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RICE UNIVERSITY

Nuclear Paramagnetism of Helium Three

by

Harry Alan Schwettman

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Abstract

The technique of adiabatic fast passage has been employed to measure within 1/3 the susceptibility of pure He$_3$ and some He$_3$ - He$_4$ mixtures for temperatures between 0.90K and 3.50K. Above 1.20K the susceptibility varies as the inverse of the temperature, indicating that the statistical and exchange correction to the Curie susceptibility is very nearly constant.

We have calculated the magnetization of an interacting system of He$_3$ particles near the classical limit. Plane waves have been used to evaluate the potential energy and the exchange energy of the particles. In this approximation potential correlation is handled by the introduction of a cutoff radius $a$. For reasonable choices of the cutoff, the statistical correction and the exchange correction are of comparable magnitude.
I. Introduction

The early measurements of the nuclear paramagnetism of \( \text{He}^3 \) were performed in the mid-fifties by W. M. Fairbank and collaborators.\(^1\) At various laboratories, all phases of the early measurements are being repeated and extended. The current investigations can be classified broadly as studies of:

1. the degenerate system..............(Duke, Illinois)
2. the approach to the classical limit...(Rice, Amherst)
3. the solid....................(Duke, Illinois, Oxford)
4. the phase separation in \( \text{He}^3-\text{He}^4 \) mixtures...(Stanford)

Noted after each area of investigation are the laboratories where these studies are being undertaken.

A. Theory

\( \text{He}^3 \) has a nuclear spin of \( \frac{1}{2} \) and therefore is a fermion. The behaviour of a system of fermions has been studied extensively in connection with the problem of electrons in metals. The resulting literature provides a basis for anticipating and interpreting the behaviour of \( \text{He}^3 \).

In a non-interacting system at the absolute zero of temperature, all single particle states are filled up to some energy \( \varepsilon_0 = kT_0 \). For temperatures much less than \( T_0 \) only those particles within \( kT \) of the Fermi surface can be excited and in this sense the system is said to be
degenerate. The degeneracy temperature $T_0$ for conduction electrons in metals is $10^4 \text{ K}$; thus even at room tempera-
ture they represent a degenerate system of fermions.

At temperatures much less than $T_0$ the thermal and magnetic properties of a non-interacting system are easily obtained. The number of excited particles is

$$N_e \sim \int_{\epsilon_0}^{\infty} \nu(\epsilon) f(\epsilon) d\epsilon \sim \nu(\epsilon_0) kT$$

where $\nu(\epsilon)$ is the density of states and $f(\epsilon)$ is the distribution function. We now have the problem of distrib-
tuting a few particles over a large number of states, which is a classical problem. The average excitation energy is $kT$ and the average magnetic moment is ($\mu^{\ast}/kT$) which gives us

$$c \sim \frac{d}{d\epsilon} (N_e \cdot kT) \sim \nu(\epsilon_0) k^2 \cdot T$$  \hspace{1cm} (1.1)

$$\mu \sim N_e \cdot \frac{\mu^{\ast}}{kT} \sim \nu(\epsilon_0) \mu^{\ast}.$$  \hspace{1cm} (1.2)

These are the well known linear specific heat and the limiting Pauli paramagnetism. We have the important result that the properties of the degenerate system are determined by the density of single particle states at the Fermi sur-
face.

The above properties of the non-interacting system can not be a general result for real interacting systems.
For example, an attractive interaction can lead to a strong pairing of particles and a subsequent boson description of the pair. Nonetheless, the linear specific heat and the limiting Pauli paramagnetism are experimentally observed for conduction electrons.

Despite the qualitative success of the non-interacting Fermi gas model, it fails to reproduce the experimental observations in an important respect. As recognized by Böttcher, the ratio of the susceptibility and the specific heat when reduced by the corresponding classical values is a pure number

\[
\lim_{T \to T_0} \frac{\chi_f}{\chi_c} = \frac{2}{\pi^2}.
\]  

This relationship is not preserved for conduction electrons in metals.

It is not difficult to account for such a discrepancy. Huang has considered a repulsive hard-core interaction as a first approximation to an imperfect Fermi gas. In a certain approximation, valid at low temperatures, the energy levels of the \( N \)-particle system are

\[
\mathcal{U} = \sum_k \epsilon_k n_k + \frac{4 \pi a^2}{m V} N^+ N^- - \mu (N^+ - N^-) \mathcal{H}
\]  

where

\[
\sum_k \epsilon_k n_k = \text{Fermi energy of system}
\]
\[
\frac{4\pi a^2}{mV} N^+ N^- = \text{exchange energy of system}
\]
\[
\alpha = \text{hard core diameter}
\]
\[
N^+ = \text{number of particles with spin up}
\]
\[
N^- = \text{number of particles with spin down}
\]
\[-\mu (N^+ - N^-) H = \text{magnetic energy}\]

The exchange term is easy to understand physically. It is smallest when the spins are aligned. This is an expression of the fact that the wave function of any pair of particles with spins aligned will be spatially antisymmetric, thus minimizing the repulsive interaction.

In the absence of an external field the exchange energy is temperature independent. Thus the specific heat of our hard-core Fermi gas is simply the specific heat of the non-interacting system; in the limit of the degenerate system it is determined by the density of single particle states at the Fermi surface.

To calculate the susceptibility we must evaluate the partition function and obtain the free energy. The usual statistical argument leads to a free energy for our N-particle system given by

\[
F = F(N^+) + F(N^-) + \frac{\mu}{3N} \left( \frac{2}{\pi} k_0 a \right) \left[ N^+ - (N^+ - N^-) \right] - \mu (N^+ - N^-) H \quad (1.5)
\]

where the value of \( N^+ \) or \( N^- \) is that which minimizes \( F \). In the expression above, \( F(N^+) \) is the Fermi free energy of
\( N^{\pm} \) particles and

\[
\varepsilon_0 = \frac{h^2}{2m} k_0^2 = \frac{h^2}{2m} \left( \frac{2\pi^2 N}{V} \right)^{\frac{2}{3}}.
\] (1.6)

But \( N^{\pm} = 2(N^+M/\mu) \) and thus the equilibrium magnetization is determined by the condition

\[
\frac{\partial F}{\partial M} = 0 = \left[ \frac{\partial F(N^+)}{\partial N^+} \frac{\partial N^+}{\partial M} + \frac{\partial F(N^-)}{\partial N^-} \frac{\partial N^-}{\partial M} \right] - \frac{2\varepsilon_0}{3N\mu^2} \left( \frac{2}{\pi} \cdot k_0 a \right) M - H. \] (1.7)

Considering the case without exchange, it is evident that the first term above is the magnetization divided by the Fermi susceptibility. Thus the derivative of equation (1.7) with respect to \( H \) gives us the susceptibility of our hard-core Fermi gas. It is

\[
\frac{1}{\gamma} = \frac{1}{\gamma_c} - \frac{2\varepsilon_0}{3N\mu^2} \left( \frac{2}{\pi} \cdot k_0 a \right). \] (1.8)

At the absolute zero \( \gamma_c = \mu(\mu) \mu^2 = 3N\mu^2/2\varepsilon_0 \) and we have

\[
\text{Limit}_{T \to 0} \, \gamma = \gamma_c \frac{\gamma_f}{\gamma_c} \quad \text{or} \quad 1 - \frac{3}{\pi} k_0 a. \] (1.9)

As opposed to the specific heat, the susceptibility is not specified by the density of single particle states at the Fermi surface.

Calculations\(^6,7\) which take detailed account of the collective behaviour of the system support the broad conclusions
of the hard-core model. The specific heat is determined by the density of single particle states at the Fermi surface; the susceptibility is not determined by the density of single particle states at the Fermi surface.

