EXPERIMENTAL STUDIES IN SOLID STATE AND
LOW TEMPERATURE PHYSICS

MASTER

Final Report for 1966-1980

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ABSTRACT

Experimental and theoretical investigations have been carried out in a broad area of low temperature and solid state physics which includes superconductivity, theory of quantum crystals (through 1973), magnetism in metals, and liquid helium. The work in superconductivity has involved investigations of the Josephson effect, studies of the pair-field susceptibility of superconductors and investigations of the thermodynamics of the superconducting phase transition. The competition between the metal-nonmetal transition and superconductivity has also been studied in random metal-rare gas systems. In the area of magnetism, magnetically ordered materials and dilute magnetic alloys have been investigated. Enhanced hyperfine nuclear magnetic ordering was discovered in PrCu_6 at about 2.5 mK. The research on liquid ^4He and $^3\text{He}/^4\text{He}$ mixtures has been directed at the quantum aspects of superfluid flow and rotation, the critical behavior near the lambda transition and the properties of the tricritical point. The theoretical program (through 1973) encompassed a broad spectrum of research on the properties of quantum liquids and solids with particular emphasis on crystalline ^3He .

TABLE OF CONTENTS

I.	INTRODUCTION	1
II.	DESCRIPTION OF RESEARCH	2
	A. SUPERCONDUCTIVITY	2
	1. Experiments Relating to the Josephson Effect and the Pair-Field Susceptibility	2
	2. Superconducting Fluctuations and the Critical Behavior of Superconductors	3
	3. Electrical Conductivity and Superconductivity in Random Metal-Gas Solids	4
	B. EXPERIMENTAL STUDIES OF MAGNETIC SYSTEMS	6
	1. Magnetically Ordered Materials	6
	2. Dilute Magnetic Alloys	7
	3. Enhanced Hyperfine Nuclear Magnetic Ordering	7
	C. EXPERIMENTS ON LIQUID HELIUM	9
	1. The Quantization of Circulation in Superfluid ^4He	9
	2. Ions in Liquid ^4He	9
	3. Dynamics of Superfluid Flow Through Fine Regular Pores	10
	4. A Search for Analogs of the Josephson Effect in Superfluid ^4He	10
	5. The Tricritical Point of $^3\text{He}/^4\text{He}$ Mixtures	11
	D. THEORETICAL STUDIES OF THE PROPERTIES OF QUANTUM LIQUIDS AND CRYSTALS	13
III.	PUBLICATIONS AND REPORTS	15

I. INTRODUCTION

The work described in this report consists of various experimental investigations in a broad area which has been called solid-state and low-temperature physics and which is now called condensed matter physics. The research was under the direction of Professors A. M. Goldman, L. H. Nosanow (until 1973), W. V. Weyhmann and W. Zimmermann, Jr. at the School of Physics and Astronomy of the University of Minnesota and was supported under contract DE-AC02-76-ERO 1569.M0003. This program began January 1, 1966 and terminated June 30, 1980.

The scientific scope of the program was quite broad with investigations concerning superconductivity, superfluidity, magnetism in metals and the properties of crystalline helium. Although these various areas of research are quite diverse, they are closely related from several points of view. In the first place most of the properties under investigation are essentially macroscopic manifestations of quantum mechanics. Secondly almost all of the systems exhibit transition to phases in which long-range order appears. Finally, the experimental and theoretical techniques for studying these systems are closely related. In fact in many instances an experiment or calculation on one of these systems has suggested another experiment or calculation in another system.

Apart from the scientific progress reported here and in the publications listed in Section III an important aspect of this program has been its role in the graduate education program at the University of Minnesota. Over the last fourteen years. A large number of capable students in solid-state and low temperature physics were supported in their doctoral research by this contract.

II. DESCRIPTION OF RESEARCH

A. SUPERCONDUCTIVITY

(Professor A. M. Goldman)

During the course of the contract, work in this area has been concentrated on a number of fundamental problems in three general areas. There have been contributions to the understanding of the Josephson Effect and to the development of the study of the pair-field susceptibility as a probe of the dynamics of superconductors. There have also been a number of investigations of superconducting fluctuations and the critical behavior of superconductors. Finally the competition between disorder related metal-nonmetal transitions and superconductivity has been probed in a series of investigations of the electrical properties of random metal-rare gas solids. In the following a brief account of the most important results of this research will be given.

