

# NBS TECHNICAL NOTE 823

# Cryogenic Physics Section, Summary of Activities 1973

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# Cryogenic Physics Section, Summary of Activities 1973

R. J. Soulen, Jr., Editor

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#### FOREWORD

This annual report covering the calendar year 1973 provides an opportunity through the medium of an NBS Technical Note to report in some detail on progress made in achieving the long-term goals of the Cryogenic Physics Section. These goals include the establishment of a thermodynamic temperature scale below 20K and developing the means for its dissemination. As to the former, we will report on nuclear orientation, noise, and nuclear magnetic resonance thermometry. Our progress in the development of superconductive fixed points relates to the latter goal. Another long-standing program is devoted to the study of paramagnetism in electronic spin systems. Research in this area was fruitful this year; we report on theoretical and experimental studies of several paramagnetic systems. Other short-term projects in the Section have been reported in the scientific literature and will not be repeated here.

R. J. Soulen, Jr., Acting Chief Cryogenic Physics Section Heat Division Institute for Basic Standards

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CRYOGENIC PHYSICS SECTION

SUMMARY OF ACTIVITIES 1973

R. J. Soulen, Jr., Editor

This report summarizes the research activities of the Cryogenic Physics Section which specifically relate to thermometry. The topics range from superconductive fixed points to nuclear orientation thermometry, as well as Josephson junction noise thermometry and paramagnetism.

Key Words: Josephson junctions; noise thermometer; nuclear orientation; paramagnetism; superconductivity; temperature.

JOSEPHSON JUNCTION NOISE THERMOMETRY - R. J. SOULEN, JR.

#### A. Noise Spectral Analysis

There were two important developments in noise thermometry in 1973. The first represents a significant advance in "debugging" the noise thermometer. The on-line minicomputer used in the experiment has now been reprogrammed to calculate several higher order moments of the variance of the frequency spectrum (see NBS Report 10 977). Spurious noise sources represented by power spectra with a  $f^{-n}$  frequency dependence are now detectable at the 1% level. Since the minicomputer prints its results on a Teletype as data are accumulated, the presence of any spurious noise spectra will be detected, and corrective measures can immediately be taken. Thus, the data acquisition system can be used much more effectively to eliminate extraneous noise sources and guarantee that the data consist only of the Johnson noise originating in the thermometer.

#### B. 10 GHz Noise Thermometer

More recently, in a project involving R. A. Kamper (NBS Boulder) and T. F. Finnegan (NBS Gaithersburg), a prototype noise thermometer was developed which should lead to an increase in the speed of temperature measurement. Basically, the technique involves pumping the thermometer (or resistive SQUID) with a 10 GHz signal instead of the more conventional 30 MHz one.

The 10 GHz resistive SQUID we tested is shown in figure 1. It consists of a section of X-band waveguide reduced to 0.25 mm in the E direction to give a characteristic impedance of 10  $\Omega$  in order to match it to the impedance of the Josephson junction. The junction is formed by rotating the adjustment lever attached to a flat niobium screw until it gently touches the sharpened end of a fixed niobium screw. The junction is placed a distance  $\lambda/4$ from the shorted end of the guide where the electric field is a maximum. The interior of



Figure 1. 10 GHz resistive SQUID. Upper drawing shows side view of the device; the lower figure shows the top view.

the guide is lined with a superconducting material (Babbitt metal). A circular groove has been machined in the upper half of the guide, thus placing a section of brass resistor ( $\sim$  3µ $\Omega$ ) in parallel with the junction. Direct current is passed through the resistor by connecting a coaxial line leading from room temperature to two solder connections shown in figure 1.

