Nuclear Magnetic Susceptibility of He^3 Vapor*

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Measurements of relative values of the nuclear magnetic susceptibility of He^3 vapor have been made by a pulsed resonance technique for several values of vapor density at 2 and 3.1°K. The results of these measurements show that deviations from Curie's law are considerably smaller than those expected for a perfect Fermi gas. A theoretical expression is derived for the susceptibility at vapor densities sufficiently low that only binary collisions are important; a modification of the calculation of the second virial coefficient is used to evaluate the effect of direct interactions. The susceptibility is related to the difference between two virial coefficients, that of He^3 and that calculated for a hypothetical He^3 with zero spin. The theoretical results are in good quantitative agreement with the experimental data. They indicate that the perfect-Fermi-gas theory is not applicable to the calculation of the susceptibility of He^3 vapor at low density except at temperatures lower than 0.002°K. Moreover, from the theoretical results it is concluded that deviations from Curie's law in the vapor are expected to be less than about 1% whenever higher order collisions do not contribute significantly to the interaction between He^3 atoms.

I. INTRODUCTION

THE nuclear magnetic susceptibility calculated for a perfect Fermi gas[1,2] characterized by parameters appropriate to He^3 exhibits pronounced degeneracy effects in the range of density and temperature at which He^3 vapor exists. If the perfect-Fermi-gas theory were applicable to He^3 vapor, the deviations from Curie's law would be large enough to be observed readily. It has been suggested by Romer[3] that the theory of a perfect Fermi gas might be valid for calculating the susceptibility of He^3 vapor at low density. The results of measurements made by Romer on the vapor at 3°K have indicated that at this temperature the perfect-Fermi-gas theory is applicable when the vapor density is less than 3×10^{-3} mole cm^{-3} and that deviations from the results of the theory occur when the density is greater than this value. In contrast, the results of measurements made in this laboratory[4] have indicated that at 3°K and at vapor densities less than 3×10^{-3} mole cm^{-3} deviations from Curie's law are much smaller than those calculated from the perfect-gas theory. In both experiments the susceptibility has been determined as a function of density at the temperature considered.

In order to estimate the effects of direct interactions, which would explain deviations from the perfect-gas theory, an approximate theoretical expression for the susceptibility of He^3 has been derived. This approximate expression for the susceptibility includes the effects of interactions between He^3 atoms when the vapor density is sufficiently low that only binary collisions are important. Measurements of the susceptibility of the vapor at 2°K have also been made in order to provide additional data for comparison with the theoretical results.

The following paragraphs comprise a description of our experimental method and results, an outline of the derivation of the theoretical expression for the susceptibility, and a discussion of the experimental and theoretical results. It is shown that our experimental results are in satisfactory agreement with the theoretical results. The theoretical results are used to establish the conditions under which the perfect-Fermi-gas theory gives a good approximation to the deviations from Curie's law. Finally, the results are used to estimate the maximum deviation from Curie's law in the vapor.

II. EXPERIMENTAL DETAILS

Relative values of the nuclear magnetic susceptibility of He^3 vapor were determined in this experiment by measuring the amplitude of the free precession signal obtained by the spin-echo technique of nuclear magnetic resonance.[5,6] Relative values of the ratio of the susceptibility to the Curie susceptibility were then calculated by utilizing the He^3 vapor P-V-T data of Keller.[7] The susceptibility was determined in this manner as a function of the vapor density at constant temperature at two different values of temperature.

