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An AC magnetic susceptibility bridge and cryostat for the temperature range 2–300 K

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Abstract. A mutual inductance bridge to measure low frequency magnetic susceptibilities of magnetic materials has been constructed. Salient features of the bridge, which uses a variable mutual inductance simulated using operational amplifiers, the cryostat and the coil assembly are described in this paper. The apparatus has been employed for accurate measurement of superconducting transition temperatures and for sensitive detection of magnetic ordering transitions in liquid helium and liquid nitrogen temperature ranges respectively. The bridge has been calibrated to determine the static susceptibility of magnetic materials as a function of temperature.

Keywords. AC magnetic susceptibility; mutual inductance bridge; superconducting transitions; magnetic ordering.

1. Introduction

The method of measuring the complex AC magnetic susceptibility of magnetic materials using a mutual inductance bridge was originally described by Casimir and DuPré (1938). In this method, the magnetic specimen which is kept inside a sample holder of, say, quartz, forms the core of a transformer through the primary of which an alternating current is passed. The resulting oscillating magnetic field of the form

$$H = H_0 + H_1 \exp (i \omega t),$$

produces a magnetisation which may be represented by:

$$M = M_0 + \chi H_1 \exp (i \omega t),$$

where $\chi = \chi' - i \chi''$, $\chi_0 = M_0 / H_0$

being the static susceptibility (Morrish 1965). The introduction of the magnetic specimen into the transformer will therefore cause a voltage to be induced at the secondary which is given by (Sydney *et al* 1976)

$$V = N H_1 \omega [(\mu_0 + \xi \chi') \sin \omega t - \xi \chi'' \cos \omega t].$$

Here N is the number of secondary turns, H_1 is the amplitude of the applied alternating magnetic field of frequency ω and ξ is a filling factor, of the order of unity.