The resistive SQUID shown in figure 1 was immersed in liquid He<sup>4</sup> at 4.2 K. The 30 MHz output is shown in figure 2. The signal at the center was proportional to the 10 GHz signal reflected from the cryostat which, in this instance, was approximately  $5 \times 10^{-11}$  W. The resistor was biased at  $2 \times 10^{-9}$  volts with a current of 0.66 mA. This voltage caused the junction to oscillate at 1 MHz. This oscillation produced a 20% amplitude modulation of the 10 GHz signal, giving rise to the two sidebands in figure 2. By varying the dc voltage, we found that the frequency of the sidebands could be continuously adjusted between 0 and 5 MHz; the upper limit was determined by the bandwidth of the intermediate frequency amplifier. We observed quantum oscillations over a power range from  $10^{-11}$  to  $10^{-9}$  W, depending on the adjustment of the point contact. The optimum power for obtaining the best signal-to-noise (S/N) for the video output was found to be  $5 \times 10^{-11}$  W. For this power level and a post-detection bandwidth of 100 kHz, the power S/N was determined to be  $\sim 28$  dB. Finally, at power levels near  $10^{-10}$  W we did observe more complicated spectra with additional side-bands corresponding to higher harmonics of the fundamental frequency.

Thus we found that a power signal-to-noise enhancement of 28 dB has been achieved in a resistive SQUID by increasing the pump frequency from 30 MHz to 10 GHz. In order to take full advantage of this demonstrated enhancement, it will be necessary to increase the bias resistance of the SQUID. These experiments are now in progress, and will be actively pursued in 1974.

#### NUCLEAR ORIENTATION THERMOMETRY - H. MARSHAK

#### A. Interregnum

The beginning of 1973 found us with a low temperature leak in the bottom part (stillheat exchangers-mixing chamber) of Dr. Soulen's dilution refrigerator. We have been using this refrigerator for our comparison measurements between a Josephson junction noise thermometer (JJNT) and a  $Co^{60}$   $\gamma$ -ray anisotropy thermometer ( $Co^{60}$   $\gamma$ -RAT). After fruitless attempts at locating the leak we decided to replace the entire bottom part. Since some funds were available we also decided to purchase rather than build a new bottom. The advantage was that the prime manufacturer at that time had had considerable experience in fabricating these units and their performance was guaranteed. As the delivery date for this unit was to be in early summer this down time proved particularly appropriate since Section 221.11 needed help in some of their calibration work.

The next six months, with a total of about four-man months, were spent calibrating and testing quartz thermometers. This work was done for the Optics Branch of the Naval Research Laboratory. In particular they requested that three newly purchased units along with six probes be calibrated in 5 °C steps from -20 to +30 °C. Since the National Bureau



Figure 2. Spectrum analysis of the 30 MHz output of the resistive SQUID at 4.2 K. Vertical scale: IF output, uncalibrated

Horizontal scale: 500 kHz per division

of Standards does not have a routine calibration service for quartz thermometers (as we do, for example, for platinum resistance thermometers), this calibration was to be viewed more as a study or test of the particular quartz thermometers involved. Each system (one unit plus two probes) was first tested for short (8-hours) and long (30-days) term stability using triple point of water cells as the constant temperature bath. Next they were calibrated over the temperature range of interest and the data were evaluated and calibration curves were generated. Finally a second calibration was undertaken to test the results of the first calibration. The results of all this work were presented in a N.B.S. Test Report (No. G42285) which was furnished to N.R.L.

### B. Recent Work in Co<sup>60</sup>

At about the end of the above work the new bottom for the dilution refrigerator was installed. In July we tested it and the results were very encouraging. Using the  $Co^{60}$   $\gamma$ -ray anisotropy thermometer we found out that we could maintain 11.8 mK continuously and 9.9 mK by the one-shot technique. Using the old bottom we were only able to obtain 20 mK continuously. This improvement will enable us to make a more critical comparison between the JJNT and the  $Co^{60}$   $\gamma$ -RAT since the sensitivity ( $\Delta$ T/T) of the latter improves between 10 and 20 mK over that above 20 mK.

During this time, y-ray background data were taken to try and understand small (but statistically significant) changes that occur in our background counts every 2 to 3 weeks. We were able to order a much needed multi-channel analyzer which will help to improve the quality of our data.

While a new noise thermometer was being built we did some experiments on the dilution refrigerator to measure the superconducting transition temperature of some new samples of beryllium and tungsten. This work is discussed in another section of this report.

