APPARATUS FOR MAGNETIC STUDIES BELOW 1° K

by

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INTRODUCTION

Below 1° K the lattice of a solid is essentially in the zero point energy state and the particles are in a state of high physical order. Certain materials such as cesium titanium alum and manganese ammonium sulfate contain ions which exhibit paramagnetic behavior. These particular ions have unpaired electron spins so that there is a resultant net magnetic moment. Although the lattice is well ordered at 1° K, the individual magnetic moments are oriented at random in the crystal. The third law of thermodynamics requires that the absolute zero is a state of complete order, so that these electron spins must become ordered if the temperature is decreased sufficiently. This thesis describes the preliminary work involving new equipment and measuring devices which will be used in a study of the ordering processes which occur in the spin systems of certain paramagnetic ions very close to the absolute zero.

For example consider the case of manganese ammonium sulphate, Mn(NH₄)₂(SO₄)₂. 6H₂O. The important consideration is the fact that the manganese ions possess unpaired electron spins and consequently exhibit electron paramagnetism. The ammonium and sulphate radicals and the waters of crystallization do not take part in the paramagnetism but only serve to "dilute" the
magnetic ions. For the experiments at hand, the salt acts like a system of weakly interacting dipoles. Below about 0.2°K and for zero external field, the interaction of the dipoles is the dominant effect. This interaction is the driving force which causes the ordering guaranteed by the third law. The manner in which the dipoles interact determines the type and mechanism of the ordering. Antiferromagnetism, relaxation and remnant magnetic moments are some of the phenomena which can be observed as the ordering takes place.

In order to extend the temperature close to the absolute zero, equipment has been constructed for the process of adiabatic demagnetization. The process involves essentially two steps: first the sample at about 1°K is placed in a large external magnetic field; and second the thermal contact between the sample and the 1° temperature bath is broken and the external field is reduced to zero adiabatically. The addition of a large external field tends to align the spins and thus reduce the entropy. Spins which had equally populated the degenerate states must now redistribute themselves over the energy levels according to a Boltzmann distribution. The temperature of the bath and the large external field determine the distribution of the spin system over the energy states. The rearrangement which occurs during the application of the external field corresponds to a
condition of lower energy, so that energy is transmitted to the bath by the spin system in this part of the process. After a short time, equilibrium is obtained and the first part of the process is complete. The second step consists of pumping the helium exchange gas out of the sample chamber so that the sample chamber is thermally isolated from the constant temperature bath. Then when the external field is removed, a condition of constant entropy exists and the sample is demagnetized adiabatically. For the paramagnetic salts of interest, the demagnetization produces a marked cooling.

Part I of this thesis considers the construction and testing of a liquid nitrogen cooled solenoid magnet capable of furnishing the large fields necessary to the magnetic cooling process. Part II discusses the measuring equipment and the low temperature apparatus associated with the general problem at hand. And Part III presents some data obtained in the temperature range $4^\circ \text{K} - 1^\circ \text{K}$ using the apparatus discussed in Part II.
PART I. LIQUID NITROGEN COOLED SOLENOID MAGNET

Since the field of a solenoid depends directly on the current and the power dissipation from resistive heating depends on the square of the current, high field magnets require prohibitively large power supplies and large cooling facilities. A cryogenic magnet is one answer to the problem for applications where experiments can be performed in the relatively short period of about 30 minutes (determined by the capacity for coolant storage). The feasibility of magnets immersed in liquid nitrogen has been studied. A cryogenic magnet takes advantage of the reduced resistance of copper at low temperatures, so that smaller power supplies and cooling facilities are required to produce a given field. Existing systems built for continuous operation require magawatts of power and use kerosene coolant. Because the specific heat of a liquid coolant is relatively low, coolant flow rate must be large or a large temperature rise across the magnet must be tolerated. In the case of a cryogenic coolant, heat is carried away by the relatively large latent heat of vaporization. The magnet described in this thesis has produced a field of 36 kilogauss with the expenditure of only 60 kW. The magnet described in reference 4 above requires at least 8 times as much power to produce the same field. The initial installation and operation of the larger power plant is obviously expensive.
1. **Description and Discussion.**

The solenoid and related equipment are described and discussed in this section.

1.1 **Solenoid.**

The physical construction of the solenoid is described elsewhere \(^5\) but a brief description is given here in order to understand the cooling system. The solenoid contains six spiral coils of copper strip 2 inches wide by 0.016 inch thick which are insulated by nylon string wound around the copper strip about one turn per lineal inch. The string must supply electrical insulation without impeding the flow of coolant between the turns. Every second coil is wound in the opposite direction so that the sense of current flow about the cylindrical axis of the solenoid is the same in each coil. Current spirals inward in the first coil, passes through a cylindrical jumper to the second coil and then spirals outward. The remaining four coils are connected in series similarly. The coils are separated by \(\frac{1}{8}\) inch thick micarda spacers cut in the shape of a spoked wheel. The spacers must support the weight of the copper without blocking the flow of coolant. The coils are stacked on a vertical axis inside a centrifugally cast aluminum casting. A cast aluminum pan bolted to the solenoid case furnishes mechanical support for the coils and also serves as a plenum for supplying coolant to the coils. The coils are
2 inches I.D. by 11 inches O.D. and the solenoid case is about 19 inches long by 13 inches O.D. The general relation between the solenoid and its associated equipment can be seen in fig. 1.