1. Experiments Relating to the Josephson Effect and the Pair-Field Susceptibility.

The work in this area evolved from studies of the Josephson effect in the mid 1960's. The early efforts involved the study of the I-V characteristics of strongly-coupled Josephson junctions, which exhibited a crossover from a Meissner state to a vortex state at a particular value of the magnetic field applied in the plane.¹ This work stimulated the definitive theoretical calculations of Owen and Scalapino.²

In this same period fluctuation rounding of the dc Josephson current was studied.³ The calculations of Ambegaokar and Halperin resulted from efforts to understand the fluctuation-rounded I-V characteristics of planar

Josephson tunneling junctions observed in this laboratory.⁴

The focus of the work in superconductivity then shifted to studies of the generalized susceptibility or pair-field susceptibility of superconductors.⁵ The latter is a quantity proportional to an excess current in the I-V characteristic of an asymmetric tunneling junction where one electrode is the superconductor of interest and the other one a metal with a higher T_C . During the course of the contract, the possibility of measuring the susceptibility was demonstrated using Sn-Pb junctions.⁶ Quantitative studies were carried out using Pb-Al junctions and it was demonstrated experimentally that the susceptibility could be observed in the superconducting as well as the normal state of the electrode of interest.⁷ When this was done, a propagating branch or charge imbalance wave was discovered. The theoretical explanation of this mode, now known as the Carlson-Goldman mode, required detailed consideration of the interaction of the disequilibria of the pairs, quasiparticles and phonons in the superconducting system.⁷

Recent studies of the pair-field susceptibility have been concerned with the effects of magnetic impurities on the dynamics of the order parameter, mode-softening of a superconductor in a current-carrying state, and the pair-field susceptibility of Cooper-limit proximity sandwiches.⁸

2. Superconducting Fluctuations and the Critical Behavior of Superconductors

The original thrust of this work was the study of dimensionality-crossover effects which were observed in NbN thin films with short coherence lengths.⁹ The focus then shifted to studies of heat capacities of superconducting films. The technique of using the resistive transition of a

film as a thermometer was used to make qualitative measurements of heat capacities of Al films near their transitions.¹⁰ Quantitative studies of amorphous-Nb₃Ge films were then carried out using Au(Fe)-Cu thermocouple thermometers and ultra-thin sapphire substrates. In this work, a broad peak in the heat capacity was observed just below the BCS transition.¹¹ The temperature-dependent resistance was also compared with several theories which predict flux-flow resistance below the BCS transition temperature due to thermally excited vortex pairs. Although the transition is broadly consistent with such a model, no feature in $R(T)$ was found that would imply a divergence in the conductivity that might be associated with a topological phase transition. The question probed in this work, that of the existence of topological phase transitions in 2-D systems has attracted significant experimental and theoretical attention in recent years.¹²

3. Electrical Conductivity and Superconductivity in Random Metal-Gas Solids

Metal-gas solids are made by co-depositing metal vapor and gas onto a substrate cooled to low temperatures. Films of Na and NH₃,¹³ Na and Ar¹⁴ and Hg and Xe¹⁵ have been prepared using a special molecular beam oven source in which the gases are mixed at high temperatures and then projected onto a surface held at 4 K.

The focus of the work has been the study of the composition dependence of the metal-nonmetal transition in the case of Na/NH₃ and Na/Ar films and on the competition between the superconducting transition and the metal-nonmetal transition in the case of the Hg/Xe films. In the Na/NH₃ system, the conductivity was found to be a nonmonotonic function of composition, a result which may be explained by what is known as polychromatic percolation.

In the Hg-Xe system, the conductivity as a function of composition appears consistent with microscopic percolation, at which point it crosses over to a regime with a negative temperature coefficient of resistance. In this regime the superconducting transition temperature drops rapidly. The fall of T_c with increasing insulator concentration may be explainable by a superconducting generalization of the random X-Y model.¹⁶

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B. EXPERIMENTAL STUDIES OF MAGNETIC SYSTEMS

(Professor W. V. Weyhmann)

During the course of this contract, several fundamental contributions to our understanding of magnetic properties of materials have been made. In the earlier years this research concentrated on the collective properties of magnetically ordered systems and as time progressed the emphasis shifted to the very low temperature, very weak magnetic properties of dilute magnetic and nuclear magnetic systems. Below we shall highlight the three most important phases of this work.