The spin-echo technique was employed to obtain the free-precession signal and two echoes associated with a 90°–180°–180° rf pulse sequence. In principle the echo technique was not essential to the measurement; however, it is easily seen that if the resonance apparatus was initially aligned to give a 90°–180°–180° pulse sequence at resonance, then a change from the initial conditions produced by a change of the magnetic field, by a drift of

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[1] E. C. Stoner, Proc. Roy. Soc. (London) **A152**, 672 (1935).
[2] J. McDougall and E. C. Stoner, Phil. Trans. Roy. Soc. London **A237**, 67 (1938).
[3] R. H. Romer, *Proceedings of the Eighth International Conference on Low Temperature Physics* (Butterworths Scientific Publications, Inc., Washington, D. C., 1963), p. 17.
[4] J. E. Opfer, K. Luszczynski, and R. E. Norberg, in *Proceedings of the Ninth International Conference on Low Temperature Physics* (Plenum Press, New York, 1965).
[5] E. L. Hahn, Phys. Rev. **80**, 580 (1950).
[6] A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, London, 1961).
[7] W. E. Keller, Phys. Rev. **98**, 1571 (1955).

the frequency of the rf oscillator, or by detuning of the transmitter circuitry would have resulted in the appearance of additional echoes. Because the magnitude of these echoes was much more sensitive to deviations from the initial condition than was the magnitude of the free precession signal, the magnitude of the echoes was used as a monitor to assure proper stability of the resonance apparatus.

The spin-echo technique was suitable for measurements of the susceptibility of He^3 vapor because with typical experimental conditions the damping of the free-precession signal was caused primarily by the inhomogeneity of the external magnetic field. In the measurements described here, the amplitude of the signal at 20.00 Mc/sec was measured 300 μsec after the pulse. The sample was located in the 3-in. gap of a 12-in. electromagnet whose field homogeneity was such that the free-precession signal persisted longer than 5 msec. Other sources of damping such as spin-spin relaxation, diffusion, and radiation damping were investigated to ascertain that changes in the vapor density did not result in measurable changes in the shape of the precession signal.

To assure that the free-precession signal was proportional to the susceptibility it was necessary that the nuclear magnetism be at its equilibrium value before the measurement. Independent observations of the relaxation of the magnetization demonstrated that a single exponential described the approach to equilibrium. The time constant T_1* varied with vapor density, temperature, and sample container. Measurements of the signal amplitude for determination of the susceptibility were made only after a time greater than $7T_1$* had elapsed since the preceding pulse sequence. Occasionally much longer waiting times were used as an additional verification that equilibrium had been obtained.

The amplitude of the free-precession signal was measured by comparing it with a calibrated reference signal induced a short time later in the sample coil. The calibration of the reference signal was based on the calibration of a set of precision attenuators (Weinschel 64A). As is shown schematically in Fig. 1, the attenuators were inserted between a circuit which matched the reference-signal circuit to the attenuators and a signal generator whose output voltage was maintained constant to within $\pm 0.1\%$. The reference-signal circuit supplied current through a 10-kΩ precision resistor to a single loop inductively coupled to the sample coil. Since the precision attenuators were variable in steps of 0.1 dB, it was possible to compare the resonance signal with a reference signal differing in amplitude by at most 1%. The gain of the receiver system was adjusted so that the amplitude of the two signals at the output of the amplifier was approximately the same for each measurement. This adjustment permitted the use of the calibrated differential comparator (Tektronix Z plug-in unit) with an oscilloscope for comparison of the two signals on an expanded scale. Corrections for the differences between the amplitudes of the two signals and the amplitude of the comparison voltage were taken from photographs of the displayed signals.

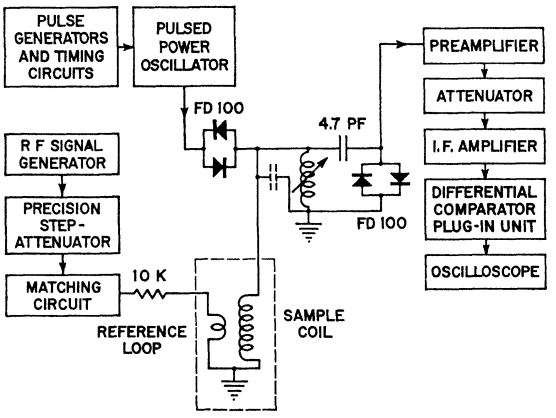

FIG. 1. Arrangement of the spin-echo apparatus and the reference-signal system.