We also started some measurements on the thermal contact between our cobalt thermometer and the mixing chamber. Our first results indicate that the response time of our present thermometer is not as good at the lowest temperatures as that of our germanium resistor thermometer. The cobalt thermometer is held on a copper mount by gold plating. Since this is a fairly old ( $\sim$  2 years) thermometer and the plating might have deteriorated we plan to try a "fresh" thermometer as well as using some different techniques to make better thermal contact.

Our new multi-channel analyzer arrived and we spent some time interfacing it with the present system. Some preliminary stability tests were made using it in the integrated mode and our single channel analyzers in the normal fashion. Since our new nuclear magnetic resonance - oriented nuclear (NMR-ON) sample was ready we postponed further stability tests till a later date. Although our new sample was no thinner ( $\sim$  75µm) than the first one we ran, it was still worthwhile to do the experiment again as we could now get colder than our previous 20 mK--thus enhancing any effect. After a few weeks of very careful and tedious measurements we had some indication of a positive effect; that is, destruction of some of the nuclear orientation by using a frequency modulated RF signal of  $\sim$  128 to 130 MHz. Further measurements showed that our initial results (which were just slightly outside

fall in the region accessible with a  ${}^{3}\text{He}{}^{-4}\text{He}$  dilution refrigerator, these two elements are worthy of some detailed experimental study. Just as importantly, we expect that their T 's can be assigned to the temperature scale defined by the noise and Co<sup>60</sup> y-ray thermometers (see NBS Report 10 977), and that they can be used as a means of dissemination of that temperature scale.

#### A. Beryllium

Four of the five beryllium samples studied, labelled Be 1, 2, 3, 4, are part of a high purity lot prepared by vapor deposition. Because of the vapor deposition technique, the samples are quite irregular in shape with approximate linear dimensions of 10, 3, and 2 mm. A fifth sample, BEFI, is a 2 x 3 x 8 mm parallelepiped spark cut from a piece of beryllium which was electron beam zone refined 12 times.

Each sample was attached to the copper mixing chamber of our dilution refrigerator. Thermal contact was obtained by tightly tying the sample with nylon thread into a bundle of 200 #43 bare copper wires which had been silver-soldered to one end of a copper stud. The other end of the stud was threaded (2-56) and screwed into the bottom of the mixing chamber. A coil set 1 cm long, composed of a 200 turn primary and a 1000 turn secondary was slipped around each sample. The transitions were detected in the usual way.  $[1]^1$  A recording typical of our results is shown in figure 3 for sample Be 1. Curves 1 and 2 represent two complete thermal cycles in which the sample was cooled from the normal (N) state until it entered the superconducting (SC) state, then warmed until it returned to the normal state. Curve 1 was taken in the smallest dc magnetic field which we could maintain (less than 1 µT), while curve 2 was taken when the magnetic field along the long axis of the sample was increased by 1 µT. The hysteresis is due to the extreme sensitivity of the SC-to-N transition to magnetic fields and is indicative of strong supercooling. To reduce the magnetic fields we used a single mu-can as well as three mutually orthogonal Helmholtz coils.

Specimens Be 2, 3, 4 displayed the same strong dependence of supercooling on ambient magnetic field and weak dependence of  $T_c$  on the same field. The sample BEFI exhibited an even greater sensitivity to magnetic fields than the other samples and was occasionally difficult to coax into the superconducting state. Its  $T_c$  lies 0.5 mK colder than the other samples and experiments are underway to investigate the cause of this discrepancy.

The results of all the  $T_c$  measurements are given in table I. These measurements give an indication of the reproducibility which presently can be obtained. All the measurements of  $T_c$  for Be 1, 2, 3, 4 fall within 0.2 mK of the average value of 22.6, mK. We assign an uncertainty of 0.5 mK to this average which reflects the present uncertainty in the absolute temperature established by our noise and anisotropy thermometers. This value differs from the  $T_c$  of 26 mK reported by Falge. [2] The latter value is undoubtedly in error due to the inherent uncertainties associated with the use of potassium chrome alum for thermometery.

<sup>&</sup>lt;sup>1</sup>Figures in brackets indicate the literature references on page 10.