1.2 Exhaust.

The maximum design input power of 100 kw will boil nitrogen at a rate of approximately 10 gallons per minute and will produce about 845 cubic feet of gas per minute at STP. The cold gas leaves the solenoid at about 77°K but it quickly mixes with the warm humid room air, producing large volumes of cold fog. The resulting moisture condensation and excess of nitrogen gas (and consequent lack of oxygen) are highly undesirable. Therefore a large capacity gas exhaust system is used.

An easily demountable plenum of corrugated paper board is placed around the solenoid to confine the cold gas somewhat. The plenum is exhausted through two 12 inch diameter flexible hoses by an 18 inch diameter squirrel cage blower mounted in a window near the solenoid. Since the top of the plenum is open to allow easy access to equipment, considerable room air is drawn into the blower along with the cold gas. The resultant mixing warms the gas stream before it enters the blower so that the blower does not have to operate at extremely low temperatures. The bearings are nevertheless purposely made loose at room temperature and a light weight lubricating oil is used.
The 2 horsepower drive motor is external to the blower and it is unaffected by the cold gas stream.

The blower seems easily capable of handling the volume of gas necessary even though it is probably not operating at highest efficiency. The flexible hose inlets are a great advantage because of their mobility but they are slightly smaller than the 16 inch blower inlet ports. The blower inlets are thus somewhat choked, but the exhaust is perfectly open. The blower RPM is slightly lower than the rated value but this is no great disadvantage. In the future as the solenoid passes out of the development stage, it may be desirable to increase the existing blower capacity by installing a duct of larger size and by choosing a new pulley for the drive motor.

1.3 Liquid Nitrogen Production, Storage, and Transfer.

After an initial cool down period of about 3 hours (depending on humidity and ambient temperature), a liquifier plant on site is capable of producing 2 tons or about 600 gallons of liquid nitrogen per 24 hour period. As a by product of oxygen produced for the local steel industry, liquid nitrogen is available locally in even larger quantities.

Since the nitrogen plant can not supply liquid nearly as fast as the solenoid requires, a reservoir is used. The liquid nitrogen is stored in a 375 gallon
aluminum tank surrounded by about 16 inches of Santocell insulation. The tank and outer hood is mounted on a farm wagon so that it is easily moved. The nitrogen is piped into the lab through a 1 inch diameter aluminum pipe covered in the most part with a 5 inch diameter styrofoam cover.

A vacuum jacketed tanker and supply line would be much more efficient. However, a vacuum supply line is too difficult to construct and alter during the early stages of a project when the designs are not always definite. In the future, vacuum lines are strongly recommended because the large quantity of gas coming through with the liquid in the present lines indicates a fairly high loss factor. However, a vacuum jacket for the tanker is prohibitively expensive and a seam split in transit is both difficult and expensive to repair. Although there are no quantitative data on the efficiency of the tanker, it seems to be good enough. Unused nitrogen from a test run remained in the tanker for nearly a week.

In initial tests it was sufficient to simply plug the tanker pressure release port and let the pressure due to normal boiling force the liquid through the transfer line. However, in an extended run the natural boiling can not maintain pressure in the tank as the liquid level decreases, so a bank of dry nitrogen cylinders, a manifold and a pressure regulator are installed. In all tests so
far a pressure of 15 PSI gauge supplied through a 3/8 inch line is sufficient to force liquid nitrogen at a large enough rate.

In order to maintain pressure in the tanker, the gas pressure system must supply enough gas to make up for the volume of liquid forced out. Assuming the gas leaves the cylinders at room temperature and there is no pressure drop along the gas line, the maximum flow rate through the pressure regulator corresponds to 6.5 gallons per minute at nitrogen temperature. The maximum liquid flow rate used so far is about 6.2 gallons per minute, for which the tanker pressure was successfully maintained during the run. The four gas cylinders on the manifold contain more than twice the volume of gas necessary to maintain a pressure of 30 PSI in the tanker (the tanker is tested to 46 PSI) but the regulator and 3/8 inch gas line are operating at almost maximum flow rate. If the solenoid is run at higher power, a second regulator and a larger gas supply line should be considered.

1.4 Nitrogen Pump and Recirculation System.

The overall cooling system is shown schematically in fig. 1. The liquid nitrogen is forced up through the annular spaces between the windings where part of the liquid is vaporized by the resistance heating. The remainder of the liquid spills over the top of the solenoid casing.
into a large stainless steel dewar vessel 18 inches in diameter by 36 inches long. A 2 inch copper pipe extending well below the liquid level in the dewar serves as an inlet for the recirculation pump and a 1 inch pipe connects the pump with the plenum at the bottom of the solenoid casing. To replace the coolant boiled away, liquid nitrogen is forced from the tanker through an insulated regulating valve into the intake line of the pump.

The pump (type C2X40) is on loan from Air Products Incorporated. A 2 horsepower motor drives the centrifugal type impeller at about 3500 RPM and thus produces a pressure of 12 PSI for a flow rate near 25 gallons per minute. The pump bowl is mounted in a large box of glass wool on a vertical axis. The impeller at nitrogen temperature is connected to the motor at room temperature by a shaft partially in contact with a stream of cold gas from boiling in the pump. The shaft is held in place by one graphite bearing in the pump bowl and by another bearing near ambient temperature. The lower bearing is lubricated by liquid nitrogen.