To calculate the properties of a non-degenerate system of fermions we must know the density of states as a function of energy. For a non-interacting system

\[ n(E) \, dE = g_s \cdot \frac{2\hbar^2}{\pi^2} \left( \frac{2m}{\pi\hbar^2} \right)^{3/2} e^{E/\beta} \, dE \]  

where \( g_s \) is the spin degeneracy. The specific heat is calculated as the temperature derivative of the internal energy

\[ C_F = \frac{d}{dT} \left[ \int_0^\infty E \, n(E) \, f(E) \, dE \right] \]

where \( f(E) \) is the Fermi distribution function. The result is

\[ C_F = Nk_B \left[ \frac{5}{2} \frac{F_{n\eta}(\eta)}{F_{n\eta}(\eta)} - \frac{1}{4} \frac{F_{n\eta}(\eta)}{F_{n\eta}(\eta)} \right] \]  

The functions \( F_{n\eta}(\eta) \) are defined as

\[ F_{n\eta}(\eta) = \int_0^\infty \frac{x^n \, dx}{e^{x-\eta} + 1} \]

where \( kT \eta \) is the chemical potential. As the system
approaches the classical limit

\[ \frac{\chi_F}{\chi_c} \sim 1 - \frac{1}{6(2\pi)^{3/2}} \left( \frac{T_0}{T} \right)^{3/2} \]  

(1.12)

The susceptibility of a non-interacting system of fermions can be obtained by evaluating the first term of equation (1.7). Expanding about \( N^+ = N^- = (N/2) \) we obtain

\[ \frac{\chi_F}{\chi_c} = \frac{F_{1b}^{'}(\eta)}{F_{1b}(\eta)} \]  

(1.13)

and as the system approaches the classical limit

\[ \frac{\chi_F}{\chi_c} \sim 1 - \frac{1}{3(2\pi)^{1/2}} \left( \frac{T_0}{T} \right)^{1/2} \]  

(1.14)

For arbitrary \( T/T_0 \) the properties of the non-interacting system can be evaluated using the extensive tables of \( \Phi_n(\eta) \) prepared by McDougall and Stoner.\(^3\)

Once again we want to consider the effect of interactions. Although the energy levels obtained in the hardcore calculation are strictly valid only in the low temperature limit, they exhibit the correct qualitative result that a repulsive interaction tends to align the spins. Thus, as an indication of what one might expect, we can extend this calculation to all temperatures. Since the exchange energy is independent of temperature, \( \mathcal{J} = \mathcal{C}_p \). The susceptibility is given by equation (1.8) which can be written in the reduced form
\[ \frac{\chi}{\chi_c} = \frac{\chi/\chi_c}{1 - \alpha \chi_c} \]  

(1.15)

where \( \alpha \) is a positive constant. This expression is plotted in figure 1 for several values of \( \alpha \). As the system approaches the classical limit we have

\[ \frac{\chi}{\chi_c} \sim 1 + \alpha \chi_c. \]  

(1.16)

Contrary to the result obtained for a non-interacting system [see equation (1.14)], the susceptibility reaches a maximum greater than the Curie law and then approaches the classical limit from above.

B. Experiment

The early measurements of the specific heat and the susceptibility (see figure 2) of helium three suggest that in the low temperature limit the behaviour of the system is very similar to that observed for electrons in metals. The linear specific heat and the limiting temperature independent paramagnetism are preserved, but the relation between them [see equation (1.3)] is not. Studies of the degenerate system in helium three are complicated by the requirement of very low temperatures. Whereas the degeneracy temperature of conduction electrons is \( 10^4 \) \( ^0K \), the calculated degeneracy temperature of non-interacting helium three atoms is \( 5^0K \); experimentally, the characteristic
Figure 1. Plot of the susceptibility of a system of fermions with exchange. Plot is given in reduced form, $\chi/\chi_c$ versus $T/T_c$. Thus the Curie susceptibility appears as a horizontal line at $\chi/\chi_c = 1$. 
Figure 2. Early susceptibility measurements of Fairbank and co-workers. Above: Plot of $\chi/\chi_0$ versus $T$ indicating onset of statistical alignment at low temperatures. Below: Detailed plot of high temperature data in the form $\chi/\chi_0$ versus $T$. Ordinate is percent deviation. Closed circle at $4.2^0K$ is gas point used in normalization.
behaviour of the degenerate system does not appear until temperatures of 0.2 °K are reached. There is a compensating feature. The degenerate He³ system can be studied as a function of density over an interval of ~40 percent which is not possible in the case of electrons in metals.

The objectives of the recent measurements undertaken at Duke¹⁰ and Illinois¹¹ are therefore:

1. The extension of measurements to lower temperatures to demonstrate unambiguously the linear temperature dependence of the specific heat and the temperature independence of the paramagnetism.

2. The investigation of the density dependence of the specific heat and the paramagnetism in the degenerate system.

There has been an additional motivation for measurements at very low temperatures. Application of the JOS theory to helium three suggests the existence of a superfluid phase at extremely low temperatures.¹² This phase has not been observed as yet.

The low degeneracy temperature makes He³ a suitable substance for the study of a Fermi system as it deviates from the classical limit. The magnetic behaviour of He³ in this region is the subject of the present thesis.

We have seen that interactions can alter significantly the magnetic behaviour of the non-degenerate system. If the exchange energy can be written
\[ E_x = J_1 - J_2 \mu^2, \]

which is the case for the hard-core calculation, then \( \frac{\mu}{4\mu} \)
events a maximum and approaches the classical limit from above. This maximum is pronounced for large values of \( J_2 \) but disappears in the limit \( J_2 \to \infty \).

In the low temperature limit the magnitude of the exchange can be estimated by comparing the observed specific heat and the observed susceptibility. Such a comparison suggests a strong exchange effect in the degenerate system, the susceptibility being increased by a factor of four. On this basis one expects to see the effect of exchange in the non-degenerate system, possibly in the form of a maximum as suggested by the hard-core calculation. In any case the Curie law does not represent an upper limit to possible values of the susceptibility.

The early magnetic measurements in He \(^3\) indicate, within rather broad limits, that the susceptibility varies inversely as the temperature above 1.2 \(^0\)K (see figure 2); this dependence is approached from below. Since the inverse temperature dependence extends to temperatures an order of magnitude lower than expected on the basis of the non-interacting Fermi gas model, interactions are important. The latitude permitted by the experimental results, however, makes interpretation difficult. The early experiment is deficient in three respects:

1. the data exhibit a 10\(^\circ\) scatter,
2. there is no check on the temperature independence of the spectrometer sensitivity,

3. no absolute measurement of the susceptibility exists; it is possible that the magnitude of the susceptibility differs appreciably from the Curie value.

Normalization of the early data was attempted by comparison to a measurement in the gas (see figure 2). It was assumed that the non-interacting Fermi gas would be an appropriate description of the physical system. However, the hard-core calculation casts doubt on this procedure. In the high temperature limit it is the exchange, not the statistics, which dominates the magnetic behaviour.

The objectives of the recent measurements performed at Amherst \(^{13}\) and Rice are therefore:

1. the completion of accurate measurements to clarify the behaviour of the high temperature susceptibility,

2. the solution of the normalization problem. It is possible that accurate measurements nearer the classical limit in conjunction with theoretical considerations might provide a method for normalization.

Solid He\(^3\) provides a very simple magnetic system which has also been investigated recently. \(^{14}\) Density variations of greater than 30% can be obtained with reasonable pressures. Three phases of the solid are known to exist. Reliable measurements in the solid have proved quite difficult but the information obtained from magnetic measurements
could be significant.

The phase separation in $\text{He}^5 - \text{He}^4$ mixtures which occurs at temperatures below $-50^\circ\text{K}$, was first observed magnetically. Subsequent observations utilizing other properties of the mixture gave slightly different phase separation curves. The magnetic measurements are being repeated at Stanford to check this discrepancy.
II. Experimental Apparatus and Procedure

We have employed the technique of adiabatic fast passage to measure the nuclear paramagnetism of H. The resonance technique offers the advantages of sensitivity and selectivity. It is sensitive because the magnetization, driven at the oscillator frequency $\omega$, induces a voltage in the pick-up coil which is proportional to $\omega^4$. It is selective because the diamagnetism does not contribute to the induced voltage.

