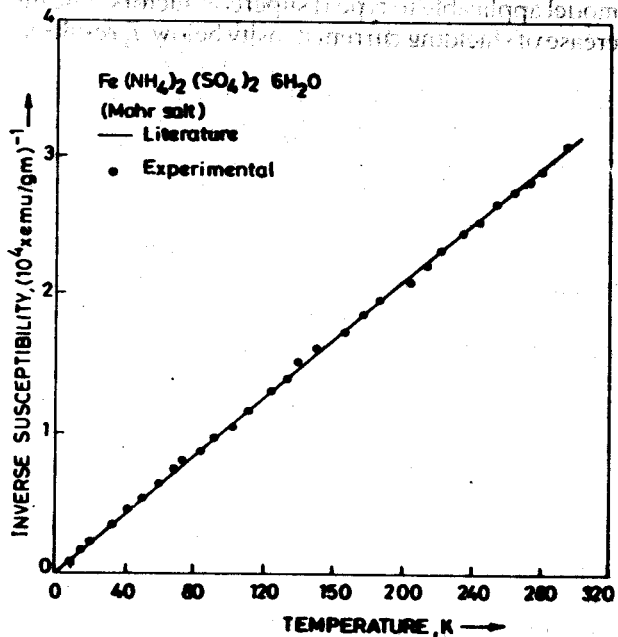

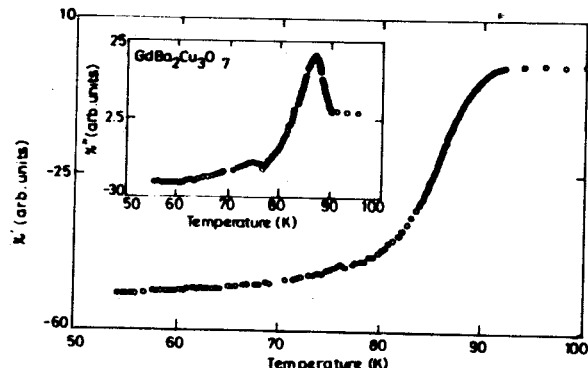Fig. 5—Inverse susceptibility of the sample $Fe(NH_4SO_4)_2 6H_2O$. (Mohr Salt)

Fig. 6—Dispersive and absorption (inset) components of the susceptibility of the superconducting compound $GdBa_2Cu_3O_{7-y}$

Fig. 4—Inverse output of the lock-in-amplifier due to the sample of Gd_2O_3 vs temperature

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ed by comparing the signal output with that of a spherical sample of Pb in which the signal may be assumed to correspond to an ideal diamagnetic susceptibility of $3/8\pi[1/4\pi$ (in cgs units) $\times$ correction factor to take into account the demagnetization due to sample shape].

However, in porous granular specimens, the porosity factor and the shape of the grains render this estimate somewhat unreliable. It must also be remembered that this expression is correct only if the grain size is large compared to the penetration depth. When this is not the case, partial or complete flux penetration may take place, leading to a reduction of the diamagnetic signal strength.

In the inset of Fig. 6 is shown the absorption component (χ'' i.e. imaginary part) of the complex susceptibility. The temperature dependence explained by Gomory and Lobotka[8] in terms of a critical state model applicable to type II superconductors. The increase of shielding current density below T_c results in
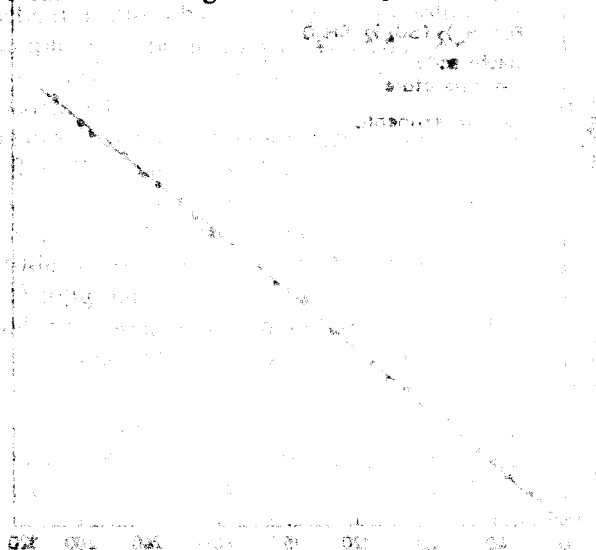
flux motion, leading to energy absorption and a maximum at a temperature below T_c. Absorption measurements are also very useful in the study of paramagnetic relaxation or domain relaxation in magnetically ordered materials and in spin glasses.

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