The glass wool insulated pump and styrofoam insulated lines constitute enough heat leak that it is necessary to continuously prime the pump. Since the pump intake line leads over the top of the large dewar
and then down to the pump, gas easily collects at the top of the inverted "U", thus breaking the liquid column. The impeller is quite loose in the pump bowl and will not pump unless the bowl is completely filled. Therefore the inlet line from the tanker empties directly into the pump intake line below the peak so that opening the transfer valve automatically primes the pump and then spills any excess liquid back into the dewar.

Tests indicate that a current larger than 1200 amperes makes the solenoid highly unstable. The corresponding power input of about 60 kw is only 60% of the 100 kw considered in the design calculations but the resultant field is 80% of the expected maximum at 1500 amperes. The transfer of heat to a liquid boiling in narrow channels as studied by Sydoriak and Roberts has a direct bearing on the effectiveness of the coolant and the resultant maximum field. A cryogenic liquid was forced through thin annular channels in a heater, closely approximating the conditions in a solenoid. It was found that there is a critical heat input depending on channel size above which the heater temperature will become unstable and rise uncontrolled. The instability seems to occur when the bubbles become large enough to completely block the flow of fresh coolant. Whereas the normal measure of effective heat transfer to a fluid is the surface area of contact, it is found that the cross section
of flowing coolant is the most important parameter for the case of boiling in narrow channels. For the solenoid at hand, the recirculation pump is used to facilitate cooling by forcing the gas out of the windings ahead of a large excess of coolant.

In any future designs the present cooling system should be made more efficient, perhaps by applying some of the suggestions to follow. The insulation between turns of the coil is nylon string about 0.010 inch in diameter. The string is carefully wrapped so that adjacent turns are separated by only one layer of insulation, thus allowing more turns per coil. At high field the radial force between conductors tends to compress the nylon string, allowing some turns to short and narrowing the cooling channels. Any modifications should include wider cooling channels at the expense of a few turns.

The solenoid windings are mechanically strong and relatively simple to construct but the resultant design is not optimized for maximum coolant flow. From the description in section 1.1 it is evident that the channel cross section through successive coils is alternately a right or left handed spiral. The resultant coolant path therefore causes turbulence and offers the possibility of trapped pockets of gas. The increased back pressure greatly limits the flow from the pump.
It is possible to wind the solenoid in a slightly different manner to reduce the resistance to coolant flow. Wind one layer of turns along the complete length of the solenoid, apply long thin spacers parallel to the axis of the solenoid and repeat for successive layers. If the separation between spacers is chosen properly and the coils are wound tightly enough, this design can be made mechanically as strong as the present solenoid. The real advantage is the complete lack of transverse obstructions in the cooling channels.

2. Field Calibration.

During previous testing\(^5\), the solenoid field was measured with a DynaLab Model D-79 gaussmeter reading up to 30 kilogauss. This meter measures the transverse voltage across a piece of semiconductor material carrying a current in the longitudinal direction (Hall effect). It is well known that the calibration is strongly affected by temperature so the flux probe was thermally shielded from the cold solenoid by a dewar vessel and the meter calibration was checked before and after each measurement. Even with these precautions the meter gave misleading results. The experimental points fall below the expected linear law for field values above 10 kilogauss as shown in fig. 2. However, the low field measurements lie on a straight line. From these considerations, the measurements
were put in question and an alternative test was sought.

The most direct field measurement involves a measuring coil and ballistic galvanometer. As the coil is quickly pulled out of the field, the change in flux through the coil induces a short current pulse and the galvanometer is ballistically deflected in direct proportion to the change in flux. The constant of proportionality can be easily determined by measuring the deflection due to a known field. The range of field to be measured is large enough to require a scale factor adjustment. A galvanometer shunt is not suitable because somewhat uncertain damping factor corrections must be applied. Therefore two calibration standards and two measuring coils are used.

The field measurements lie in three principle regions: low field 0-400 gauss, medium field 6,000-9,000 gauss, high field above 9,000 gauss. In the low field region the solenoid can be run for long periods of time at room temperature because external cooling is not necessary for a dissipation of about 80 watts. The necessary current of approximately 15 amperes can be supplied either by storage batteries or by a small DC generator. Both of these sources drift but the storage batteries are the more stable, lacking rapid fluctuations due to brush noise etc. For these measurements the power source is connected in series with the solenoid, a 0.1
ohm standard resistor and a standard solenoid. The current is determined by the voltage drop across the standard resistor as measured by a Leeds and Northrup type K2 potentiometer and the field is determined by the ratio of deflections from the nitrogen solenoid and standard respectively. The standard is 17.55 \pm 0.01 gauss per ampere. Since the standard begins to heat at currents above 10 amperes, it is used for direct comparison at fields less than 200 gauss. The Hall effect meter, independently calibrated against a 600 gauss permanent magnet supplied with the meter, agrees with the calibration of the standard solenoid but the drift and relatively coarse adjustment controls make the Hall meter readings uncertain by perhaps several percent. In general, the ballistic measurements and the Hall measurements agree at low field but the ballistic readings seem to be much more accurate.

In the medium field region current is supplied by the diesel generator run at idle speed, producing a minimum current of 200 amperes through the solenoid. This current corresponds to a power of approximately 2 kw if the solenoid is immersed in liquid nitrogen, but the rate of nitrogen boil off is small enough that the pump and recirculation system are not necessary. The measurements in this range are relatively easy to make because both the generator and the solenoid are extremely
stable. The measuring coil used for these measurements consists of 10 turns wound on a nylon spool about 5/8 inch in diameter. The coil produces a deflection of 1.91 divisions per kilogauss on a galvanometer of 120 divisions full scale so the reading accuracy of 1/4 division is therefore about ± 125 gauss or 1.5% at 10 kilogauss. The measuring coil is calibrated by the transverse field of a 12 inch Varian nuclear magnetic resonance magnet at a field of approximately 10 kilogauss.