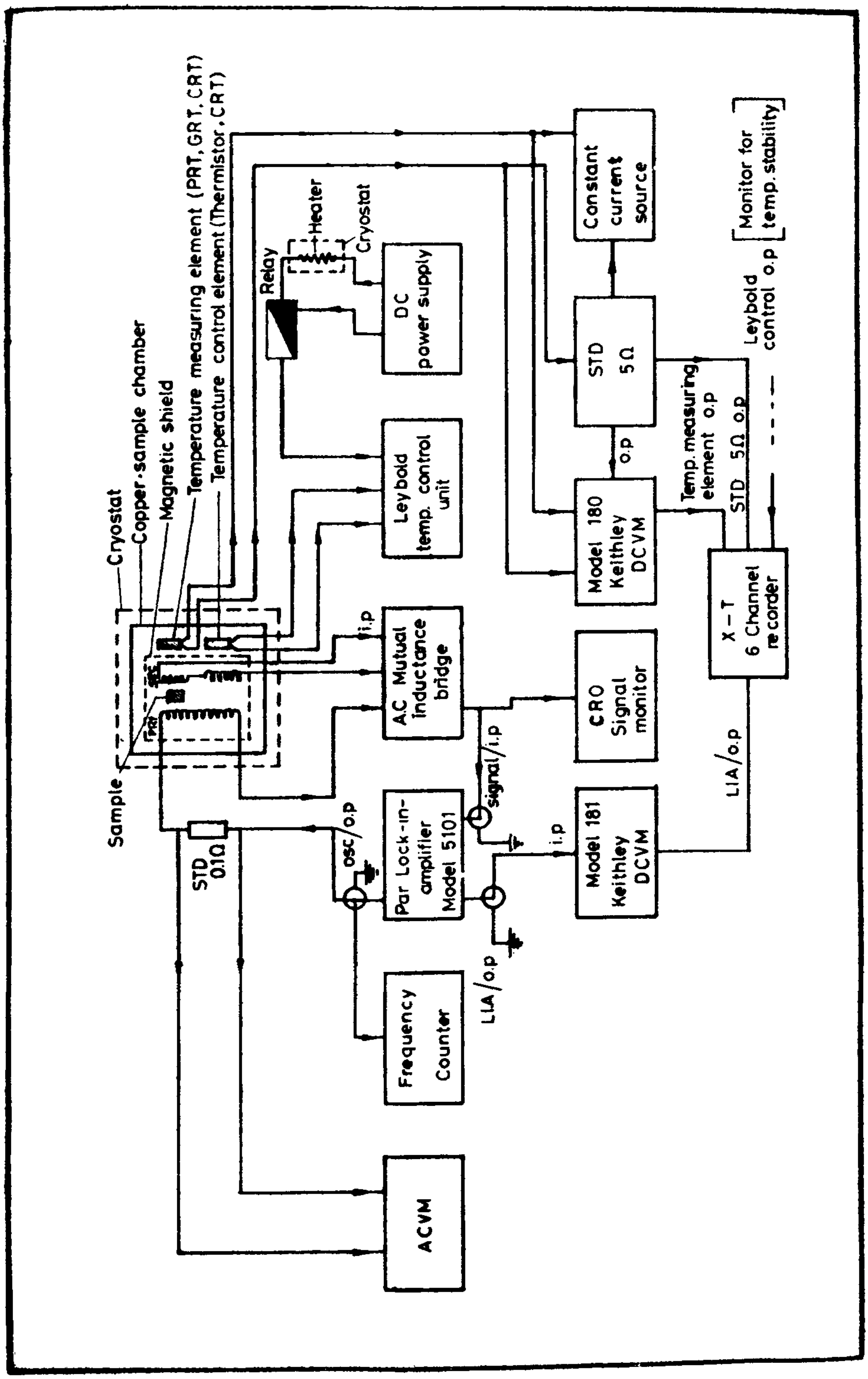


Figure 1. AC magnetic susceptibility bridge block diagram.

Using a phase-sensitive detector, these components can be measured and related to the real and imaginary parts of the susceptibility.

2. Description of apparatus

2.1 *The mutual inductance bridge*

For maximum sensitivity in the method described above, it is essential to have a zero output at the secondary of the transformer in the absence of the magnetic specimen. This is usually achieved by splitting the secondary coil into two balanced sections which are wound antiphase. The specimen is then inserted into one half of the coil. It is however found that for specimens with widely differing magnetic susceptibilities, this balancing method is not satisfactory. An additional balancing mechanism is provided by including a variable mutual inductance (Casimir *et al* 1939) or a ratio transformer (Maxwell 1965) in series with the sample coil. In addition, a resistive balancing network is also required (Erickson *et al* 1954). But this has the disadvantage that the resistive network directly connects the primary and the secondary cir-