1. Magnetically Ordered Materials

The discovery that spin reorientation is in some cases a second order phase transition was the thesis work of R. W. Houghton¹ and the interpretation of this result was provided by H. Horner and C. M. Varma,² working at the time at Minnesota on this contract (with Professor Nosalow). The experimental signature of the phase transition was the disappearance of the quadrupole modulation of the nmr spin echo signal, which was interpreted as indicating a fluctuation in the direction of the magnetization. The classical (Landau) theory of the transition predicted a double step (up and then down as temperature increased) in the magnetic contribution to the heat capacity, the steps occurring at the two endpoints of the rotation of the magnetization. This form of the heat capacity was confirmed in an experiment Weyhmann performed in collaboration with G. Sjolander and Prof. M. Moldover.³ While the sharpness of the steps is not yet established, this is one of the few cases (the superconducting transition is another) in which the classical Landau behavior is observed and the only one in which two back-to-back second order transitions are intrinsic to the process.

2. Dilute Magnetic Alloys

The magnetization of dilute magnetic impurities in nonmagnetic, metallic hosts has been an important part of our work for several years. Holliday and Weyhmann⁴ showed that Co in Au has a surprisingly large Knight shift and J. Babcock et al.⁵ showed that Mn in Al remains nonmagnetic at very low temperatures. Another aspect of the work on dilute systems has been the precision measurement of the magnetic saturation behavior of Mn in Cu and Ag, most notably by D. Bakalyar et al.⁶ To the best of our knowledge, this is the only extensive and precise determination of the saturation of a Kondo system in a high magnetic field. All of this work was carried on using nuclear orientation at temperatures between 8 and 30 mK.

3. Enhanced Hyperfine Nuclear Magnetic Ordering

In order to continue these studies at still lower temperatures and to broaden the spectrum of orientation studies that can be pursued, we began an investigation of the magnetic properties of enhanced hyperfine nuclear coolants, particularly PrCu₆. Recently we observed the nuclear ordering transition in PrCu₆ at about 2.5 mK, measuring the magnetization and heat capacity behavior in magnetic fields.⁷ Over the last few months we have observed anomalies in the electrical impedance of a small rod of this material and have noticed what appears to be a second phase transition at very low fields (less than 10 Oe) and slightly higher temperature.⁸ This work opens the door to intensive investigation of the nuclear ordered state in metals where complete thermal equilibrium can be achieved (as opposed to ³He or dipole-dipole ordering in metals such as Cu).

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C. EXPERIMENTS ON LIQUID HELIUM

(Professor William Zimmermann, Jr.)

The research on liquid ^4He and $^3\text{He}/^4\text{He}$ mixtures has been directed at quantum aspects of superfluid flow and rotation, critical behavior near the λ transition, and the properties of the tricritical point.

1. The Quantization of Circulation in Superfluid ^4He

One of the fundamental tenets of superfluidity in helium is quantization of circulation of the superfluid. In order to obtain direct clear-cut evidence in support of this hypothesis, a series of experiments were undertaken using the method first employed by W. F. Vinen. In these experiments studies were made of the superfluid circulation around a fine wire. These experiments culminated in the observation of well-defined stable plateaus of circulation at the predicted quantum values up to 4 quantum units in magnitude.^{1,2} The value of the circulation as a function of angular velocity of rotation of the apparatus was shown to be in general agreement with minimum-free-energy arguments based on quantization of circulation around the wire and the existence of singly-quantized superfluid vortex lines in the liquid.³

2. Ions in Liquid ^4He

Another aspect of such quantum-vortex lines was investigated by means of direct measurements of the lifetime of negative ions trapped on the lines in the rotating liquid as a function of temperature and pressure.⁴ By this means it was possible to estimate the depth of the well in which the ion was trapped and, as a result, the size of the

negative ion, an electron-filled bubble in the helium. That the ion size so determined compares well with other determinations gives indirect support for the existence of vortex lines in the rotating superfluid having one quantum of circulation.