The measurements at high fields are less accurate because of ± 20 ampere current fluctuations at a current of about 1,000 amperes. High frequency fluctuations from brush noise etc. can cause errors in ballistic field measurements but only variations of time constant greater than about 1/2 second can be observed on the current meter easily. Very slow drift can be compensated by manually adjusting the generator current.

Considering all things, the overall accuracy of the field measurement is probably about 2-3%. Refer to fig. 2 for a plot of all field measurements made in the medium and high field regions. The low field data have not been plotted because of the compressed scale. The low and medium field measurements are weighted more heavily in determining the straight line because the high field values are more uncertain. The nitrogen solenoid
calibration is then $30.5 \pm 1.0$ gauss per ampere for a liberal error of about $3\%$.

The experimental calibration curve can be compared with design calculations. Adair\textsuperscript{5} considers the solenoid to be equivalent to a single layer coil of the same turns ratio $N$ and length by choosing the equivalent radius and applying equation (1), page 18, at the center of the solenoid. Adair picks the equivalent radius by taking the average of the cosines of the angles to the inner and outer turns of the solenoid respectively and the result is 29.4 gauss per ampere. If one picks the equivalent radius by taking the arithmetic mean of the inner and outer radius of the coils $B$ equals 30.1 gauss per ampere. Adair also uses a relation due to Cockcroft which considers the finite volume of the current distribution and finds that the field is 33.4 gauss per ampere. The values computed by these various methods involve a certain amount of approximation, so they compare favorably with the observed 30.5 gauss per ampere calibration.

3. **Field Homogeneity.**

It is interesting to compare the homogeneity of the nitrogen solenoid with some simple calculations. The field of a single layer solenoid along its axis has the form\textsuperscript{7}
\[ B = \frac{1}{2} \mu NI (\cos \beta_2 + \cos \beta_1) \]  
\[ (1) \]

\[ y = \frac{a}{L} \quad x = \frac{b}{L} \]

\[ B(0) = B_0 = \frac{\mu NI}{\sqrt{1+y^2}} \]

\[ \frac{B}{B_0} = \frac{\sqrt{1+y^2}}{2} \left( \frac{1+x}{\sqrt{(1+x^2) + y^2}} + \frac{1-x}{\sqrt{(1-x^2) + y^2}} \right) \]  
\[ (2) \]

N is the number of turns per unit length, I is the current, \( \mu \) is a dimensional constant and the point of observation is defined by the angles \( \beta_1 \) and \( \beta_2 \). For ease of computation, equation (1) can be put into dimensionless form and normalized to the maximum field.

Equation (2) is plotted in fig. 4. In these units the center of the solenoid is the point \( x = 0 \) and the end is the point \( x = 1 \). The theoretical curves represent diameter to length ratios \( y \) of 1.0 and 0.5 respectively. The nitrogen solenoid can be compared with these curves by...
FIELD HOMOGENEITY

$\frac{B}{B_0}$

Fig. 4

Y = 1.0

Y = 0.5

OBSERVED
computing an average value of $y$ for the solenoid. The simple arithmetic mean of the outer and inner diameter of the coils yields a value of $y = 0.96$.

The axial field dependence actually observed with the Hall effect meter at low fields is plotted with the theoretical curves. Because the maximum field for these readings is about 250 gauss, the fluxmeter reads in the lower quarter scale with an accuracy probably no better than 5%. Within reading accuracy the field is constant for 4 centimeters along the axis of the solenoid and the field is constant across the cross section of the working space.
PART II. MEASURING EQUIPMENT AND LOW TEMPERATURE APPARATUS

1. Description.

The apparatus used to produce the low initial temperatures necessary for the magnetic cooling process is shown schematically in fig. 5. Plate I is a photograph of the cryostat.

1.1 Liquid Helium System.

The glass liquid helium dewar is supported from above by a large brass tee joint. The lower portion of the dewar is less than 2 inches O.D. and the upper portion is about 4.5 inches O.D. The "tail" must be small enough to fit into the working space of the solenoid described in Part I of this thesis and the upper portion must be large enough to hold a reasonable supply of liquid helium. The horizontal opening in the tee joint is connected to a large mechanical vacuum pump (NRC, model 30-S) by a 3 inch pumping line. Several vacuum valves in the pumping line serve as a means of regulating the temperature of the helium bath by controlling the pumping speed of the pump. The temperature of the helium bath is determined by measuring the pressure above the bath. For relatively high temperatures the height of mercury in a manometer is measured with a cathetometer calibrated to 0.005 cm., and for the lower temperatures the pressure is measured directly on a McLeod gauge. A liquid nitrogen trap is included in the pressure measuring line to prevent volatile contamination
LOW TEMPERATURE APPARATUS

TO HIGH VACUUM

VEECO VALVE

ION T.C.

TO NRC PUMP

NRC VALVE

BYPASS VALVES

INLET PORT

FILL PORT

TO PRESSURE GAUGES

EXCHANGE GAS VALVE

SAMPLE

HELIUM DEWAR

PYREX PEDESTAL

RADIATION TRAP

SAMPLE CHAMBER

NYLON PLUG

FIG 5
in the line from affecting the pressure reading.