The physical quantity of interest is the susceptibility of a fixed number of particles. Since measurements are made at constant volume, an accurate determination of the susceptibility requires an accurate measurement of the magnetization, an accurate measurement of the temperature, and an accurate assignment of the molar volume.

a. Measurement of the Magnetization

1. Adiabatic Fast Passage

The behaviour of the magnetization of liquid helium three is adequately described by the phenomenological equations of Bloch

$$
\begin{align*}
\dot{M}_x &= -\gamma (M_y H_z - M_z H_y) + \frac{1}{T_2} M_x = 0 \\
\dot{M}_y &= -\gamma (M_x H_z - M_z H_x) + \frac{1}{T_1} M_y = 0 \\
\dot{M}_z &= -\gamma (M_x H_y - M_y H_x) + \frac{1}{T_2} M_z = \frac{1}{T_1} M_0 .
\end{align*}
$$

(3.1)

Here $\gamma$ is the magneto-gyric ratio, $T_1$ is the longitudinal
relaxation time, and $T_2$ is the transverse relaxation time.

The application of an external field

$$H_x = H_x \cos \omega t; \quad H_y = i \H_t \sin \omega t; \quad H_z = H_0(t) \quad H_1 \ll H_0$$

leads to a magnetic resonance for adiabatic variation of $H_0$. The resonance condition is $\theta H_0 = \omega$.

If $H_1$ is sufficiently large, it is possible to sweep through resonance fast enough that relaxation effects can be neglected, but still slow enough to satisfy the adiabatic condition. Such a passage is known as an adiabatic fast passage and the conditions to be satisfied are

$$\frac{H_1}{H_2} \ll \left| \frac{dH_0}{dt} \right| \ll |\omega| H_2$$

Denoting the resonance field by $H^*$ and requiring that $H_0$ is initially greater than $H^*$, the appropriate solutions to the Bloch equations are

$$M_x = \frac{M}{(1 + \delta^2)^{1/2}} \cos \omega t$$
$$M_y = \frac{M}{(1 + \delta^2)^{1/2}} \sin \omega t$$
$$M_z = \frac{M_0}{(1 + \delta^2)^{1/2}}$$

where

$$\delta = \frac{H_0 - H^*}{H_1}$$
and $M$ is the value of the magnetization at the instant passage is begun. As resonance is approached, the magnetization driven at the frequency $\omega$ deviates from the $z$-direction. The magnetization passes through the transverse plane when $\delta = 0$ and for large negative values of $\delta$ is inverted from its initial direction.

The magnetic resonance can be observed as a voltage induced in a coil in the transverse plane. Since this voltage is proportional to the time rate of change of the magnetization along the axis of the coil, the observed signal is out of phase with the driving field, is proportional to $M$, and is "amplified" by the factor $\omega$.

2. Resonance Equipment

The longitudinal field $H_0$ employed in the adiabatic fast passage measurements is produced by a 16 inch Varian magnet. This field can be swept several hundred gauss through the region of resonance by feeding suitable voltages into the sweep input circuit of the magnet power supply. Both the sweep rate and the sweep amplitude are variable.

The scheme for detection of the adiabatic fast passage signal has been described by Low and Rorsmarch. This scheme (see figure 3) employs an eight wave transmission line which transforms the impedance of the tank circuit, $Z_L$, to an impedance $Z_T$ at the top of the line. If the transmission line is terminated by its characteristic impedance, $Z_0$, then
Figure 3. Block diagram of resonance equipment used for detection of the adiabatic fast passage signal.
\[ z_T = z_0 \quad \text{and} \quad \delta z_T = -j \delta z. \]

Thus the reactive impedance change associated with the resonance is transformed to an equal resistive change. This change is detected by a balanced detector, amplified and recorded.

It proves advantageous to use a crystal controlled oscillator for the susceptibility measurements. Since the d.c. output from the detector is proportional to the difference between the oscillator frequency and the tank circuit resonance frequency, this provides a continuous check that the conditions of measurement remain constant. Use of a crystal controlled oscillator creates one problem; the tank circuit resonance is not tuneable in our experiment, and thus a set of crystals is required to match the oscillator frequency to the tank circuit resonance. Fortunately, the Citizens band crystals, at 10 kc intervals around 27 mc, can be used.

For measurements below the lambda temperature in He\(^4\) the tank circuit is surrounded by the He\(^4\) bath. The superfluid bath is very effective in carrying away the heat produced in the resonating tank circuit. Above the lambda temperature, however, the density variation of He\(^4\) requires that the bath be excluded from the region of the tank circuit. It is well known from the design of rf coils\(^{19}\) that the dielectric character of the immediate environment of a coil determines its self-capacitance, and that the
self-capacitance has an apparent effect on its resistance, inductance, and Q. It is observed in our experiment that the density variation of the He$_4$ bath when in contact with the tank circuit results in a large frequency shift. The still larger density variation of the He$_3$ sample, located inside the coil, produces no observable shift.

Above the lambda temperature an exchange gas provides cooling for the sample. With the rf power on, the sample will establish equilibrium at some temperature greater than the bath temperature. To avoid this situation a coaxial switch is inserted between the oscillator and the transmission line. The oscillator output is fed into a 50 ohm termination, and then switched to feed the tank circuit only long enough to make a measurement.

The resonance signal is recorded on a Sanborn model 320 dual channel d. c. amplifier-recorder. The detector output is amplified in one channel and then fed into a potential divider. The signal is then fed to the second channel. The potential divider is set prior to each measurement so that the signal amplitude recorded on the second channel will be nearly constant. This measure eliminates the problem of non-linearity in the recorder.

3. Procedure

The magnetization is permitted to grow to its equilibrium value prior to passage through resonance. If the sample is initially unmagnetized, a steady state
magnetization will be approached according to the expression (see Appendix A)

\[
\frac{M(t)}{M_0} \rightarrow \left[1 - \frac{T_1/T_2}{\delta^2}\right]\left[1 - e^{-t/T_1}\right]
\]

(3.6)

where \(\delta\) is defined in equation (3.5). After waiting a time \(6T_1\), the magnetization has grown to within 0.25\% of its steady state value. This steady state value, however, is the equilibrium value only if \(\delta' \gg T_1/T_2\). For our experiment \(T_1/T_2 \sim 10\) and \(\delta \sim 100\), and thus there is a negligible difference between the equilibrium magnetization and the steady state magnetization.

Having established equilibrium, the large field \(H_0\) is swept through its resonance value subject to the conditions of equation (3.3). Under the least favorable conditions \(T_2 \sim 3\) sec., \((dH_0/dt) \sim 25\) gauss/sec., and \(H_1 \sim 1/3\) gauss. Thus with \(\theta \sim 2 \times 10^4\) equation (3.3) becomes

\[.1 \ll .25 \times 10^2 \ll .2 \times 10^4\]

and the conditions of adiabatic fast passage are satisfied by two orders of magnitude. The methods of measuring the relaxation times and \(H_1\) are described in Appendix A.

The temperature independence of the spectrometer sensitivity has been checked in our experiment. The fact that the tank circuit frequency is observed to be constant during
a set of measurements augurs well for the temperature independence of the $Q$. We have made a direct check on this by measuring the strength of the proton signal as a function of temperature (see figure 7). The nylon sample chamber provides a convenient proton sample.

B. Cryogenics

The dewar system is designed to obtain a one inch working space in the one and three quarters inch magnet gap. The capacity of the helium dewar is one and one half liters.

Significant contributions to the total energy transfer into the helium bath arise from room temperature radiation funnelled down the helium dewar and from conduction along the transmission line. The radiation is limited by a shield located just below the ring seal of the helium dewar. Conduction along the transmission line is minimized by stainless steel construction. With this system temperatures as low as $0.92^{°}K$ can be maintained twenty hours.

The temperature of the sample can be inferred from the vapor pressure of the helium bath. The pressure is sensed as near the liquid level as practicable. With a three-eights inch diameter sensing line the thermomolecular pressure difference yields a temperature correction of less than one-half percent above $1.10^{°}K$. Above $1.5^{°}K$ the pressure is measured with a mercury manometer and a cathetometer; below this temperature the pressure is measured with a
Stokes-McLeod gauge.

Above the lambda point, temperature measurement is complicated by the presence of thermal gradients in the bath. If the bath temperature is lowered monotonically during the course of an experiment, cooling of the liquid below the surface proceeds first by evaporation and then by convection. Conduction is too slow to be important.