The general arrangement of the spin-echo apparatus is shown schematically in Fig. 1. The relatively simple arrangement of the spin-echo apparatus was made possible by the use of two pairs of crossed diodes (Fairchild FD-100). One pair decoupled the pulsed oscillator from the remainder of the system; the other pair of diodes limited the voltage across the first tube of the preamplifier during the rf pulse.

The sample container used in this experiment consisted of a cylindrical cavity ($\frac{1}{2}$-in. i.d.$\times\frac{1}{2}$-in. high) in a nylon rod sealed to a copper block by a differential contraction seal similar to that described previously.[8] Thermal contact between the block and the He^3 vapor in the cavity was achieved by means of a sapphire rod attached to the block by an epoxy resin-indium joint. The sapphire rod, the bottom end of which terminated in the sample cavity, excluded vapor from all but a small channel above the cavity. A relatively high vacuum was maintained in the region surrounding the sample coil and reference loop. Neither coil was in direct thermal contact with the nylon sample container.

The pressure of the He^3 vapor was measured in this experiment by utilizing a transducer-manometer combination. The pressure of nitrogen gas contained in a ballast cylinder was measured with a mercury manometer; a differential pressure transducer was then used to measure the difference between the nitrogen pressure in the cylinder and the pressure of the He^3 vapor. Sample pressure changes falling within the range of the transducer (± 5 Torr) were measured quickly and accurately because of the small "dead volume" of the transducer (0.1 cm^3).

The sample temperature was determined by means of a carbon resistor used in conjunction with a Wien bridge

[8] K. Luszczynski, R. E. Norberg, and J. E. Opfer, Phys. Rev. 128, 186 (1962).

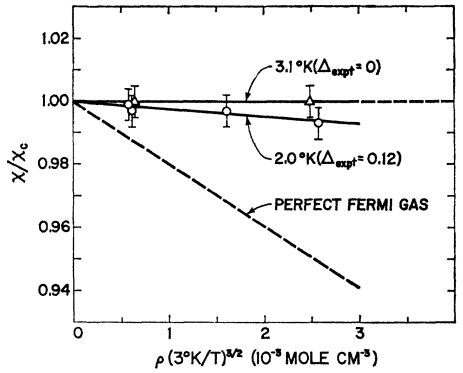

FIG. 2. Ratio of the susceptibility χ of He^3 vapor to the Curie susceptibility χ_C as a function of the quantity $\rho\,(3°K/T)^{3/2}$, where ρ is the density of He^3. The abscissa is chosen so that a single curve represents the corresponding ratio for a perfect Fermi gas. Triangles and circles represent data at 3.1 and 2.0°K, respectively. The data for each temperature are normalized independently to extrapolate to $\chi/\chi_C=1$ at zero density. The quantity Δ_{expt} for each temperature is the ratio of the slope of the straight line drawn through the experimental points to the slope of the curve for the perfect Fermi gas in the limit of zero density.

oscillator.[9] Calibration of the thermometer was accomplished by measuring the pressure of the He^4 bath and checked by measurements of the vapor pressure of liquid He^3 in the sample container (T_{58} He^4 and T_{62} He^3 temperature scales). The He^3 gas used in the experiment was sufficiently pure (<0.15% He^4) to make He^3 vapor pressure thermometry practical. The carbon resistance thermometer was sensitive to temperature changes smaller than 0.001°K although the error in measurement of the absolute value of the temperature was an order of magnitude larger.