TUDDO T	TA	BL	E	I
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Sample	<u> </u>
Be 1	22.80
Bę 1	<sup>22.8</sup> 1
Be 2	22.7 <sub>9</sub>
Ве 3	22.60
Ве 3	<sup>22,5</sup> 5
Be 3	22.5 <sub>9</sub>
Be 4	22.6 <sub>0</sub>
BEFI	22.00
BEFI	22.3

In conclusion, the evidence presented here indicates that pure beryllium should serve as a useful superconducting fixed point for calibration procedures provided that sufficient care is taken to eliminate ambient magnetic fields. Furthermore, our value of  $T_c$  is probably very close to the <u>absolute temperature</u> of the transition since both the noise and anisotropy thermometers do not depend upon any calibration or extrapolation; only on some accurately and independently determined parameters of each system.

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#### B. Tungsten

To date we have only examined the transitions of one high-purity W sample which has a residual resistance ratio of 20,000. Our experimental data are quite preliminary, but indicate that it has a sharp  $T_c$  (width of  $\sim$  .00001 K) near 15 mK. Its supercooling is even more sensitive to ambient dc fields than that of Be. More data are clearly necessarv before much can be said about its role as a superconductive thermometric fixed point.

# C. Calibration of Paramagnetic Salts Using Superconductive Fixed Points

I. INTRODUCTION

At low temperatures, a paramagnetic material may be used as a sensitive thermometer

because its susceptibility varies inversely with temperature. Use of a paramagnetic thermometer is quite straightforward: a signal, M, proportional to the susceptibility is measured by any one of several techniques [1-3]<sup>1</sup> and is fitted to the Curie Law

$$M = AT^{-1} + B \tag{1}$$

where T is the temperature, and A, B are coefficients which must be determined from a minimum of two calibration points. We have shown that the coefficients A and B are easily determined by calibration at the narrow superconductive transitions of several pure elements. Certain of these transitions have been assigned values so that they form a set of thermometric reference points suitable for just such an application. As a particular demonstration of the technique, we will report on some experiments in which the paramagnetic salt cerous magnesium nitrate (abbreviated CMN) was calibrated from 0.5 to 1 K in order to determine the superconductive transition temperature and temperature dependence of the critical magnetic field of pure iridium. Because of space limitations in the cryostat, we chose to conduct the experiment in two parts. First, the CMN thermometer was calibrated at the aluminum, zinc, and cadmium points and the CMN thermometer was used in turn to calibrate a germanium resistance thermometer in the temperature range 0.03 - 1.0 K. Second, the T and supercooling properties of several iridium samples were determined as functions of the calibrated germanium thermometer resistance in subsequent experiments. Thus, while in principle the experiments need not have involved the use of a germanium resistance thermometer, its use was dictated by the geometry of our cryostat.

#### SUPERCONDUCTIVE FIXED POINT DEVICE

The CMN thermometer was calibrated using a superconductive fixed point device, SRM 767, obtained from the NBS Office of Standard Reference Materials, [4] which consists of a set of five samples (Pb, In, Al, Zn, and Cd) varnished into a copper stud and enclosed by a bakelite cover and two copper coils. As a sample undergoes its superconductive transition, the mutual inductance of the coil set changes and is easily detected with a Hartshorn bridge circuit. [5] The midpoint of the mutual inductance change is identified as the superconductive transition temperature,  $T_c$ . All five samples exhibit narrow and reproductible transitions and thus serve as thermometric reference points with a stated precision of + 1 mK.

The SRM 767 unit provides a total of five reference temperatures. However, owing to lack of sensitivity of the CMN thermometer at higher temperatures as well as departures from Curie-law behavior above 3 K, [6] only the three superconductors with the lowest transitions were used. The stated transition temperatures for Al, Zn and Cd are  $1.174_6$ , 0.844, and 0.515 respectively. [7] The widths of the same transitions for this particular unit (Serial #1) were 0.8 mK, 4.5 mK and 0.8 mK. (The transition width is defined as that

<sup>&</sup>lt;sup>1</sup>Figures in brackets indicate the literature references on page 14.

temperature range over which the central 80 percent of the change in mutual inductance occurs). Calibration data were always taken within  $\pm$  10% of the midpoint of each transition which leads to an estimated uncertainty of  $\pm$  0.2 mK,  $\pm$  0.5 mK, and  $\pm$  0.3 mK in achieving the superconductive transitions of Al, Zn and Cd.