3. Dynamics of Superfluid Flow Through Fine Regular Pores

Superfluid flow through the fine regular channels of Nuclepore filters has been studied near the λ transition for both pure ^4He and $^3\text{He}/^4\text{He}$ mixtures using a superfluid Helmholtz resonator.^{5,6} This work has involved measurements of both the effective superfluid density in the pores and the critical mass current as functions of temperature at constant ^3He mole fraction. Near the λ transition the superfluid density in the pores is depressed below the bulk value, and it tends to vanish at a temperature below the transition whose distance from the transition increases as the channel size decreases or as the ^3He mole fraction increases. The onset of nonzero superfluid density in the pores appears to be governed in general by the ratio of the bulk coherence length to the pore diameter attaining some particular value. The temperature at which the critical mass current goes to zero appears to be the same as the temperature at which the superfluid density vanishes.

4. Search for Analogs of the Josephson Effect in Superfluid ^4He

Recent experiments on superfluid flow have concerned a search for analogs to the Josephson effects in superconductors.⁷ A flow path in the form of a closed loop interrupted by a single weak link having the form of a tiny orifice in a thin, free-standing film is incorporated into a superfluid Helmholtz resonator. Transitions in the circulation state

of the loop are sensed by measuring the Q of the resonance, the operation of this experiment being in strong analogy to that of an rf-biased SQUID. Preliminary results have shown a pronounced step in the resonance response-versus-drive characteristic at the point where dissipation first occurs in the orifice, and one orifice has shown two additional steps which are suggestive of the expected staircase pattern.

5. The Tricritical Point of $^3\text{He}/^4\text{He}$ Mixtures

Research on the tricritical point that occurs in $^3\text{He}/^4\text{He}$ mixtures at saturated vapor pressure at a temperature of 0.867 K and a ^3He mole fraction of 0.675 has consisted of two parts. The first involved an extensive analysis of measurements of the specific heat $C_{p,x}$ that had been performed in Finland.^{8,9} Here p is pressure and x is the mole fraction of ^3He . While $C_{p,x}$ as a function of temperature T at constant x becomes increasingly featureless as x approaches the tricritical value, it was shown that the complementary specific heat $C_{p,\phi}$ along a path of constant ϕ was strongly divergent at the tricritical point as was the concentration susceptibility $(\partial x/\partial \phi)_{T,p}$ along the two branches of the coexistence curve. Here $\phi = \mu_3 - \mu_4$ is the difference between chemical potentials of the ^3He and ^4He components. The second part of the research involved measurements of $(\partial x/\partial \phi)_{T,p}$ near the tricritical point by means of differential osmotic pressure measurements.¹⁰ These measurements revealed a pronounced and divergent or nearly-divergent cusp at the λ transition which had been missed in other work. Subsequent measurements of the gravitational concentration gradient by others confirmed the existence of this cusp and yielded results in good agreement with ours.

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D. THEORETICAL STUDIES OF THE PROPERTIES OF QUANTUM LIQUIDS AND CRYSTALS

(Professor L. H. Nosanow (through 1973))

The work in this field encompassed a broad program of theoretical research on the properties of quantum liquids and crystals. Included in this effort were calculations of the ground state properties of nucleon matter using Monte-Carlo techniques, studies of the liquid-crystal phase transition in large quantum systems using the quantum theorem of corresponding states, investigations of quantum crystal alloys, and of the effects of the exchange operator on the properties of the nuclear spin system of crystalline He^3 . Calculations of the ground state energy and exchange integrals of crystalline He^3 were also carried out. Finally, the superconducting phase transition in a two-component fermion system was investigated.

The work on the ground state properties of nucleon matter was originally motivated by the description of pulsars as rapidly rotating neutron stars. Properties of these stars are sensitive to the equation of state of neutron matter. Some proposed explanations of anomalies in the observed frequencies of pulsation require that neutron matter crystallize somewhere in the density range $3 \times 10^{14} \text{ g/cm}^3$ to $5 \times 10^{15} \text{ g/cm}^3$.