1.2 Sample Chamber.

A brass plate which will be called the cryostat head is vacuum sealed to the top of the tee joint. The helium fill tube, the helium bath pressure measuring line and the sample chamber are all connected to the cryostat head. The sample chamber is made of 3/4 inch thin wall nickel silver tubing to keep the heat leak to the bath small. Above the sample chamber is a small brass radiation trap to keep energy from radiating down the tube to the sample. The radiation trap is soldered to a 1/2 inch nickel silver tube which is connected to the cryostat head. A high vacuum system is connected to the sample chamber through a Veeco vacuum valve. The high vacuum system is used to remove the helium exchange gas before the second part of the magnetic cooling cycle is initiated.

1.3 Susceptibility Measuring Equipment.

The magnetic susceptibility of the paramagnetic sample is found by measuring the self inductance of a measuring coil which contains the sample. The balance conditions for the Owen self inductance bridge are found in Appendix A.

A block diagram of the measuring circuit is shown in fig. 6. An audio oscillator (Hewlett Packard, model 200 CD) is coupled to the self inductance bridge (General Radio, type 1632-A) by a 25:1 step down impedance matching
The output signal from the bridge is amplified by a low noise amplifier (Tektronix, type 122 low level preamplifier), and filtered (Krohn-Hite variable bandpass filter, model 310-AB) before being detected on an AC millivolt meter (Hewlett Packard, model 400 H). The measuring coil consists of 2128 turns of #40 copper wire wound directly on the sample chamber over a distance of 2 inches. The leads connecting the coil to the cryostat head are #18 constantan wire and all other connections are made with shielded cables and connectors.

2. Discussion of Experimental Procedure.

The steps necessary to perform an experiment are given and the procedure is evaluated.

2.1 Sample Preparation and Mounting.

Pressed powder samples are prepared from recrystallized salt. Since the salt can easily lose some of its waters of crystallization during storage the salt is first dissolved in doubly distilled water. The saturated solution is allowed to evaporate in a covered container, resulting in hundreds of little crystals. The recrystallized salt is ground to a fine powder. The powder is placed in a cylindrical press and a force of several tons is exerted by a hydraulic press. The mechanical strength and homogeneity of the sample is determined by the fineness of the powder and the magnitude of the pressure. It is desirable to produce a sample with a density greater than about 93% of
the single crystal density in order to have good homogeneity. For example, forces of 10 and 20 tons on a 5/8 inch plunger produced samples of density 94.2% and 97.7% of single crystal density respectively. The cylinder which comes from the press is filed to the desired ellipsoidal shape using a set of templates as a guide.

In fig. 5 a sample is shown mounted on top of a pyrex pedestal. The large part of the pedestal slips into the sample chamber and acts as a guide to keep the sample from touching the sides. For the magnetic cooling process the sample must have a minimum of mechanical contact with its surroundings in order to keep the heat leak small. The heat leak is limited by the small pyrex stem (3 mm. tubing) and the poor thermal contact with the stem. A screw adjustment at the base of the pedestal is used to center the sample vertically in the measuring coil.

After the sample is mounted on the sample holder it is slipped up into the bottom of the sample chamber, a brass plug is put into place, and vacuum sealed with cadmium solder. This particular type of solder is chosen because it is not superconducting. Supercurrents in ordinary tin-lead solder can make magnetic measurements impossible.
2.2 Cooling.

Once the sample is mounted, the sample chamber is inserted into the dewar and the cryostat head is bolted to the tee joint. The Veeco high vacuum valve and the exchange gas valve are closed, and the helium fill port is plugged with a rubber stopper. After the bath space is evacuated by the NRC pump, the NRC valve is closed and helium gas is allowed to enter through the inlet port valve. The dewar is thus flushed with helium gas at least twice, leaving a slight overpressure of helium. Since the dewar is now free from water vapor and other condensable gases, liquid nitrogen is transferred into the outer dewar (not shown). For a pressure of 20-50 microns of air in the helium dewar jacket, the sample reaches nitrogen temperature in about two hours. The approximate temperature of the sample chamber can be determined by measuring the resistance of the measuring coil. The resistance of the coil and leads is approximately 400 ohms at room temperature and approximately 70 ohms at nitrogen temperature.

Because the sample will dehydrate in a vacuum at room temperature it is not possible to test the high vacuum system until the salt has cooled. At this time the Veeco valve is opened and the vacuum system is turned on. There is usually quite a bit of outgassing due to the solder flux left when the sample is sealed into the chamber.
For this reason it is important to pump on the sample chamber for about 24 hours before helium is transferred if good thermal isolation is desired during a demagnetization experiment.

If the sample chamber is properly cooled, the Veeco valve is closed, the helium dewar jacket is evacuated and helium is transferred. The inlet port valve can be used to vent the gas which is blown off during the transfer. When the transfer is complete, the exchange gas valve is opened and a small amount of helium gas enters the sample chamber from the bath space. This gas cools the sample to the temperature of the bath.