#### CMN THERMOMETER

The CMN thermometer was a sphere (1.892 cm diameter) machined from a compress of CMN crystals, silver wire and AgCl powder. [8] A silver rod extending from the sphere was clamped to a gold-plated copper section of the mixing chamber with a nylon compression ring. [9] This particular arrangement produced satisfactory thermal contact between the CMN pill and the mixing chamber down to 0.030 K. For the choice of coils and the Hartshorn bridge used, the calculated sensitivity of the system was  $\pm$  0.3 mK at the transition temperature of Al,  $\pm$  0.2 mK at the T<sub>c</sub> of Zn and  $\pm$  0.1 mK at the T<sub>c</sub> of Cd. Repeated bridge measurements at a given T<sub>c</sub> produce readings which did not exceed twice these values.

#### **II. CALIBRATION PROCEDURE**

The coefficients A and B in equation 1 were determined for this CMN thermometer using the three temperatures defined by the superconductive transitions of Al, Zn, and Cd. The calibration procedure began with cancellation of the three components of the ambient magnetic field. Then the cryostat was stabilized at the midpoint of the mutual inductance change caused by one of the superconductors and the value of the germanium resistance was recorded. The Hartshorn bridge was switched to measure the susceptibility of the CMN while the germanium resistor was monitored to guarantee that the CMN was calibrated to within  $\pm$ 10% of the center of the mutual inductance change. This procedure was repeated for the other two superconductors. After calibration of the CMN, the dilution refrigerator was permitted to cool slowly and was stabilized at several temperatures from 0.030 to 1 K in order to calibrate the germanium resistance using the CMN. Several data points were taken in the vicinity of the T<sub>c</sub> of iridium (0.1 K). The complete experiment was repeated a second time after warming the cryostat to room temperature. This procedure thus led to a temperature scale transferred to the germanium resistor which depended on a CMN sphere calibrated with the aid of superconductive thermometric reference points.

#### **III. CALIBRATION RESULTS**

The quality of the calibration of the CMN thermometer is shown in table II. The first column identifies the superconductor, the second the value of its defined  $T_c$ . The third column gives the value calculated from equation 1 after A and B had been determined by a least squares fit of M to all three superconducting fixed points. The last column gives the results of the second experiment carried out a month later. The scatter in the data is well within the  $\pm$  1 mK precision claimed for the SRM 767 units. In fact the scatter is less than would be expected from the combined uncertainties of the measurement of M and

determination of  $T_c$ . The data suggest that the precision of an individual SRM unit could be considerably better than 1 mK.

Defining	Defining T <sub>c</sub>	Least Square Values Expt I	Least Square Values Expt II
Superconductor	<u>(K)</u>	(K)	(K)
Aluminum	1.1746	1.1741	1.175
Zinc	0.844	0.8442	0.8445
Cadmium	0.515	0.5149	0.515

TABLE II. RESULTS OF CMN CALIBRATION

## IV. APPLICATION: SUPERCONDUCTIVE PROPERTIES OF IRIDIUM

A study of the superconductive properties of iridium and some of its alloys has recently been reported. [10, 11] The apparatus described in this article was used to measure  $T_c$  and supercooling effects, while magnetization curves and critical magnetic field data were taken in another dilution refrigerator. The temperature scale used in these experiments depended ultimately on the CMN - SRM 767 temperature scale discussed earlier in this article. However, as we noted earlier, the iridium samples were not mounted on the dilution refrigerator in the same experiments with the CMN and SRM 767 because the CMN sphere and coil set occupied too much space in the cryostat. The experiments were conducted in the following way. Data on the  $T_c$  of each iridium sample were recorded along with a value of the germanium resistance. Data on the superconductive transitions were found to be quite reproducible, [10] they were used to transfer a calibration to the apparatus used for the magnetic measurements so that, at this stage, all data were recorded versus a set of germanium resistance values. Finally, in the two later calibration experiments described in this article, the germanium resistance values were converted to temperature.