Large temperature gradients are reduced by evaporation. The pressure at any level \( h \) below the surface is:

\[
P = P_{\text{top}} + \rho g h.
\]

Thus evaporation is an effective means of heat transfer as long as the gradient \( \frac{dT}{dh} \) satisfies the condition

\[
\frac{dT}{dh} > \rho g \left( \frac{dT}{dP} \right)_{\text{Vapor Pressure}}.
\]

(3.7)

Convection can reduce the temperature gradient further. We have

\[
\frac{dP}{dh} = \left( \frac{\partial P}{\partial P} \right)_T \frac{dP}{dh} + \left( \frac{\partial P}{\partial T} \right)_P \frac{dT}{dh}
\]

\[
= \rho \kappa \cdot \rho g - \rho \beta \cdot \frac{dT}{dh}
\]

and convection is an effective means of heat transfer so long as

\[
\frac{dT}{dh} > \rho g \frac{\kappa}{\beta}.
\]

(3.8)

where

\[
\kappa = -\frac{1}{V} \left( \frac{\partial V}{\partial P} \right)_T \quad \text{and} \quad \beta = \frac{1}{V} \left( \frac{\partial V}{\partial T} \right)_P.
\]
The actual temperature gradient in a particular experiment depends on the heat input but normally is bounded by the values given in equations (3.7) and (3.8). In He\textsuperscript{4} the gradient given by equation (3.7) is greater than that given by (3.8). We have assumed the full value

$$\Delta T = \rho g h \left( \frac{\Delta T}{\Delta P} \right)_{\text{vapor Pressure}}$$

in correcting our temperature measurements. The correction is less than 1.5.

Below the lambda point the problem of thermal gradients is solved by the appearance of superfluidity. Unfortunately, superfluidity is accompanied by the mobile film and the consequent problems of film reflux.\textsuperscript{20} Furthermore, at lower temperatures the vapor pressure of He\textsuperscript{4} becomes quite small. Therefore in the temperature interval 0.9\textdegree K - 2.0\textdegree K He\textsuperscript{3} vapor pressure thermometry is used. At 1.0\textdegree K the vapor pressure of He\textsuperscript{3} is two orders of magnitude greater than He\textsuperscript{4}, and there is no mobile film.

The He\textsuperscript{3} thermometer is a closed system consisting of a vapor pressure bulb, a pressure-sensing tube, and a manometer. The dimensions of the system and the quantity of gas are fixed such that liquefaction of the He\textsuperscript{3} begins at 2.0\textdegree K and the bulb is eight-tenths full at 0.9\textdegree K. The diameter of the pressure-sensing tube is .030 inches, and thus above 0.90\textdegree K the temperature correction arising from thermomolecular pressure difference is a small fraction of
one percent. Vapor pressure measurements of the bath serve as a check of the temperature measurement.

C. Sample

To the extent that $\text{He}_4$ represents an inert matrix in which $\text{He}_3$ can be diluted, the magnetic measurements can be extended toward the classical limit through the use of $\text{He}_3 - \text{He}_4$ mixtures. Measurements have been made in pure $\text{He}_3$ under its saturated vapor pressure from 0.92°K to 2.9°K. Measurements also have been made in mixtures of 81% $\text{He}_3$ and 70% $\text{He}_3$ over the same temperature range.

As indicated in figure 4 the concentration of the mixtures was selected to provide a density overlap. For this purpose it is unnecessary to prepare mixtures of an exact predetermined concentration. A Toepler pump with accomodations for a $\text{He}_3$ source, a $\text{He}_4$ source, a sample container and a line for evacuating the system proves adequate. Using the Toepler pump as a manometer, the quantity of $\text{He}_3$ and $\text{He}_4$ pumped into the sample container can be measured and the concentration fixed within two percent. Samples of 250 milliliters were prepared in this way.

Since the susceptibility measurements are relative, it is unnecessary to know the sample concentration with great accuracy. The temperature variation of the molar volume is a strong function of concentration only in the region of high temperature and low $\text{He}_4$ content.
Figure 4. Molar volume of the He³ component for the three mixtures used in this experiment.
Concentrations were therefore inferred within one percent from vapor pressure measurements. In the mixtures of low He content the concentrations were checked with a mass spectrometer. The concentration measurements are enumerated in Table 1.

Table 1

<table>
<thead>
<tr>
<th>Sample</th>
<th>Concentration (% He³)</th>
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<tr>
<td>Vapor Pressure</td>
<td>Mass Spectrometer</td>
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<td>A</td>
<td>100</td>
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<td>B</td>
<td>50</td>
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Figure 5 is a schematic of the gas handling system. When the bath temperature has been reduced to 2.0 K (vapor pressure less than 150 mm Hg) gas is admitted to the sample chamber. Condensable vapors are eliminated by a helium trap in the gas line. When a sufficient amount of gas has condensed, the valve to the sample chamber is closed, the trap warmed, and the excess gas in the trap returned to the storage container using the U-tube as a Toepler pump. The valve to the storage container is closed, the valve to the sample chamber opened, and the experiment is begun. The U-tube is now used to monitor the sample pressure.

The sample chamber, illustrated in figure 6, is constructed of nylon. Considerable difficulty was experienced
Figure 5. Schematic of the gas handling system.
Figure 6. Scale drawing of nylon sample chamber.
in the construction of a chamber which was reliable. The nylon chamber illustrated is force fit over the brass plug and has proved quite satisfactory.

Molar volume data for pure He\textsuperscript{3} has been taken from the work of Sherman and Edeskuty.\textsuperscript{21} Data for the mixtures have been taken from the work of Kerr.\textsuperscript{22} Molar volumes for mixtures of He\textsuperscript{3} content greater than 50\% are presented in Table 2. These volumes are unpublished and represent interpolation from a limited amount of data. The estimated error in the molar volumes is typically 0.3\%.

Although the accuracy of the molar volume data is quite good, significant errors can arise in the process of assigning volumes in particular experiments. It is not always possible to maintain the ideal thermal conditions of the molar volume experiments. Two errors are relevant to the present work. First, an error in the temperature measurement produces an improper assignment of the molar volume. Unfortunately, too high a temperature results in assigning too large a molar volume. In the limit of high temperature and high He\textsuperscript{3} content the error in assigning the molar volume is larger than the original temperature error. It is for this reason that our measurements extend to temperatures no higher than 2.9\textdegree K. Second, considerable errors can arise from variations of concentration during the course of an experiment. There is a very strong distillation effect in He\textsuperscript{5} - He\textsuperscript{4} mixtures and the literature
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presents numerous examples where significant discrepancies can be attributed to variations in concentration. \(^{24}\) Sydoriak and Roberts \(^{20}\) have enumerated some of the conditions which can lead to variations of concentration. The procedure of lowering the temperature monotonically must be adhered to rigorously!
III. Experimental Results and Discussion

It is conventional to plot the susceptibility data relative to the Curie law. Since the normalization is arbitrary, our data are presented in the form $\chi T$ versus $T$. The ordinate is percent deviation.

For each of the three samples studied (100\% He\textsuperscript{3}, 81\% He\textsuperscript{3}, 70\% He\textsuperscript{3}) the susceptibility exhibits an inverse temperature dependence above 2\textdegree K. As shown in figure 7, the scatter of the data about the inverse temperature dependence is ± 1\%. Each point represents the average of three or more measurements; the scatter in the individual measurements is typically ± 0.5\%. In this region the temperature measurement and the molar volume assignment represent the principal sources of error. The proton measurements used to check the temperature independence and the linearity of our spectrometer also are plotted in figure 7.

Measurements from 0.92\textdegree K to 2.0\textdegree K indicate that the susceptibility approaches the inverse temperature dependence from below. Data for the same three samples are presented in figure 8. The deviation from an inverse temperature dependence is very small above 1.2\textdegree K. At 1.2\textdegree K the data suggest a possible 2\% deviation. Below this temperature $\chi T$ falls more rapidly. Within experimental error the temperature dependence is the same for each of the three
Figure 7. Plot of high temperature data (in form $\chi T$ versus $T$) indicating inverse temperature dependence of the susceptibility. Ordinate is percent deviation. The proton measurements serve as a check of the temperature independence and the linearity of our spectrometer.
Figure 8. Approach to the inverse temperature dependence. Plotted as $\chi T$ versus $T$. Ordinate is percent deviation.
samples. The temperature measurements and molar volume assignment are improved in this region; the scatter in the data is \( \pm \frac{1}{2} \% \).