2.3 Susceptibility Measurement.

Let the self inductance of a coil in vacuum be $L_0$. Then, if a material of permeability $\mu$ is placed inside the coil the self inductance becomes

$$L = \mu L_0$$

(3)

The permeability is related to the magnetic susceptibility $\chi$ which is the ratio of the dipole moment $M$ to the magnetizing force $H$. In CGS units the relations are

$$\chi = \frac{M}{H} \quad \mu = 1 + 4\pi \chi$$

(4)

For the particular geometry at hand, the sample does not fill all of the volume of the coil. Therefore the value of $\chi$ is diminished by the factor $f = V_s/V_c$, where the
subscripts refer to the volume of sample and coil respectively. Therefore

$$L = L_0 (1 + 4\pi f X)$$  \hspace{1cm} (5)$$

In the helium temperature region, the susceptibility of a paramagnetic salt is inversely proportional to the absolute temperature according to the Curie law. If $C$ is the usual Curie constant, equation (5) becomes

$$X = C \quad \quad L = L_0 \left(1 + 4\pi f \frac{C}{T}\right)$$  \hspace{1cm} (6)$$

The expression for the susceptibility must be altered slightly to take into consideration the fact that the field inside the sample is not exactly equal to the external field. The Lorentz approximation gives

$$X = \frac{C}{T - \Theta}$$  \hspace{1cm} (7)$$

where $\Theta$ is defined by equation (8).

$$\Theta = \left(\frac{4}{3}\pi - \alpha\right)FC$$  \hspace{1cm} (8)$$

$F$ is the ratio of sample density to single crystal density and $\alpha$ is the demagnetization factor for the particular sample shape used. For a sphere $\alpha = \frac{4\pi}{3}$ and for an infinitely long cylinder $\alpha = 0$. Dempsey gives the value $\alpha = 2.15$ for an ellipsoid of length to diameter ratio 2:1 (used in this thesis). Equations (7) and (6) combine to give the general form
In the helium temperature region measurements of the self inductance $L$ are made. $L_0$ is measured at the temperature of liquid nitrogen because the temperature dependent term in (9) can easily be neglected there. For the case of manganese ammonium sulphate the Curie constant is well known and therefore $\Theta$ can be computed. Then a graph of $L$ versus $\frac{1}{T-\Theta}$ yields a straight line which gives the constants $A$ and $B$. For a magnetic cooling experiment the measurements in the helium range are a calibration used to determine the relation between $L$ and $\chi$ for the particular geometry and coil used. Once this relation is known, $\chi$ can be calculated from the measured value of $L$ at any temperature.

The major difficulty in experimentally determining the susceptibility is the fact that the temperature dependence of the susceptibility causes a very small percentage change in the measured value of $L$. For example $L$ changes by only 74 parts in about 2850 over the range 4°-1° K for the manganese salt, and the change is only 45 parts in about 27875 for cesium titanium alum. The change is this small because the Curie constant in equation (8) is very small compared to 1. Therefore $L$ must be measured with extreme accuracy in order to see any temperature dependence at all.
The self inductance bridge used in these experiments can be balanced to six decimal places but absolute measurements made in this way are only accurate to 0.1%. That is, each decade is calibrated to that accuracy. However, in comparing two six decimal numbers which differ only in the last three figures, the result is still within 0.1%. This means that the bridge is well suited to the type of measurements made for this thesis, where only changes are important.

The accuracy of the method is not limited by the calibration of the bridge. The most important factor is the sensitivity of the circuit in determining the balance condition. In particular the bridge is very sensitive to changes in resistance. The fourth place resistance drift experienced in a preliminary equipment test was sufficient to mask the inductance measurement below the fourth place and the third place was uncertain. The balance of the bridge of course depends on the simultaneous balance of both the inductance and the resistance. In the present set up the resistance of the coaxial lead from the cryostat head to the bridge is kept to a minimum by using a cable with a very large center conductor. The temperature dependence of resistance of the leads from the coil to the cryostat head is decreased by using constantan wire. The drift due to temperature variation is by no means minimized but the drift is small
enough to be tolerable. In a recent experiment the resistance drifted slowly by only 50 parts in $12350$ over a period of nearly 7 hours. For this experiment the resistance drift was small enough to have almost negligible effect on the balance of the inductance controls.

Before any demagnetization experiments are performed it will be necessary to make the resistance measurements reproducible by completely eliminating the effect of the leads. At the temperatures encountered after a demagnetization (roughly .1-.001° K) energy absorption can occur. In this case it is convenient to write the susceptibility as the sum of a real and an imaginary part.

$$\chi = \chi' - j\chi''$$

(10)

The real part $\chi'$ corresponds to the ordinary susceptibility and the imaginary part $\chi''$ corresponds to an energy absorption term. In order to determine $\chi''$ it is necessary to make absolute measurements of resistance $R_\chi$ as can be seen from (17) in Appendix A. Therefore the resistance measurements must be reproducible.
Part III. PRESENTATION OF DATA

The equipment described above has been subject to various tests to check the function of each part separately. However, the measuring equipment in particular is best tested in an actual situation. Since the low temperature properties of manganese ammonium sulphate are well known\textsuperscript{11}, this salt is well suited to preliminary experiments. Two low temperature experiments have been performed on this salt and the results compare favorably with data in the literature. A third experiment has been performed on Cs Ti alum. The cesium salt is chemically unstable and somewhat difficult to work with. Consequently, the only data on Cs Ti alum below 1°K which appears in the literature is of a preliminary nature. It is hoped that a careful study of this material in the magnetic cooling range can be made in the future.

1. Manganese Ammonium Sulphate - I.

Preliminary calculations show that the self inductance measurements described above can be very accurate if the bridge is used to its maximum capability. However, external disturbances can easily destroy the accuracy and make the measurements impossible. The measuring coil designed for this experiment will not be described in any detail because it is not satisfactory. Essentially it has only one layer of turns of #36 copper wire. The coil resistance in liquid helium is therefore
small enough to make lead effects very important. The thermal variation of lead resistance was large enough to make the bridge balance very difficult to obtain. However, by waiting a long time the drift becomes smaller and measurements are possible.