III. EXPERIMENTAL RESULTS

Measurements of the susceptibility of He^3 vapor and liquid were reported[4] earlier to present several results. First, it was demonstrated that the susceptibility of the vapor at 3°K did not deviate from Curie's law by more than ±1% at vapor densities in the range ($3.3\times10^{-4}<\rho<3.3\times10^{-3}$ mole cm^{-3}). Second, this result was shown to be reproducible with different experimental conditions involving changes in probe design, changes in sample container size, and changes in the sample and reference coils. Third, it was shown that the same experimental procedure gave reproducible results for the ratio of the liquid susceptibility to the vapor susceptibility at 3°K. It was concluded from this measured ratio and from the behavior of the vapor susceptibility that the susceptibility of liquid He^3 at 3°K was given by Curie's law to within ±1%. This was a reasonable result because earlier measurements by Fairbank *et al.*[10] had demonstrated the liquid susceptibility above 2°K to be proportional to the inverse temperature to within their experimental error. Since no anomalous results were obtained by extending the range of density an order of magnitude to the liquid, the probability of large errors in the measurement technique was reduced. The measurements considered above were made at various stages of the development of the apparatus and the measuring technique. The results to be reported here were obtained from the latest measurements made under the most favorable experimental conditions.

The experimental results are shown in Fig. 2 which is a plot of the ratio of the susceptibility χ of He^3 vapor to the Curie susceptibility χ_C versus $\rho\,(3°K/T)^{3/2}$ where ρ is the density of He^3. This choice of abscissa is made so that a single curve represents χ_{FD}, the susceptibility calculated for a perfect Fermi gas. The two sets of data at 3.1 and 2.0°K are normalized independently in such a way that a straight line drawn through the data points for each temperature passes through the point $(\chi/\chi_C)=1$ at zero density. In effect the only information which is derived from the results is the slope of the straight line drawn through the experimental points. It is convenient to express this slope in terms of the results of the perfect-Fermi-gas theory by defining a parameter Δ_{expt} as the ratio of $[\partial(\chi/\chi_C)/\partial\rho]_{\rho=0}$, determined experimentally, to $[\partial(\chi_{FD}/\chi_C)/\partial\rho]_{\rho=0}$. The parameter Δ_{expt} is defined to facilitate comparison with the theoretical results. The experimental results can now be summarized by stating that the values of Δ_{expt} determined from Fig. 2 are $\Delta_{expt}=0.00\pm0.25$ at 3.1°K and $\Delta_{expt}=0.12\pm0.25$ at 2.0°K. The estimated limit of error in Δ_{expt} is obtained from the combined effects of arbitrary normalization, the ±0.5% error limit estimated for each experimental point, and the limited range of densities over which the data were taken. The major part of this estimated error is due to limits of error assigned to the calibration of the apparatus and cannot be reduced simply by taking more data.

IV. THEORETICAL CONSIDERATIONS

A system of N He^3 atoms is considered to be enclosed in a container of volume V and situated in an external magnetic field H directed along the positive z axis. The static magnetic properties of the system are determined by the number of nuclei in each of the two nuclear spin states. The nuclear magnetization M_z and the susceptibility χ per atom are given by

$$M_z=\tfrac{1}{2}\gamma\hbar(N_+-N_-)/V\,, \tag{1}$$

$$\chi/N=(\tfrac{1}{2}\gamma\hbar/H)(N_+-N_-)/NV\,, \tag{2}$$

where $\frac{1}{2}\hbar$ is the spin angular momentum of He^3 nucleus, $\frac{1}{2}\gamma\hbar$ is the dipolar magnetic moment of He^3 nucleus, N_+ is the number of He^3 nuclei in spin state $m_z=+\frac{1}{2}$, N_- is the number of He^3 nuclei in spin state $m_z=-\frac{1}{2}$, $(N=N_++N_-)$.

An approximate expression for the susceptibility in the limit of low density is derived by minimizing the

[9] B. J. Sandlin and J. C. Thompson, Rev. Sci. Instr. **30**, 659 (1959).