The work on the quantum theory of corresponding states was directed at predicting the solidification pressure and density of neutron matter. The statistics have been found to have a marked effect on the crystallization pressure.

In the investigations of the properties of quantum crystal alloys a statistical mechanical ensemble was found. Using this ensemble, previous

work, which had been thought to depend on the two-body approximation to the cluster expansion and an assumption of randomness, can be seen as a Landau theory of the phase separation with quantum crystal effects taken into account.

III. PUBLICATIONS AND REPORTS

1. C00-1569-1 "University of Minnesota, Experimental and Theoretical Studies in Solid State and Low Temperature Physics Progress Report for 1966."
2. C00-1569-2 "Theory of Quantum Crystals" (L. H. Nosanow).
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15. C00-1569-15 "Progress Report" (1967).
16. C00-1569-16 "A Critique of the Saunders Theory of Solid ^3He at 0°K^+ " (W. J. Mullin) Submitted to Physics.
17. C00-1569-17 "Theory of Quantum Crystals" (L. H. Nosanow, W. J. Mullin and J. H. Hetherington) Presented at the 10th International Conference on Low Temperature Physics.
18. C00-1569-18 "Spin Hamiltonian, Exchange and Spin-Lattice Relaxation in Crystalline ^3He ". (L. H. Nosanow and C. M. Varma) Doctoral dissertation, U of Minn. 1968, unpublished.
19. C00-1569-19 "Shape Dependent Thermodynamic Quantities in Magnetic Systems with Dipole Interaction". (Heinz Horner) To be published.
20. C00-1569-20 "Spin-Lattice Relaxation from Negative Temperatures in Solid ^3He " (C. M. Varma) Phys. Rev. Letters 20 1146 (1968).
21. C00-1569-21 "Exchange in Crystalline ^3He " (L. H. Nosanow and C. M. Varma) Phys. Rev. Letters 20 912 (1968).
22. C00-1569-22 "Nuclear Spin Echo Measurements of Undoped and Cr-Doped Mn_2Sb " (R. W. Houghton and W. Weyhmann) Abstract of Paper presented at the January American Physical Society Meeting.
23. C00-1569-23 "Nature of Spin-Reorientation Transitions" (H. Horner and S. M. Varma) Phys. Rev. Letters 20 845 (1968).
24. C00-1569-24 "Thermodynamic Green's Function Methods in Many Body Theory" (Heinz Horner) Lecture notes, U of Minnesota 1968. Unpublished.
25. C00-1569-25 "Lectures on the Theory of Phase Transitions" (N. G. van Kampen). Lecture notes, U of Minnesota 1968. Unpublished.
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30. C00-1569-30 "Short-Range Correlations and Exchange in Crystalline He^3 " (L. H. Nosanow) Invited talk presented at the International Conference of Pure and Applied Physics at St. Andrews, Scotland.
31. C00-1569-31 "A Nuclear Spin Echo Study of $Mn_{2-x}Cr_xSb$ " (W. Houghton and W. Weyhmann) Abstract submitted to the 14th Conference on Magnetism and Magnetic Materials, New York, November 1968. J. Appl. Phys. 40, 1410 (1969).
32. C00-1569-32 "Progress Report"
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38. C00-1569-38 "Theory of Exchange in Crystalline 3He " (L. H. Nosanow and C. M. Varma) Phys. Rev 187 660 (1969).
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40. C00-1569-40 "Thermal Fluctuations and the Josephson Super-Current" (J. T. Anderson and A. M. Goldman), Phys. Rev. Lett 23 128(1969).
41. C00-1569-41 "Thermal Fluctuations and the Josephson Supercurrent" (J. T. Anderson and A. M. Goldman) Physica 55 256(1971).
42. C00-1569-42 "Lifetimes of Persistent Currents in Superconductive Loops Interrupted by Josephson Junctions" (A. M. Goldman) J. Low Temp Phys 3 27(1970).
43. C00-1569-43 "Many-Body Perturbation Theory for Quantum Crystals" (Heinz Horner).
44. C00-1569-44 "Many-Body Perturbation Theory for Quantum Crystals II. Ladder Approximation (H. Horner).
45. C00-1569-45 "Progress Report (1969).
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