Fig. 7 shows the data taken in this experiment. The points on the line are considered more certain because the helium bath is a superfluid at these temperatures and good thermal equilibrium is most probable. The point below the line can be explained by a nonequilibrium thermal condition and the points above the line can be explained by poor equilibrium in the bath pressure measuring line.

The empirical relation between $L$ and $\chi$ is given on the graph. $\Theta$ is computed from the measured value of $F$ and the known value of $C$. The Curie constant per unit volume $C$ for manganese ammonium sulphate is $0.0205^\circ$ K. From the dimensions of the sample (1 inch by 1/2 inch ellipsoid) and the measured packing factor $f$, the constant $B$ in equation (9) can be calculated by comparing (6) and (9). The calculated value of $B = 1180 \mu$H compares with the measured value of $B = 1250 \mu$H (on graph). The results indicate that the method is accurate enough but that the measuring coil needs modification.

In order to check the feasibility of adiabatic demagnetization experiments, one demagnetization was performed on sample 1. The temperature of the salt rises after a demagnetization because of inevitable heat
CURIE LAW REGION

\[ \text{M}_N\text{(NH}_4\text{)}_2\text{(SO}_4\text{)}_2\cdot6\text{H}_2\text{O} \]

SAMPLE 1
COIL 1

\[ L = 643.0 + 1250 \times \frac{1}{T - 0.041} \, \mu\text{H} \]

\[ L = 643.0 + \frac{257}{T - 0.041} \, \mu\text{H} \]

FIG.7

SELF INDUCTANCE \( \mu\text{H} \)

\[ \frac{1}{T - 0.041} \, \text{oK}^{-1} \]
leaks to the sample. Therefore the susceptibility is changing with time and it is important to be able to make inductance readings very quickly. Since the entropy is known accurately only immediately after the demagnetization it is necessary to measure the susceptibility as a function of time after the demagnetization and then extrapolate back to zero time. The extrapolated demagnetization data shown in fig. 8 determine one point on a $\chi$ versus entropy curve. They are hardly acceptable because the first reading occurs too long after the demagnetization and the extrapolation is uncertain. However, the general information gained indicates that the method is quite accurate if the bridge is preset very close to the balance condition before the demagnetization. With some practice, the readings can be taken immediately after the demagnetization and every few seconds thereafter. It would be desirable to separate the resistance and inductance balance controls because of the difficulty in maintaining a simultaneous adjustment of two quantities which are changing rapidly with time. Perhaps with enough practice the operator can balance both controls in the region where $\chi''$ is important. More tests along this line are indicated before one can say that the method is entirely satisfactory.

2. **Manganese Ammonium Sulphate - II.**

Because of the difficulties encountered in the
ADIABATIC DEMAGNETIZATION
OF
\[ \text{Mn}_2\text{(SO}_4\text{)}\text{(NH}_4\text{)}_2\text{(SO}_4\text{)} \cdot \text{6H}_2\text{O} \]
SAMPLE 1
COIL 1
\[ S/R = 1.16 \]

FIG. 8
SELF INDUCTANCE \( \mu \)H
MINUTES AFTER DEMAG.
first experiment a new measuring coil was constructed. The coil is described in Part II above. In order to be certain that the sample was fresh a new sample was prepared. The results of the second experiment are given in fig. 9.

Especially at the temperatures above the superfluid point of the helium bath the temperature is hard to keep precisely constant. The next best thing is to make the drift very small. The data used in plotting fig. 9 are much more accurate than the scale indicates. Several of the points shown are actually an average of several readings taken quite close together in order to check for thermal equilibrium. The points not on the line are nearly all too low, indicating poor thermal equilibrium. The points at the extreme right side of the graph correspond to the lowest temperatures reached. These readings were taken about 7 hours after the helium transfer and the helium level is probably below the sample. Therefore temperature equilibrium is uncertain for these points. The inductance varies according to the expected $1/T$ law but the experimentally determined Curie constant is about 5% lower than the accepted value. Since the measurements are certainly good to a fraction of 1% the discrepancy must be found somewhere else. The largest error is probably in the sample preparation. It is quite difficult to form an ellipsoid with exactly the correct shape. Because these measurements are preliminary, the sample was not shaped
as carefully as possible. In any future tests more care should be taken in preparing the sample. Because the deviations from the expected Curie constant for the two samples are in opposite directions the deviation is definitely due to some sort of experimental error. This deviation should be checked in more detail.

3. **Cesium Titanium Alum.**

Cs Ti alum is an example of a spin 1/2 system. The paramagnetic ion is Ti$^{3+}$. Unfortunately the salt is chemically unstable and the titanium oxidizes in air to diamagnetic Ti$^{4+}$. The sample was prepared in a nitrogen atmosphere to prevent oxidation. The salt has been stored in a sealed glass tube at 0° C since it was first prepared. Because of the instability of the salt it was decided not to subject it to the severe mechanical processes used in preparing the manganese salt. However it was found necessary to grind the crystals moderately because the sample would not hold together otherwise. The salt was pressed in a small hand press so that there would be no danger of squeezing out any waters of hydration. Perhaps the precautions were overdone. The resultant sample is extremely soft and very difficult to mount in the usual manner. In the future it is suggested that the salt be tamped into a glass container in some inert atmosphere and sealed. In this manner the sample is not exposed to the atmosphere and it does not have to be mechanically strong.
The present sample is coated with a heavy coating of stopcock grease to prevent oxidation.