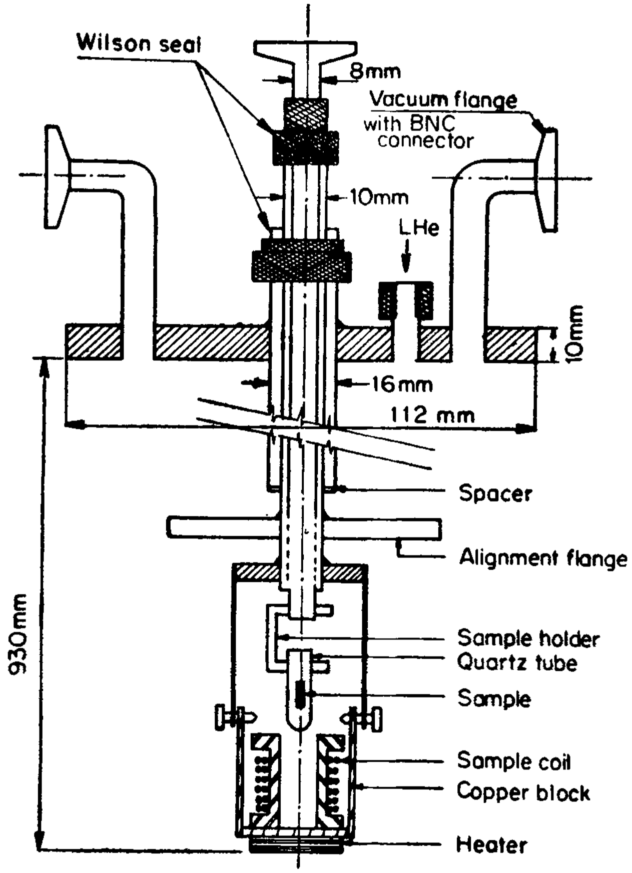


Figure 2. Cryostat and sample assembly.

cuits. As a result, the operation of the mutual inductance bridge becomes difficult, especially when employed over a wide frequency and/or temperature range. The availability of operational amplifiers has led to an elegant solution of this problem through a simulated variable mutual inductance and such a circuit has already been described by Brodbeck *et al* (1978). We have adopted this method in building our bridge, a simplified schematic of which is shown in figure 1. The null signal at balance is displayed on an oscilloscope and further sensitivity for null detection is obtained by using a PAR model 5101 lock-in-amplifier.

2.2 *The cryostat and coil assembly*

A simplified diagram of the cryostat is shown in figure 2. The measuring coil is wound on a former of PTFE of diameter 3 cm and length 8 cm, which fits closely around a quartz tube of outer diameter 6 mm. The primary of the mutual inductance coil has a continuous winding of 3000 turns of SWG 38 copper wire. The two halves of the secondaries are of 1200 turns each, of the same copper wire. The specimen coil is embedded inside a copper block, with a mu-metal shield separating them. The copper block is then attached to a long thin-walled stainless steel tube of inner diameter 10 mm which is held in position by a Wilson seal near the top flange of the glass dewar containing liquid helium. Thus the block inside the dewar can be lifted above the liquid helium level by about 6 to 8 cm; this facilitates the temperature of the block being varied without boiling off the liquid helium. This arrangement is particularly useful in repeated monitoring of superconducting or magnetic transition temperatures by warming up or cooling down the sample.

The specimen is inserted into or removed from the transformer coil by attaching the specimen holder to another thin-walled stainless steel tube of 8 mm inner diameter by means of a strip of teflon. This stainless steel tube in turn is held in position by another Wilson seal. With this arrangement it is possible to position the specimen reproducibly inside the coil to within 1 mm. All screws and other mechanical components used in the vicinity of the specimen coil are of non-magnetic materials.

A constantan wire heater (20 Ω), connected to a relay and a DC power supply, and wound around the bottom of the copper block, enables the block to be heated. A Leybold temperature controller which uses a calibrated thermistor and a carbon resistor actuates this relay and thus regulates the temperature of the block to better than 0·05 K. The temperature measurement is carried out by using (Lake shore Cryotronics Inc.) calibrated germanium/platinum thermometers. The voltage across the thermometer due to the passage of a constant current (delivered by a Keithley model 225 current source) is measured using a Keithley model 180/181 nanovoltmeter.

3. Calibration and performance

Since no special effort had been made to ensure a homogeneous field inside the specimen coil, it was essential to first locate a point in the sample space in the upper half of the secondary of the transformer at which the induced voltage due to the introduction of the specimen is a maximum. This would then ensure that maximum sensitivity for a given sample dimension was achieved and also that the specimen is moved between two positions which were precisely located. It was found that at a distance of about 24 mm from the top of the coil there was a region 8 mm in length over which the variation of the bridge output with respect to the position of the specimen was less than 1 %. This ensured that measurement errors due to variation in the position of the specimen were negligible, provided the samples are kept within this region during different measurement runs.