The data for the three samples are not experimentally normalized relative to one another. However, the considerable density variation in each sample as a function of temperature and the considerable density overlap between samples suggests that the susceptibility is independent of density over the region studied. This conclusion is supported by measurements of the susceptibility as a function of pressure at 1.20K; no density dependence is observed from the vapor pressure curve to the liquid-solid curve.

Interpretation of these measurements is complicated by the absence of normalization relative to the Curie law. But regardless of normalization, the results present an interesting problem. Let us write the susceptibility as

\[
\chi = \chi_c \left[ 1 - f(\rho, T) \right]
\]

where \( f(\rho, T) \) represents the statistical and exchange correction to the Curie susceptibility. The experimental results then require that \( f(\rho, T) \) be very nearly a constant for greater than 100\% variations of density and temperature. And this must be true in a region where one expects large deviations from the Curie susceptibility on the basis of a non-interacting Fermi gas model.

Recent measurements by Romer\(^{13}\) suggest that detailed
study of the susceptibility in the gas will provide a method for normalization. For low densities at 300K the susceptibility exhibits the same density dependence as the non-interacting Fermi gas (see figure 9). Extrapolation to zero density should then give the Curie value. If Romer extends his measurements to liquid densities, all data can be normalized relative to the susceptibility obtained from the zero density extrapolation.
Figure 9. Romer's susceptibility measurements in the gas at 2.99 K.
IV. **Theory**

We have calculated the magnetization of an interacting system of $^3$He particles near the classical limit. Plane waves have been used to evaluate the potential energy and the exchange energy of the particles. In this approximation potential correlation is handled by the introduction of a cutoff radius $a$.

The occupation numbers may be obtained from the variation of the free energy subject to the condition that the total number of particles is $N$. The variational function is

\[
\sum_k n_k^+ \left[ e_k + \frac{1}{2} \sum_{k'} n_{k'}^+ \left[ V + J(k, k') \right] - \mu \right] - \lambda
\]

\[
+ kT \sum_k \left[ n_k^+ \ln n_k^+ + (1 - n_k^+) \ln (1 - n_k^+) \right]
\]

\[
+ \sum_k n_k^- \left[ e_k + \frac{1}{2} \sum_{k'} n_{k'}^- \left[ V + J(k, k') \right] + \mu \right] - \lambda
\]

\[
+ kT \sum_k \left[ n_k^- \ln n_k^- + (1 - n_k^-) \ln (1 - n_k^-) \right]
\]

(5.1)

where

$\vec{n}_k^+$ = occupation number for state $k$ and spin up

$\vec{n}_k^-$ = occupation number for state $k$ and spin down

* This function has been obtained by a slight generalization of the expression for the free energy of an ideal gas to include interactions between particles in the plane wave states $K$ and $K'$. The direct and exchange interactions are given by $V$ and $J(K, K')$, respectively.
\[ \epsilon_k = \frac{\hbar^2 k^2}{2m} \]
\[ V = -\frac{4\pi}{V} \int_0^\infty V(r) r^2 dr \]
\[ J(k, k') = -\frac{4\pi}{V} \int_0^\infty \frac{\sin |k-k'| r}{|k-k'| r} V(r) r^2 dr \]
\[ \lambda = \text{chemical potential}. \]

The variation of equation (5.1) with respect to the occupation numbers is

\[ \sum_k \left[ \epsilon_k + \frac{1}{2}(N-1)V + \frac{1}{2} \sum \frac{\theta}{e^\theta - 1} J(k, k') \right] = \mu H - \lambda + kT \ln \frac{n_{k}^\pm}{1-n_{k}^\pm} \]

and setting this equal to zero we obtain

\[ kT \ln \frac{n_{k}^\pm}{1-n_{k}^\pm} + \epsilon_k + \mu H - \lambda' = -\frac{1}{2} \sum \frac{\theta}{e^\theta - 1} J(k, k') \]

where

\[ \lambda' = \lambda - \frac{1}{2}(N-1)V. \]

The occupation numbers may be determined by iteration of equation (5.4). Near the classical limit with \( J \approx 0 \) the occupation numbers are

\[ n_{k}^\pm = \exp \left[ \frac{\theta}{kT} (-\epsilon_k \mp \mu H + \lambda') \right] - \exp \left[ \frac{\theta}{kT} (-\epsilon_k \mp \mu H + \lambda') \right]. \]
Iterating this we obtain

\[ n^e_k = \exp \left[ \frac{-\epsilon_k + \lambda'}{kT} \right] \left\{ \left[ 1 - \frac{1}{kT} \cdot \frac{1}{2} \sum_k \exp \left[ \frac{-\epsilon_k + \lambda'}{kT} \right] J(k,k') - \exp \left[ \frac{-\epsilon_k + \lambda'}{kT} \right] \right] \right\} \]

\[ \pm \frac{N^H}{kT} \left[ 1 - \frac{2}{kT} \cdot \frac{1}{2} \sum_k \exp \left[ \frac{-\epsilon_k + \lambda'}{kT} \right] J(k,k') - 2 \exp \left[ \frac{-\epsilon_k + \lambda'}{kT} \right] \right] \]

(5.6)

To calculate the magnetization to first order, terms must be retained to first order in \( n^e_k + n^e_k \) and to second order in \( n^e_k - n^e_k \). Now \( \lambda' \) is determined by the relation

\[ N = \sum_k (n^e_k + n^e_k) = 2 \sum_k \exp \left[ \frac{-\epsilon_k + \lambda'}{kT} \right] - \frac{2E_x}{kT} - 2 \sum_k \exp \left[ \frac{-\epsilon_k + \lambda'}{kT} \right] \]

(5.7)

where the exchange energy \( E_x \) is

\[ E_x = \frac{1}{2} \sum_k \exp \left[ \frac{-\epsilon_k + \lambda'}{kT} \right] \sum_k \exp \left[ \frac{-\epsilon_k + \lambda'}{kT} \right] J(k,k') . \]

(5.8)

The magnetization is

\[ M = \mu \sum_k (n^e_k - n^e_k) = \frac{N^H}{kT} \left[ 2 \sum_k \exp \left[ \frac{-\epsilon_k + \lambda'}{kT} \right] - \frac{4E_x}{kT} - 4 \sum_k \exp \left[ \frac{-\epsilon_k + \lambda'}{kT} \right] \right] \]

(5.9)

and using equation (5.7) we have to first order

\[ M = \frac{N^H}{kT} \left[ 1 - \frac{2E_x}{NK^T} - \frac{2}{N} \sum_k \exp \left[ \frac{-\epsilon_k + \lambda'}{kT} \right] \right] . \]

(5.10)
Here we recognize $N \mu^2 H / K T$ as the Curie magnetization, $-2E_x / N K T$ as the exchange correction and $\frac{-1}{N} \sum \exp \left[ \frac{2}{K T} (-\epsilon_x + \lambda) \right]$ as the statistical correction. The zero order expression for $\exp (\lambda' / K T)$

$$\exp (\lambda' / K T) \sim N \sqrt{\frac{2}{N K T}} \exp (-\epsilon_x / K T)$$ (5.11)

now may be used to calculate our first order corrections to the magnetization. The statistical correction is

$$\frac{-2N}{\sqrt{N K T}} \left[ \frac{k_b^3}{32 (\pi m k_b^2)^{3/2} T^{1/2}} \right] \approx \frac{-2N}{\sqrt{N K T}} \left[ \frac{1.24 \times 10^{-26}}{T^{1/2}} \right].$$ (5.12)

To calculate the exchange correction we must evaluate the total exchange energy $E_x$

$$E_x = \frac{-\pi N^2 k_b}{2V (2\pi m k_b^2)^2} \int dE_1 \exp (-p^2 k_1^2) \int dE_2 \exp (-p^2 k_2^2) \int_0^{2\pi} \frac{\sin k_z k_2 r}{k_2 k_z} V(r) \, dr. \quad (5.13)$$

Here we use the Slater-Kirkwood potential $V(r)$

$$V(r) = A e^{-\alpha r} - B r^{-6} \quad (5.14)$$

where
\[ A = 77 \times 10^{-11} \text{ erg} \]
\[ B = 0.149 \times 10^{-59} \text{ erg} \]
\[ \alpha = 4.60 \times 10^6 \text{ cm}^{-1}. \]