[10] W. M. Fairbank, W. B. Ard, and G. K. Walters, Phys. Rev. **95**, 566 (1954).

free energy of the system with respect to changes of the parameter N_+ at constant N (c.f., calculation by K. Huang[11]). In order to evaluate this expression it is necessary to evaluate the term which depends on the nature of the direct interactions. This term is shown to be related to the difference between the second virial coefficient calculated for He^3 and the second virial coefficient calculated for a hypothetical He^3 with zero spin by employing Bose-Einstein statistics. In this manner a simple relationship is established between the susceptibility and the two virial coefficients. It is convenient to use this relationship because the difference between the two virial coefficients has been calculated by Kilpatrick, Keller, Hammel, and Metropolis[12] (this paper is referred to in the following as KKHM)

The free energy of the system can be written as the sum of three terms in the form

$$F(N,N_+)=F_p(N,N_+)+F_{\rm int}(N,N_+) -\tfrac{1}{2}\gamma\hbar(2N_+-N)H, \quad (3)$$

where $F_p(N,N_+)$ is the free energy of a perfect Fermi gas of N particles with N_+ nuclei in $+\frac{1}{2}$ spin state and N_- nuclei in $-\frac{1}{2}$ state; $F_{\rm int}(N,N_+)$ is the contribution to the free energy arising from the direct interaction between He^3 atoms; and $-\frac{1}{2}\gamma\hbar(2N_+-N)H$ is the interaction energy of the magnetic moment of the system with the external magnetic field. If a minimum value for $F(N,N_+)$ exists for some of N_+ such that $0<N_+<N$, it is required that

$$\partial F(N,N_+)/\partial N_+=0 \quad (4)$$

or

$$\partial F_p(N,N_+)/\partial N_++\partial F_{\rm int}(N,N_+)/\partial N_+-\gamma\hbar H=0. \quad (5)$$

After expressions for $\partial F_p(N,N_+)/\partial N_+$ and $\partial F_{\rm int}(N,N_+)/\partial N_+$ are derived it can be verified that the condition that $\partial^2F(N,N_+)/\partial N_+^2$ must be positive imposes no additional restriction on the range of density for which the final expression for the susceptibility is valid. From the properties of a perfect Fermi gas[1,13] it is seen that the first term in Eq. (5) can be written

$$\partial F_p(N,N_+)/\partial N_+=\mu(N_+)-\mu(N_-), \quad (6)$$

where $\mu(N')$ is the chemical potential calculated for a perfect gas of N' fermions with *no spin degeneracy* of the energy levels. The function $\mu(N')$ is the inverse of the function $N'(\mu)$ which can be obtained from the results of McDougall and Stoner[2] in the form

$$N'(\mu)=2\pi^{-1/2}V\lambda^{-3}F_{1/2}(\eta), \quad (7)$$

where

$$\lambda=(h^2/2\pi mkT)^{1/2},$$

$$\eta=\mu/kT,$$

$$F_{1/2}(\eta)=\int_0^\infty \frac{x^{1/2}dx}{\exp(x-\eta)+1},$$

m is the mass of He^3 atom; other symbols have their usual meaning. If it is assumed that the system is weakly degenerate, then e^η is small and hence $F_{1/2}(\eta)$ can be represented by the approximation

$$F_{1/2}(\eta)\approx\tfrac{1}{2}\pi^{1/2}e^\eta(1-2^{-3/2}e^\eta). \quad (8)$$

An approximation for $\mu(N')$ obtained by utilizing this expression for $F_{1/2}(\eta)$ and assuming that e^η is small is given by

$$\mu(N')\approx kT[\ln(N'\lambda^3/V)+2^{-3/2}\lambda^3N'/V]. \quad (9)$$

If only first-order terms in the density are retained, the approximation for $\mu(N')$ permits rewriting Eq. (6) to give the result

$$\partial F_p(N,N_+)/\partial N_+=kT[\ln(N_+/N_-) +2^{-3/2}\lambda^3(N_+-N_-)/V]. \quad (10)$$