Since in the experiments described here, the frequency of the applied alternating magnetic field was chosen to be 74 Hz, the measured susceptibility would correspond essentially to the static value. This was also checked by the absence of a detectable in-phase induced voltage. The output of the mutual inductance bridge was measured by introducing a large number of paramagnetic substances of known static magnetic susceptibilities and standard dimensions into the coil. The results of these measurements are shown in figure 3 and it is found that the output is linear over a wide range of susceptibility values (10^{-5} — 10^{-3} emu/g) at room temperature. The slope of this graph gives the constant of proportionality between the bridge output and the magnetic susceptibility for the given set of operating conditions.

When the experiment was performed at different temperatures down to 77 K, it was found that the constant of proportionality was different at different temperatures. It changed by a factor of 2·33 around 180 K and by a factor of 1·6 at 77 K as compared with the room temperature value. These changes arise from the differential thermal contraction of the sample coil and the teflon former in this temperature interval.

Using a sample of pure Gd_2O_3 as the standard, the constant of proportionality was then determined at different temperatures between 77 K and room temperature.

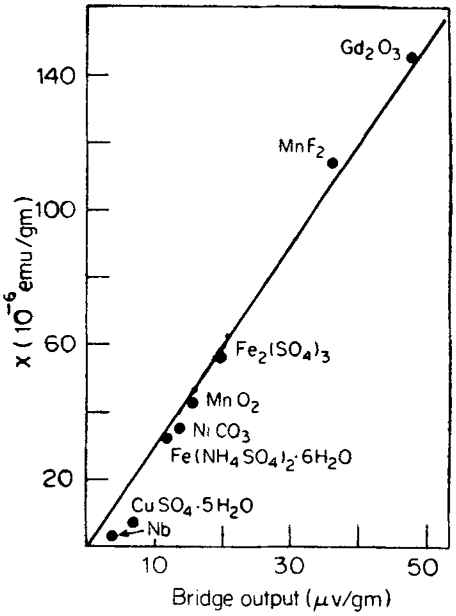


Figure 3. Measured bridge output *vs* static susceptibility of different paramagnetic materials.

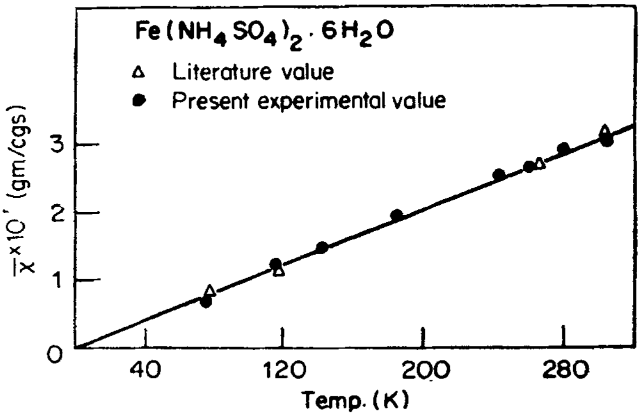


Figure 4. Temperature variation of the magnetic susceptibility of Fe $(NH_4SO_4)_2 \cdot 6H_2O$.

The temperature variation of the calibration constant was obtained by drawing a smooth curve through these experimental values. The magnetic susceptibility of $Fe(NH_4)_2(SO_4)_2 \cdot 6H_2O$ (Mohr salt) was next determined using this calibration curve. In figure 4 our measured values for Mohr salt are plotted as a function of the temperature and these are found to be in satisfactory agreement with the values derived from data reported in the literature (Martin 1967). For all further measurements, the calibration constant at a particular temperature was therefore read off from this calibration curve.