The variables \( k_1 \) and \( k_2 \) can be separated by use of the Gegenbauer Addition Theorem

\[
\frac{\sin|k_1-k_2|r}{|k_1-k_2|r} = \frac{2}{r} \sum_{m=0}^{\infty} (m+\frac{1}{2}) \frac{J_{m+\frac{1}{2}}(kr)}{J_{m+\frac{1}{2}}(k_1)} \frac{J_{m+\frac{1}{2}}(kr)}{J_{m+\frac{1}{2}}(k_2)} P_m (\cos \theta) \quad (5.15)
\]

where \( \theta \) is angle between \( k_1 \) and \( k_2 \). If we choose a coordinate system with \( k_1 \) along the z-axis, the angular integration in \( k_2 \) gives \( 4\pi \delta_{m,0} \). The angular integration in \( k_1 \) gives \( 4\pi \) and we obtain

\[
E_x = \frac{-N^2 \hbar^2}{V(mkT)^2} \int_0^{\infty} r^2 dr \int_0^{2\pi} d\theta \sin \theta \left[ \int_0^{\infty} J_{\frac{1}{2}}(kr) \exp \left( -\frac{\hbar k^2}{2m} \right) r^2 \frac{dr}{r} \right]^2. \quad (5.16)
\]

The integral over \( k \) is

\[
\int_0^{\infty} J_{\nu}(at) \exp (-p^2 t^2) t^{\nu-1} dt = \frac{\Gamma(\frac{\nu+1}{2})}{2p^{\nu-1}} \frac{\Gamma(\nu+1)}{\Gamma(\nu+1)} \cdot F_1 \left( \frac{\nu+1}{2}; \frac{\nu+1}{2}; \frac{-a^2}{4p^2} \right). \quad (5.17)
\]

In our case \( \nu = \frac{1}{2}, a = r, t = k, p^2 = \hbar^2 / 2mkT, \mu = 5/2, \) and \( F_1 = \exp (-p^2/4p^2). \) The exchange energy is

\[
E_x = \frac{-N^2}{V} [G] \quad (5.18)
\]
and the exchange correction to the magnetization is

$$
\frac{+2N}{VkT} [G]
$$

(5.19)

Here

$$
[G] = A \int_a^\infty r^2 dr \exp \left[-\alpha r -(r^2/2\beta^2) \right] - B \int_a^\infty \frac{dr}{r^4} \exp \left(-r^2/2\beta^2 \right)
$$

(5.20)

which upon integrating and expanding to lowest order becomes

$$
[G] = A e^{-\alpha a} \left[\frac{a^2}{2\alpha} + \frac{2a}{\alpha^2} + \frac{2}{\alpha^3} \right] - \frac{B}{\beta} \cdot \frac{1}{a^2}
$$

(5.21)

$[G]$ has been calculated as a function of $a$. The results are shown in figure 10. For reasonable choices of the cutoff, the statistical correction and the exchange correction are of comparable magnitude. Thus there exists a reasonable possibility that the magnetic behaviour of a low density system of $\text{He}^3$ can be explained in this approximation. A realistic comparison to experiment, however, would require an independent determination of the cutoff radius, "$a". This could be done by considering "$a" as a variational parameter chosen to minimize the total energy. Alternatively, the two-body collision problem could be solved to obtain more realistic wave-functions, which could then be used to calculate the exchange without the introduction of a cutoff.
Figure 10. Plot of \(-[G]\) as a function of the cutoff radius "a".
Appendix

A. Strong Field Solutions to Bloch Equations

If the transformation

\[ \begin{align*}
    \mathbf{V} &= \frac{1}{M_0} (M_x \cos \omega t + M_y \sin \omega t) \\
    \mathbf{W} &= \frac{M_0}{M} \\
    \mathbf{V} &= \frac{1}{M_0} (-M_x \sin \omega t + M_y \cos \omega t)
\end{align*} \]

is applied, the Bloch equations can be written

\[ \begin{align*}
    \dot{\mathbf{U}} + p \mathbf{U} + \delta \mathbf{V} &= 0 \\
    \dot{\mathbf{V}} + \rho \mathbf{V} - \delta \mathbf{U} + \mathbf{W} &= 0 \\
    \dot{\mathbf{W}} + \mathbf{W} - \mathbf{V} &= \alpha.
\end{align*} \]

Here

\[ \begin{align*}
    \mathbf{J} &= \mathbf{B} \mathbf{H}_1 \mathbf{T} ; \quad \alpha = \frac{1}{\mathbf{B} \mathbf{H}_1 \mathbf{T}_1} ; \quad \rho = \frac{1}{\mathbf{B} \mathbf{H}_1 \mathbf{T}_2} ; \quad \delta = (\mathbf{B} \mathbf{H}_0 - \mathbf{w}) / \mathbf{B} \mathbf{H}_1
\end{align*} \]

and the derivative is taken with respect to \( \mathbf{J} \).

The Bloch equations can be solved by Laplace transform assuming \( \delta = 0 \). The solutions will be valid for adiabatic variation of \( \delta \). For large \( \mathbf{H}_1 \), or the strong field case, \( \alpha \) and \( \rho \) are much less than one and the solutions to zero order are
\[ U(t) = \frac{\delta}{\delta^* + \beta \omega} (1 - e^{-\delta^* t}) + B \sin \theta \ e^{-\delta^* t} + A \cos \theta \ \cos(\omega t + 4) \ e^{-\beta s} \]

\[ V(t) = C(\beta) + C(\omega - \beta) + \cos(\omega t + 4) \ e^{-\beta s} \]

\[ W(t) = \frac{\delta^*}{\delta^* + \beta \omega} (1 - e^{-\delta^* t}) + B \cos \theta \ e^{-\delta^* t} - A \sin \theta \ \cos(\omega t + 4) \ e^{-\beta s} \]

where

\[ A = \left[ V_0^2 + (U_0 \cos \theta - W_0 \sin \theta)^2 \right]^{1/2} \]

\[ B = W_0 \cos \theta + U_0 \sin \theta \]

\[ \psi = \tan^{-1} \frac{V_0}{U_0 \cos \theta - W_0 \sin \theta} \]

\[ s = (1 + \delta^*) \theta \]

\[ \delta = e^{\gamma \theta} \]

\[ a = \frac{\delta^* + T_1/T_2}{\delta^* + 1} \]

\[ \beta = a^{-1} \]

\[ \beta = \beta \left[ 1 - \frac{1 - T_2/T_1}{z(\delta^* + 1)} \right] \]

If passage through resonance is very slow compared to the relaxation times, the exponentials in equations (a.4) approach zero and we have the slow passage solution

\[ U = \frac{\delta}{\delta^* + \beta \omega} \quad V = 0 \quad W = \frac{\delta^*}{\delta^* + \beta \omega} \]

The deviation of the magnetization

\[ M_0 (U^2 + V^2 + W^2)^{1/2} \sim M_0 \left[ \delta(1 + \delta^*)^{1/2} / (\delta^* + T_1/T_2) \right] \]
from the $z$-direction is

$$\tan \phi = \left( \frac{U^* + V^*}{W} \right)^{1/2} \sim \frac{1}{\delta} \quad .$$  \hspace{1cm} (A.8)

For $\delta \gg T_1/T_2 \gg 1$ the magnetization is $M_0$ and $\phi = 0$.

As $\delta$ approaches zero the magnetization is reduced in magnitude and deviated from the $z$-direction. At resonance

$\delta = 0$, the magnetization is zero, and $\phi = \pi/2$.

If $H_1$ is sufficiently large, it is possible to satisfy the adiabatic condition and still traverse resonance in a time short compared to the relaxation times. The conditions for this adiabatic fast passage are

$$\rho = \frac{\delta}{\rho} \approx 1 \quad .$$  \hspace{1cm} (A.9)

Then $\exp(-a \tau)$, $\exp(-b \tau) \sim 1$ and we look for a solution of the form $U = U_0$, $V = V_0$, $W = W_0$. We have

$$U = \frac{M}{M_0} \sin \theta \quad ; \quad V = 0 \quad ; \quad W = \frac{M}{M_0} \cos \theta \quad .$$  \hspace{1cm} (A.10)

where $M$ is the magnetization at the instant passage is begun.