If the density of the gas is now considered to be sufficiently low so that only binary collisions are important, then $F_{\rm int}(N,N_+)$ can be written as

$$F_{\rm int}(N,N_+)=(kTN^2/N_AV)B_{\rm int}(N_+/N). \quad (11)$$

In Eq. (11) $B_{\rm int}(N_+/N)$ is the contribution to the second virial coefficient arising from the direct interaction between atoms; N_A is Avogadro's number which is present in the expression for $F_{\rm int}$ because it is assumed here that the virial expansion is made in terms of the molar volume. The method of calculating $B_{\rm int}(N_+/N)$ parallels the earlier calculations[12,14] of the second virial coefficient of He^3. The results of KKHM can be utilized here to give the following equation for $B_{\rm int}(N_+/N)$:

$$B_{\rm int}(N_+/N)=-2^{-3/2}N_A\lambda^3(16/\pi q_0^2) \times\int_0^\infty(\Sigma)\exp(-q^2/q_0^2)qdq, \quad (12)$$

where

$$(\Sigma)=p_s\textstyle\sum_{l\ \rm odd}(2l+1)\eta_l+p_a\sum_{l\ \rm even}(2l+1)\eta_l,$$

q^2 is the parameter proportional to the relative energy of colliding atoms, q_0^2 the parameter proportional to kT, l the angular-momentum quantum number, η_l the phase shift of the wave function, p_s the probability of a symmetric spin state of nuclear spins of two colliding atoms, and p_a the probability of an antisymmetric spin state. In general p_s and p_a are determined from the equations

$$p_s=(N_+N_++N_+N_-+N_-N_-)/N^2 \quad (13a)$$

[11] K. Huang, *Statistical Mechanics* (John Wiley & Sons, Inc., New York, 1963), p. 282.

[12] J. E. Kilpatrick, W. E. Keller, E. F. Hammel, and N. Metropolis, Phys. Rev. **94**, 1103 (1954).

[13] L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Addison-Wesley Publishing Company, Inc., Reading, Massachusetts, 1958).

[14] J. De Boer, Physica **16**, 545 (1950).

and

$$p_a=N_+N_-/N^2. \tag{13b}$$

In the calculation of the ordinary virial coefficient of He^3, N_+ and N_- are taken to be equal; in this case $p_s=\frac{3}{4}$ and $p_a=\frac{1}{4}$, results for p_s and p_a which correspond, respectively, to $(s+1)/(2s+1)$ and $s/(2s+1)$ for $s=\frac{1}{2}$. The results of the calculation assume a more compact form if three quantities I_0, I_e, and Δ are defined in the following way:

$$I_0\equiv(1/q_0^2)\int_0^\infty[\textstyle\sum_{l\ \mathrm{odd}}(2l+1)\eta_l]\times\exp(-q^2/q_0^2)qdq, \tag{14a}$$

$$I_e\equiv(1/q_0^2)\int_0^\infty[\textstyle\sum_{l\ \mathrm{even}}(2l+1)\eta_l]\times\exp(-q^2/q_0^2)qdq, \tag{14b}$$

$$\Delta\equiv 1-(16/\pi)(I_0-I_e). \tag{14c}$$

In terms of these quantities Eqs. (12) and (11) give for $B_{\mathrm{int}}(N_+/N)$ and $\partial F_{\mathrm{int}}(N,N_+)/\partial N_+$

$$B_{\mathrm{int}}\left(\frac{N_+}{N}\right)=-2^{-5/2}N_A\lambda^3\left(\frac{16}{\pi}\right)\times\left[(I_0+I_e)+(I_0-I_e)\frac{N_+^2+(N-N_+)^2}{N^2}\right] \tag{15}$$

$$\partial F_{\mathrm{int}}(N,N_+)/\partial N_+ = -2^{-3/2}kT\lambda^3V^{-1}(N_+-N_-)(1-\Delta). \tag{16}$$

The results of Eqs. (10) and (16) when combined with Eq. (5) give

$$kT[\ln(N_+/N_-)+2^{-3/2}\lambda^3\Delta(N_+-N_-)/V]-\gamma\hbar H=0. \tag{17}$$