The helium temperature data on Cs Ti alum are shown in fig. 10. A detailed analysis is not made because of the scatter of points. Further checks on data reproducibility are desired. If one considers the points above the helium superfluid temperature to be uncertain, the "best" points indicate a $1/T - \Theta$ dependence with a small negative $\Theta$. Because the Curie constant is so small the $\Theta$ correction in equation (7) is negligible.

Benzie and Cooke\textsuperscript{12} give the molar susceptibility of Cs Ti alum as 0.118. This figure is converted to CGS units by multiplying by the density (about 2) and dividing by the molecular weight (589). The result is $4.0 \times 10^{-4}$ (per cm\(^3\)). Because of the difficulty in preparing the sample the weight and dimensions are not very certain. The packing factor is determined as about 0.9 and the sample volume is about 8.4 cm\(^3\). The coil volume is 14.5 cm\(^3\). The relation between the constant $BC$ in equation (9) is $BC = 4\pi f L_o C$. From the experimental value of $BC$ (on the graph) and the values of $L_o$ and $f$, an experimental value can be calculated for the Curie constant. This calculated value is $3.9 \times 10^{-4}$. Therefore there is at least qualitative agreement with other experiments. A low value of the Curie constant can be explained by a lower density of paramagnetic ions due to
CURIE LAW REGION

$T_i \cdot C_8(SO_4)_2 \cdot 12H_2O$

$L = 27.83 + \frac{0.775}{T} \text{ mH}$

FIG. 10
oxidation. The outside of the sample shows discoloration which might be caused by oxidation.

The general conclusion which can be drawn from this set of experiments is that the self inductance method is sufficiently accurate to determine the susceptibility of Cs Ti alum. The present measuring coil is quite adequate but the temperature dependence of the lead resistance must be compensated. Sample preparation techniques must be improved for the cesium salt possibly going to a sealed glass container. Further data on the cesium salt are necessary to verify the observed temperature dependence.
APPENDIX A

Owen Bridge

The Owen bridge is shown schematically in fig. 6. The series resistance $R_x$ and the inductive reactance $X_x$ of an unknown inductor are determined by the balance condition obtained by adjusting $R_n$ and $X_n$ for a null reading on the detector. The balance condition is obtained by equating the ratios of the complex impedances.

$$\frac{Z_a}{Z_a} = \frac{Z_x}{Z_b}$$

$Z_a = jX_a$
$Z_b = R_b$
$Z_n = R_n + jX_n$
$Z_x = R_x + jX_x$

Equate the real and imaginary parts separately and use the definitions of impedance to determine the desired quantities.

$$\frac{X_n}{X_a} = \frac{R_n}{R_b} \quad \frac{-R_n}{X_a} = \frac{X_x}{X_b}$$

$$X_a = -\frac{1}{\omega C_a} \quad X_n = -\frac{1}{\omega C_n} \quad X_x = \omega L_x$$

$$R_x = \frac{C_a R_b}{C_n} \quad L_x = R_b R_n C_n$$

For the particular instrument at hand, the components $C_n$ and $R_n$ serve as a fine adjustment and the components $C_a$ and $R_b$ serve as a coarse range setting. The values of $L_x$ and $G_x = 1/R_x$ are read directly off the dials of the instrument.

Consider the case in which the unknown inductor
contains a material of susceptibility \( \chi \). Let A, B and \( R_X \) be known constants and define the quantities \( \chi' \) and \( \chi'' \) below.

\[
L_x = A + B \chi \quad \chi = \chi' + \chi'' \tag{15}
\]

Similarly the balance readings L and G are

\[
L = A + B \chi' \quad \frac{1}{G} = R_x + \omega B \chi'' \tag{16}
\]

\[
\chi' = \frac{L - A}{B} \quad \chi'' = \frac{1}{\omega} - \frac{R_x G}{B G} \tag{17}
\]

The coaxial cable which connects the unknown inductor to the bridge looks like a capacitance of about 450 \( \mu \)F in parallel with the inductor. In order to compute the effect of the cable let \( R_X = 10 \Omega \), \( L_X = 30 \text{ mH} \) and \( \omega = 450 \pi \).

\[
X_X = 42.4 \Omega \quad R_X = 10.5 \Omega \quad X_C = 1.57 \times 10^6 \Omega
\]

\[
\frac{1}{Z_x} = \frac{1}{j X_C} + \frac{1}{R_X + j X_C}
\]

\[
Z_x = \frac{X_C \left( R_X X_C + j \left[ R_X^2 + X_X (X_X + X_C) \right] \right)}{R_X + (X_X + X_C)^2} \tag{18}
\]

For the parameters at hand a reasonable approximation is to neglect quantities which are of the order of \( X_X^2 \), \( R_X^2 \), \( X_X R_X \) in comparison to \( X_C^2 \). The result is a first order correction for the effect of the cable.

\[
Z_x = \left\{ \frac{1}{1 + 2 X_X X_C / X_C} \right\} (R_X + j X_X) \tag{19}
\]
Therefore the values of $R_x$ and $L_x$ determined by (4) should be corrected by the multiplicative factor in curly brackets above if absolute readings to one part in $10^5$ are desired. However, the derived quantities determined by (17) contain ratios of quantities with the curly bracket as a factor. To a high order the readings are thus unaffected by the capacitance of the cable.
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