For an adiabatic fast passage the initial conditions are $B = M$, $A = 0$. If we choose $B \neq 0$ and $A \neq 0$, the final terms of equations (A.4) correspond to a precession about the adiabatic fast passage solution with the frequency
\[ \Omega = (1 + \delta)^{1/2} H_1. \]

Various parts of the strong field solution can be applied to the measurement of \( H_1, T_2, \) and \( T_1 \). Those methods which prove most convenient in the case of \( \delta \) will be discussed below.

Measurement of \( H_1 \): From equations (A.10) and (A.5) we see that the adiabatic fast passage solution is proportional to \((1 + \delta)^{-1/2}\) and that \( \delta = (H_0 - H^*)/H_1 \). Thus \( H_1 \) can be determined from the width of the adiabatic fast passage signal.

Measurement of \( T_2 \): From equations (A.4) and (A.5) we see that for \( \delta = 0 \) the adiabatic fast passage solution decays as \( \exp(-t/T_2) \). If \( H_1 \) is swept adiabatically to its resonance value, the transverse relaxation time \( T_2 \) can be observed as the decay of the signal amplitude.

Measurement of \( T_1 \): From equations (A.4) and (A.5) we see that for \( \delta \ll T_1/T_2 \) the slow passage solution grows toward its equilibrium value as \( [1 - \exp(-t/T_1)] \). By maintaining \( \delta = 0 \) for a time long compared to \( T_2 \) the magnetization can be saturated. If \( \delta \) is made large compared to \( T_1/T_2 \) the magnetization will grow toward its equilibrium value. By performing an adiabatic fast passage the magnetization can be sampled at some later time \( t \). A set of such measurements for different \( t \) gives \( T_1 \).
5. Magnetic Properties of an Imperfect Fermi Gas

MAGNETIC PROPERTIES OF AN IMPERFECT FERMI GAS

KERSON HUANG

1. DEFINITION OF THE MODEL

We consider $N$ particles, each of mass $m$ and spin $(1/2)\hbar$, obeying Fermi statistics, and enclosed in a box of volume $V$. The particles interact with one another through repulsive hard cores of diameter $a$. The Hamiltonian of this system can, in a certain approximation, be written as follows:

$$H = \sum_{i=1}^{N} \frac{p_i^2}{2m} + \frac{4\pi a^2}{m} \sum_{i=1}^{N} \frac{1}{r_i} \frac{\partial}{\partial r_i} \cdot r_i.$$  \hspace{1cm} (1)

This approximation is valid if the core diameter $a$ satisfies the condition

$$ka << 1$$  \hspace{1cm} (2)

where $k$ is the relative wave number of any pair of interacting particles. It is therefore a low-temperature approximation.

Treating the interaction term in (1) as a perturbation, and calculating the energy levels of the $N$-particle system to first order in $a$, one easily obtains the result:

$$E = \sum_{n} \varepsilon_n n_s + \frac{4\pi a^2}{mV} N_s N_-,$$  \hspace{1cm} (3)

where

- $\varepsilon_n$ = unperturbed single-particle energies $= \hbar^2 n^2 / 2m$
- $n_s$ = occupation number of the single-particle energy level $\varepsilon_n$ in the unperturbed wave function.
- $N_s$ = total number of particles with spin "up",
- $N_- = N_s N_-$ = total number of particles with spin "down".

Formula (3) is easy to understand physically. It is smallest when the particle spins are aligned. This is an expression of the fact that the wave function of any pair of particles with spins aligned will be spatially antisymmetric, thus minimizing the repulsive interaction.

The model under consideration consists of a fictitious system whose energy levels are given exactly by (3). Furthermore, the particles shall each have a magnetic moment $\mu$. In the presence of an external magnetic field $H$, the energy levels are

* Department of Physics and Laboratory for Nuclear Science, Massachusetts Institute of Technology, Cambridge, Massachusetts, U.S.A.
accordingly:

\[ E = \sum_{s} e_{s} n_{s} + \frac{4\pi a h^{4}}{m V} N_{+} N_{-} - (N_{+} - N_{-}) \mu H. \]  

(4)

Introducing the parameter

\[ r = (N_{+} - N_{-})/N, \quad (-1 \leq r \leq +1), \]  

(5)

we can also write

\[ E = \sum_{s} e_{s} n_{s} + \frac{2}{3} N e_{r} \left( \frac{k T a}{\pi} \right) (1 - r^{2}) - \mu H N r, \]  

(6)

where

\[ e_{r} = \frac{\hbar^{2} k^{2}}{2m} = \frac{\hbar^{2}}{2m} \left( \frac{3\pi^{2} N^{\frac{2}{3}}}{V} \right) \]  

(7)

is the Fermi energy.

We shall evaluate the partition function of the system, and calculate the average value of \( r \) at a given temperature. (As no confusion can arise, we denote the thermodynamic average of \( r \) by the same symbol: \( r \).) The magnetization of the system is then given by

\[ M = \mu r. \]  

(8)

The magnetic susceptibility is given by

\[ \chi = \mu \frac{\partial r}{\partial H}. \]  

(9)

At low temperatures, and for small values of \( a \), the model is the hard-core Fermi gas. At high temperatures and for arbitrary values of \( a \), this is not necessarily true, since the validity of the energy levels (3) does not extend to these latter cases. However, the general results may still be of interest, since, as we have seen, formula (3) contains the correct qualitative feature that a repulsive interaction tends to align the spins of the particles.

2. THE PARTITION FUNCTION

The partition function for the system is

\[ \exp(-\beta F_{\text{tot}}) = \sum_{n_{s}} \exp\left\{ -\beta\left[ \sum_{s} e_{s} n_{s} - \frac{4\pi a h^{4}}{m V} N_{+} (N_{+} - N_{+}) + (2N_{+} - N_{-}) \mu H \right] \right\}. \]  

(10)

where \( \beta = (kT)^{-1} \), \( k \) being Boltzmann's constant, and \( T \) the absolute temperature, and the partition sum extends over all sets of occupation numbers (\( n_{s} \)) satisfying the condition

\[ \sum_{s} n_{s} = N. \]  

(11)

Let the partition function for an ideal Fermi gas of \( N \) particles be denoted by \( \exp(-\beta F_{\text{tot}}) \). Then we may rewrite (10) in the form

\[ \exp(-\beta F_{\text{tot}}) = \sum_{n_{s}=0}^{N} \exp\left\{ -\beta\left[ f_{s_{0}} + f_{s_{-}} - \frac{4\pi a h^{4}}{m V} N_{+} (N_{+} - N_{+}) + (2N_{+} - N_{-}) \mu H \right] \right\}. \]  

(12)
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It is well known that the logarithm of a sum such as the one above is equal to the logarithm of the largest term in the sum, within an error of the order \(N^{-1} \log N\), which becomes negligibly small as \(N\) increases without limit. Therefore

\[
F_s = f_{s+} + f_{s-} = \frac{4\pi a \hbar^2}{mV} N_+ (N_+ - N_0) + (2N_+ - N) \mu H, \tag{13}
\]

where \(N_+\), appearing in (13), is that value which maximizes \(F_s\). That is, it is the root of

\[
\frac{\partial F_s}{\partial N_+} = 0, \tag{14}
\]

if \(F_s\) possesses a maximum. The last condition may be rewritten

\[
\nu(N_+) - \nu(N_-) = \beta [2\mu H + \frac{8\pi a \hbar^2}{mV} \frac{N_+ - N_-}{2}]. \tag{15}
\]

where

\[
\nu(N) = \beta \frac{\partial f_s}{\partial N} \tag{16}
\]

is the chemical potential of the ideal Fermi gas. Its low and high temperature behaviors are well-known:

\[
\begin{align*}
\nu\left(\frac{N}{2}\right) & \sim \frac{\pi^2}{12} \left(\frac{\hbar T}{\epsilon_0}\right)^2, \\
\nu\left(\frac{N}{2}\right) & \sim \log\left(\frac{N \hbar^2}{2\epsilon_0 V}\right). \tag{17}
\end{align*}
\]

where

\[
\lambda = \left(\frac{2\pi \hbar^2}{mkT}\right)^{1/2}. \tag{19}
\]

From (5) we have

\[
\begin{align*}
N_+ &= \frac{N}{2} (1+r) \\
N_- &= \frac{N}{2} (1-r). \tag{20}
\end{align*}
\]

Hence (15) can also be written

\[
\nu\left(\frac{N}{2} (1+r)\right) - \nu\left(\frac{N}{2} (1-r)\right) = \frac{2\mu H}{kT} + \frac{2a \hbar^2 N}{V} \cdot r. \tag{21}
\]

Solving this equation for \(r\) would yield the magnetic susceptibility \(\chi\), by virtue of (9).