Equation (17) is simplified by neglecting saturation effects which is equivalent to making the approximation

$$\ln(N_+/N_-)\approx 2(N_+-N_-)/N. \tag{18}$$

With the aid of this approximation Eq. (17) can be solved for χ because from Eq. (2) it follows that

$$N_+-N_-=2VH\chi/(\gamma\hbar). \tag{19}$$

The result for χ obtained from Eq. (17) is

$$\chi/\chi_C=1-2^{-5/2}\lambda^3\Delta(N/V)+O[(N/V)^2], \tag{20}$$

where χ_C is the Curie susceptibility given by

$$\chi_C=N(\gamma\hbar)^2/(4kTV). \tag{21}$$

It is seen in the approximation considered here that if $\Delta=0$, the susceptibility obeys Curie's law; the condition for perfect-Fermi-gas behavior is that $\Delta=+1$, a condition which exists when there are no direct interactions.

The preceding results are now to be expressed in terms of the second virial coefficients defined in the following way: $B_{\mathrm{FD}}{}^{1/2}$ is the second virial coefficient calculated for a real gas of He^3 atoms with spin $\frac{1}{2}$ obeying Fermi-Dirac statistics, $B_{\mathrm{BE}}{}^0$ the second virial coefficient calculated for a hypothetical He^3 gas with particle spin zero obeying Bose-Einstein statistics; b the absolute value of $B_{\mathrm{BE}}{}^0$ calculated by assuming that the Bose-Einstein gas is perfect. It can be readily shown[12] that

$$B_{\mathrm{ED}}{}^{1/2}=b[\tfrac{1}{2}-(32/\pi)(\tfrac{3}{4}I_0+\tfrac{1}{4}I_e)], \tag{22a}$$

$$B_{\mathrm{BE}}{}^0=b[-1-(32/\pi)I_e], \tag{22b}$$

where

$$b=2^{-5/2}N_A\lambda^3=109T^{-3/2}\ \mathrm{cm^3\ mole^{-1}}. \tag{23}$$

It follows that the quantity Δ defined by Eq. (14c) can be written in the form

$$\Delta=\tfrac{2}{3}(B_{\mathrm{FD}}{}^{1/2}-B_{\mathrm{BE}}{}^0)/b. \tag{24}$$

The two results given in Eq. (20) and Eq. (24) comprise the anticipated relationship between the susceptibility and the virial coefficients. Equation (24) correlates deviations from Curie's law with the difference between the two virial coefficients $B_{\mathrm{FD}}{}^{1/2}$ and $B_{\mathrm{BE}}{}^0$.

The quantity Δ may be evaluated with the aid of Eq. (24) if calculated values for the difference between the two virial coefficients are available, or it may be evaluated directly from Eqs. (14a)–(14c) by utilizing calculated values of $\eta_l(q)$. Both procedures were employed in determining the values of $\Delta\equiv\Delta_{\mathrm{theo}}$ presented in Table I. Results for $B_{\mathrm{FD}}{}^{1/2}-B_{\mathrm{BE}}{}^0$ were taken from KKHM in which a Lennard-Jones 12–6 potential (LJ1)[12] was assumed for the interaction potential between He^3 atoms. These results for $B_{\mathrm{FD}}{}^{1/2}-B_{\mathrm{BE}}{}^0$ and the values for Δ_{theo} calculated for the LJ1 potential from Eq. (24) are shown in Table I for temperatures of 1, 2, 3, and 4°K. (The phase shifts[15] calculated by KKHM

TABLE I. Experimental and theoretical results[a] for the density dependence of the susceptibility of He^3 vapor as a function of temperature.

T °K	b cm³/mole	$B_{\mathrm{FD}}{}^{1/2}-B_{\mathrm{BE}}{}^0$ cm³/mole (LJ1)	Δ_{theo} (LJ1)	Δ_{theo} (MR5)	Δ_{expt}
0.002	...	...	...	1.2	...
0.01	...	...	...	1.4	...
0.10	...	...	...	1.7	...
1.00	109	107	0.65	0.56	...
2.00	39	12	0.21	0.15	0.12±0.25
3.00	21	2.0	0.06	0.03	...
3.10	...	...	...	0.03	0.00±0.25
4.00	14	0.4	0.02	...	...