One iridium sample was studied in detail because its purity was quite high (residual resistivity ratio  $\geq 2000$ ). This sample exhibited reversible, Type I magnetization curves, and on the basis of the temperature dependence of the critical magnetic field, it was characterized as a BCS superconductor with a critical magnetic field at T = 0 of 16.00 ( $\pm$  .05) x 10<sup>-4</sup>T, a T<sub>c</sub> cf 0.1125 ( $\pm$  0.0005) K, and an average gap anisotropy parameter of ( $a^2$ ) = 0.048 ( $\pm$  0.005). The internal consistency of these data as well as consistency with the supercooling data on this sample [11] give good testimony to the adequacy of the temperature scale used throughout the experiments.

# V. CONCLUSIONS AND SUGGESTIONS

The data summarized in table II clearly show that a paramagnetic material (in this case CMN) can be calibrated using superconductive thermometric fixed points with a precision exceeding that of the thermometric fixed points. The paramagnet can then be used to calibrate other secondary thermometers (in the experiments described here, a germanium resistance thermometer) or to provide a direct in situ temperature scale for experiments. In the example given here, the temperature scale calibration and application were divided into separate experiments only because of geometric considerations. One means of reducing the bulk of the CMN system without loss in temperature resolution is to use a SQUID, [12] a Josephson junction device, for the measurement of susceptibility. Moreover, the somewhat clumsy procedure of switching the same electronic system between the paramagnet and the SRM 767 unit can be avoided if the detection of the fixed points is carried out with a very simple electronic bridge recently developed. [13] Thus a streamlined experiment could simply include a small CMN salt and SQUID detection system, and an SRM 767 device, mounted on a common apparatus with the rest of the experiment. Finally, we remark that this procedure is not limited to the choice of CMN as the paramagnet: copper and platinum nuclei serve as Curie Law thermometers [3, 14] at low temperatures, while neodymium ethylsulphate is well suited for work at higher temperatures. [15]

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- 15. R. P. Hudson, private communication.



Figure 4. Scale drawing of calibration apparatus. The primary coil used to measure the susceptibility of CMN is compensated with end windings in order to produce a magnetic field uniformity of 3% over the volume of the sphere. The secondaries were wound as series opposites with equal area-turns so that the mutual inductance of the coil set was small at 4 K. Insulated copper wires not shown in the figure were wrapped around the CMN coils to keep them in thermal equilibrium with the mixing chamber. The copper mixing chamber has an extension which is clamped around the 3 mm diameter Ag rod with a nylon compression ring. A nylon set screw prevents slippage of the clamp at room temperature. Electrical leads have not been shown for clarity.

#### A. Dipole-Dipole Interactions in Magnetic Crystals

As pointed out in the previous report (NBS Report 10 977 ) this project is concerned with the high temperature expansion of the partition function for salts that contain paramagnetic spins. During the period from July 1 till the end of the year the work was focused on a refinement of the cerous magnesium nitrate (CMN) calculations. This salt has a crystallographic structure in which the planes in the z direction are almost equidistant. In the earlier work the distances were taken to be equal. It may be of importance to study the influence of this small change, in particular since the experimental results are still slightly different from the calculated results. The introduction of unequal spacing results in a larger unit cell containing four, rather than one, atoms per unit cell. It was with this in mind that our programs were redesigned this past year. Results with this new program will be obtained in 1974.

The work on Erbium Phosphate mentioned in the previous report is completed and is being prepared for publication.

#### B. Quantum Mechanical Cluster Model for Cerous Magnesium Nitrate

This project has been carried out in cooperation with Dr. Niemeijer of Delft University. The ground state for the totally symmetric state of CMN (with respect to the permutation group) was obtained from a 46 by 46 matrix. This ground state is about twice as low as the ground state from the Hartree-like result obtained by the Luttinger-Tisza method. There are reasons to believe that this totally symmetric ground state is not the lowest state for the system and thus we are continuing to calculate matrix elements for the seven non-symmetrical spin states. The work is about half completed. Dr. Niemeijer will be a visiting professor at Temple University this coming year and we will use the opportunity to complete this work.