3. SPONTANEOUS MAGNETIZATION

We consider the case of no external field (\(H=0\)). Consider first the low temperature limit \((T \to 0)\). The condition (21) becomes, upon using (17):

\[
(1+r)^{1/2} - (1-r)^{1/2} = \zeta r. \tag{22}
\]
where
\[ \zeta = \frac{8}{3\pi}k_r a. \] (23)

The equation is invariant under a change of sign of \( r \). This is to be expected, for in the absence of field no absolute meaning can be attached to "up" or "down". Thus it is sufficient to consider \( r \geq 0 \). It is seen that \( r = 0 \) is always a solution. By graphical inspection, it is further seen that if \( \frac{4}{3} < \zeta < 2^{2/3} \), then there is an additional solution between 0 and 1. It is this value that maximizes \( F_r \), as we can easily show. When \( \zeta > 2^{2/3} \), (22) gives only one solution namely \( r = 0 \). But going back to (13), one finds that for \( \zeta > 2^{2/3} \) the correct solution is actually \( r = 1 \). The value of \( r \) at absolute zero, as a function of the repulsive strength \( \zeta \) may be summarized as follows:

\[
\begin{align*}
    r &= 0 \quad \text{for} \quad \zeta < 4/3 \quad \text{(no magnetization)} \\
    0 &< r < 1 \quad \text{for} \quad \frac{4}{3} < \zeta < 2^{2/3} \quad \text{(partial spontaneous magnetization)} \\
    r &= 1 \quad \text{for} \quad \zeta > 2^{2/3} \quad \text{(saturated spontaneous magnetization)}
\end{align*}
\] (24)

That is, if the repulsive strength as measured by \( \zeta \) becomes sufficiently strong, the system becomes ferromagnetic. The critical value of at which ferromagnetism first sets in (\( \zeta = 4/3 \)) corresponds to

\[ k_r a = \pi/2. \] (25)

The foregoing results hold at absolute zero. At a finite but small temperature, one has to also take into account the second term of (17), obtaining instead of (22) the following:

\[ (1+r)^{2/3} - (1-r)^{2/3} = \frac{\pi^2}{12} \left( \frac{kT_e}{\epsilon_r} \right)^2 [((1+r)^{1/3} - (1-r)^{1/3}] = \zeta r. \] (26)

Suppose \( \zeta > 4/3 \), so that we have a ferromagnetic system at absolute zero. It can be shown from (25) that the ferromagnetism disappears at a temperature \( T_e \). That is, above \( T_e \) the only solution to (26) is \( r = 0 \). One finds

\[ \frac{kT_e}{\epsilon_r} = \frac{3}{\pi} \left( \zeta - \frac{4}{3} \right)^{1/3} = \frac{2}{\pi \sqrt{3}} \left( \frac{2}{\pi} k_r a - 1 \right)^{1/3}. \] (27)

This result is valid for \( \zeta - \frac{4}{3} \ll 1 \). The temperature \( T_e \) is clearly what is known as the Curie temperature.

Next consider the high temperature limit in the absence of external field. Equation (21) combining with (18) gives

\[ r = \tanh \left[ \frac{4 \pi a h^2}{m} \frac{N}{\sqrt{V k_T}} \right], \] (28)

the only solution of which is \( r = 0 \), as expected.

4. MAGNETIC SUSCEPTIBILITY

In the presence of a weak external magnetic field \( H \), we calculate \( r \) from (21) to first order in \( H \):
MAGNETIC PROPERTIES OF AN IMPERFECT FERMI GAS

\[ r = r_e + \frac{\mu}{\mu} \cdot H, \]  

where \( r_e \) is the root of \( r \) in the absence of field \( (H=0) \) but at arbitrary temperature:

\[ \nu \left( \frac{N}{2} (1+r_e) \right) - \nu \left( \frac{N}{2} (1-r_e) \right) = 4a^2 \frac{N}{V} r_e. \]  

We find the magnetic susceptibility to be

\[ \chi = 2\mu^2 \left( \frac{N a T}{2} \left[ \nu \left( \frac{N}{2} (1+r_e) \right) + \nu \left( \frac{N}{2} (1-r_e) \right) \right] - \frac{8}{3\pi} (k_B a) \right)^{-1}, \]  

where

\[ \nu(N) = \frac{\partial \nu(N)}{\partial N}. \]  

The low and high temperature limits for \( \chi \) are

\[ \frac{\chi}{T} \rightarrow \frac{3\mu^2}{e} \left[ (1+r_e)^{-3} + (1-r_e)^{-3} - \frac{4}{\pi} - \frac{k_B a}{T} \right]^{-1} \]  

\[ \frac{\chi}{T} \rightarrow \mu^2 / k_B T \]  

Hence Curie's constant is

\[ C = \mu^2 / k_B. \]  

Note that \( r_e \) depends on \( k_B a \). It approaches unity when \( k_B a \) exceeds a certain value. Thus it can be seen from (31) that in general \( \chi > 0 \). The system is either ferromagnetic or paramagnetic, never diamagnetic.

Consider now the case of paramagnetism. That is, we require \( r_e = 0 \) for all temperatures. This means that

\[ k_B a < \frac{\pi}{2}. \]  

In this case (31) becomes

\[ \frac{T \chi}{C} = \frac{3}{2} \frac{k_B T}{e r} \left[ f - \frac{2}{\pi} k_B a \right]^{-1} \]  

where

\[ f = \frac{3}{2} \frac{k_B T}{e r} \left( \frac{N}{2} \right) \nu \left( \frac{N}{2} \right). \]  

The function (37) rises linearly at \( T = 0 \), with a slope of

\[ \frac{\partial}{\partial T} \left( \frac{T \chi}{C} \right)_{T=0} = \frac{3}{2} \left( 1 - \frac{2}{\pi} k_B a \right)^{-1}. \]  

It reaches a maximum value, which is greater than unity, at \( k_B T / e r \sim 1 \). Then it approaches unity as \( T \rightarrow \infty \).

If one calculates \( \chi \) for an ideal Fermi gas endowed with the same magnetic moment, one would find the slope

\[ \frac{\partial}{\partial T} \left( \frac{T \chi}{C} \right)_{T=0} = \frac{3}{2} \] (ideal gas).
The imperfect gas has a steeper slope, as (39) shows. This is again a reflection of the enhancement of spin alignment by the repulsive interaction. This result is sometimes described by saying that imperfect gas behaves like an ideal gas with a higher Fermi energy.

5. SPECIFIC HEAT

As a by-product of the calculation, one can evaluate the specific heat $C_v$ of the system, the result is (for $H=0$):

$$\frac{C_v}{N} = -2\left[2kT\frac{\partial r}{\partial T} + kT^2 \frac{\partial^2 r}{\partial T^2}\right] I(r) - 2kT \frac{\partial r}{\partial T} \frac{\partial I(r)}{\partial T} + \frac{32\pi ah}{m} N \frac{1}{V} T \left[\left(\frac{\partial r}{\partial T}\right)^2 + r \frac{\partial^2 r}{\partial T^2}\right]$$

where

$$I(r) = \nu\left(\frac{N}{2} (1+r)\right) - \nu\left(\frac{N}{2} (1-r)\right)$$

In the paramagnetic case $r=0$. One finds

$$C_v = (C_v) \text{ ideal gas}$$

Thus in this model the interpretation that the imperfect gas behave like an ideal gas with a higher Fermi energy cannot be consistently maintained. In liquid $H_2$, one encounters such a situation to which one should perhaps show no surprise.

REFERENCES

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References


5. K. Huang; a reprint of this article can be found in Appendix B.