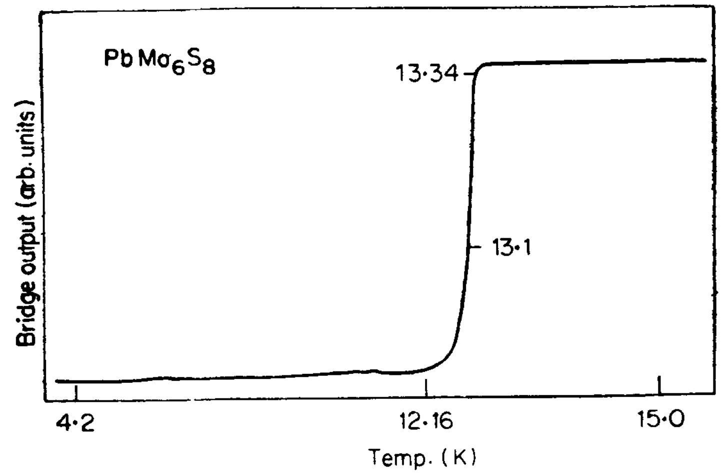
4. Test results

4.1 *Superconducting transitions*

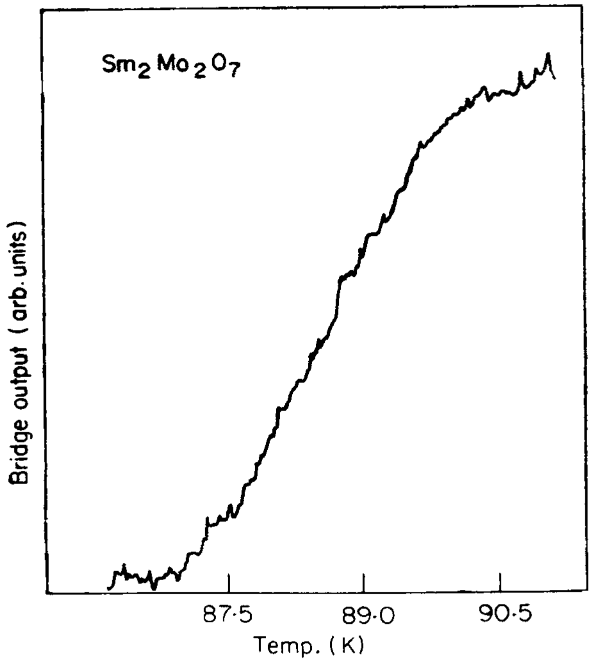
The superconducting state is characterised by a transition into a state of perfect diamagnetism. Hence, the magnetic susceptibility of any material which becomes super-conducting would change abruptly to a negative value at the transition temperature, thus providing a method of sensitively detecting the superconducting transition. As compared with the resistive method, this has the advantage (i) of being a contactless method (ii) of detecting the onset of superconductivity in the bulk material. We used a sample of 185·45 mg of a polycrystalline sintered pellet of the chevrel phase superconducting compound, $PbMO_6S_8$. The observed susceptibility transition at T_c is shown in figure 5a and a T_c value of 13·1 K ± 0·05 K, corresponding to the mid-point of the transition, was obtained, in good agreement with the reported value of 13·29 K (Delk and Sienko 1980).

4.2 *Magnetic transitions*

The rare earth molybdate, $Sm_2MO_2O_7$, belonging to the pyrochlore structure has been recently synthesised and found to be magnetically ordered (Subramanian 1982). The magnetic ordering transition in this compound is shown in figure 5b. The transition temperature, corresponding to an abrupt variation of the susceptibility, was found to be 89 K.



(a)



(b)

Figure 5a. Variation of susceptibility at the superconducting transition temperature of $PbMO_6S_8$.

Figure 5b. Variation of susceptibility at the magnetic transition temperature of $Sm_2MO_2O_7$.

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It is a pleasure to acknowledge valuable suggestions given by Prof. Schönberg, during his visit to IIT, Madras, regarding the constructional aspects of the cryostat and coil assembly. We would also like to thank Prof. R Srinivasan for his interest in this work and Prof. G V Subba Rao for providing the samples used in these measurements.

Note added in Proof: This value agrees well with the magnetic susceptibility measurements reported by Manthiram and Gopalakrishnan (Manthiram A and Gopalakrishnan 1980 *Indian J. Chem.* **19A** 1042)

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