#### C. Crystal Field Calculation on Paramagnetic Compounds

The calculation for the susceptibility of spin doped compounds based on their crystal field parameters has been completed and was submitted for publication. The paper is entitled: "Magnetic Properties of Paramagnetically Doped Crystals: Fe<sup>3+</sup>, Nd<sup>3+</sup>, and Ho<sup>3+</sup> In Various Compounds."

A second paper, "Exact Calculation of the Energy and Heat Capacity for the Triangular Lattice With Three Different Coupling Constants," will appear in the January, 1974 issue of J. Math. Phys.

#### D. Other

The helium work mentioned in last year's report was published in the Proceedings of the 13th International Conference on Low Temperature Physics. An extended version of this paper is being written. The project leader spent the first half of the calendar year at the Ecole de Physique et Chimie (University of Paris IV) on a project to study supercooled liquids. The liquid in question, carbon tetrachloride, was studied by means of a laser experiment. The widths of the Kayleigh and Brillouin peaks were followed as a function of the temperature, the scattering angle, and the wavelength. Most of the time of the visit was devoted to obtaining low temperature coefficients from the existing data at higher temperatures in order to obtain data to compare the experiment with the region above the fusion point.

# LOW TEMPERATURE PARAMAGNETISM (EXPERIMENTAL) - R. P. HUDSON AND E. R. PFEIFFER

One of the most interesting features emerging from work on the magnetic temperature scale for CNN has been that the leading term in the high-temperature expansion of the specific heat  $(C/R = \sum_{2}^{\infty} a_n T^{-n})$ , viz.  $b/T^2$ , is *smaller* than one calculates on the basis of dipolar interaction theory.

Because doubts always remain as to the accuracy of involved low-temperature experiments, we thought it worthwhile to make a determined effort to measure the coefficient b in the relatively high 1 K region. We performed this with the crystal used in our very lowtemperature experiments, and used a method which was quite different from conventional calorimetry and capable, in principle, of high accuracy - namely, the Casimir and Du Pré spin-lattice relaxation method. This latter involves measuring the isothermal susceptibility,  $\chi_{\rm T}$ , and the adiabatic susceptibility,  $\chi_{\rm S}$  (determined by employing an ac frequency  $\omega$  such that  $\omega \tau \gg 1$ , where  $\tau$  is the spin-lattice relaxation time) in the presence of a dc field H.

Then

$$C_2 = \frac{\lambda H^2}{T^2} - \frac{\chi_S}{\chi_T - \chi_S}$$

where  $\lambda$  is the Curie constant.

Special attention was paid to achieving good temperature stability, to accurate measurement of T and H, and to realistic assignments of uncertainties in the processing of the data. The value of b which we obtained,  $(5.99 \pm 0.02)10^{-6}K^2$ , confirms our most recent magnetic cooling result, and is slightly smaller than that obtained recently by Abraham et al. (who also used the Casimir-Du Pré method), viz.,  $(6.16 \pm .01)10^{-6}K^2$ .

The theoretical value, as found essentially by all workers (when reduced to the same basis, i.e., of lattice parameters and value for  $g_{\downarrow}$ ) is about 6.6 x 10<sup>-6</sup>, i.e., some 10 percent higher. No other case is known where the experimental value of the magnetic contribution to the heat capacity (now on a very firm footing for CMN) is *less* than the theoretical value. Something appears to be lacking in the simple dipole-dipole coupling theory.

During a five week visit to NBS by B. M. Abraham of Argonne National Laboratory, demagnetization experiments were performed on compacted powder specimens of four different compounds. Pure and lanthanum-diluted CMN were studied along with two new compounds formed

from cerium iodide and antipyrine and from cerium thiocyanate and triphenyl phosphine oxide. The magnetic temperatures,  $T^*$  were determined from measurements of the magnetic susceptibility perpendicular to the axis of the initial magnetizing field. Both the lanthanum-diluted CMN and the triphenyl phosphine oxide compounds yielded significantly lower values of  $T^*$  than were obtained with CMN. A paper describing the results has been accepted for publication by the Journal of Low Temperature Physics.

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