[a] The results are expressed in terms of the results of the perfect-Fermi-gas theory by giving values of Δ_{theo} and Δ_{expt}; the parameters Δ_{theo} and Δ_{expt} are, respectively, the theoretically and experimentally determined ratios of $[\partial(\chi/\chi_C)/\partial\rho]_{\rho=0}$ to $[\partial(\chi_{FD}/\partial\chi_C)/\partial\rho]_{\rho=0}$ where χ is the susceptibility of He^3 vapor, χ_C is the Curie susceptibility, χ_{FD} is the susceptibility of a perfect Fermi gas, and ρ is the vapor density. The relationship between Δ_{theo} and the virial coefficients is discussed in the text.

[15] Document No. 5064, ADI Auxiliary Publications Project Photoduplications Service, Library of Congress, Washington 25, D. C. A description of the contents of the document is contained in a paper by Keller: W. E. Keller, Phys. Rev. **105**, 41 (1957). The document contains the quantity $\eta_l(q)$ calculated for the LJ1 potential as well as for the MR5 potential.

were used to determine that the value of $B_{FD}^{1/2}-B_{BE}^{0}$ at 2°K should be 12 cm³/mole instead of the published value of 22.) In a later calculation of the second virial coefficient of He^3 Kilpatrick *et al.*[16] assumed an exponential-six potential (MR5) for the interaction potential. Values for Δ_{theo} were calculated for this potential (MR5) by integrating numerically the expressions (14a) and (14b) using the quantities $\eta_l(q)$ which were available[15] from the calculations of Kilpatrick *et al.* These values of Δ_{theo} calculated for the MR5 potential for several temperatures in the range $0.002°K<T<3.1°K$ are also given in Table I.

V. DISCUSSION OF RESULTS

In the preceding section a theoretical expression has been obtained for the susceptibility of He^3 vapor at low density. Before these results are compared with the experimental results it is necessary to justify the assumption that only the first two terms for χ/χ_C in Eq. (20) are important in the range of density for which measurements were made. As is indicated in Fig. 2 the relationship between χ_{FD}/χ_C and ρ is nearly linear in this range; consequently it is assured that the density is sufficiently low to permit the approximation [Eq. (10)] made for the contribution arising from the free energy of the perfect gas. The assumption that only binary collisions are important cannot be justified rigorously because the theoretical calculation does not give an estimate of the magnitude of higher order terms. Instead of a more rigorous justification it can be stated that the term involving the third virial coefficient is small compared to the term involving the second virial coefficient in the inverse volume expansion of the equation of state. The third-order term is less than 5% of the second-order term at 3.1°K and less than 2% of the second-order term at 2°K for densities at which measurements were made. It is now seen that the assumption of a linear relation between the experimentally determined ratio χ/χ_C and the density was made because an approximately linear relation is expected from the theoretical results.

The theoretical result given by Eq. (20) can be expressed in a different form in the following way:

$$\Delta_{theo}\equiv\Delta=[(\partial(\chi/\chi_C)/\partial(N/V))/(\partial(\chi_{FD}/\chi_C)/\partial(N/V))]_{(N/V)=0}, \quad (25)$$

where χ_{FD} is the susceptibility calculated for a perfect Fermi gas (i.e., $\Delta=1$). This expression for Δ_{theo} is analogous to that given in Sec. III for Δ_{expt} and therefore permits the comparison of the two quantities. It is evident from Table I that there is good agreement between the experimental and theoretical results for the two temperatures at which measurements were made. At both 2.0 and 3.1°K, Δ is small compared to the perfect-Fermi-gas value of unity. It should be noted that the second virial coefficients calculated for He^3 by assuming the MR5 potential are in better agreement with the measured[7] virial coefficients than are those calculated by assuming the LJ1 potential. It is therefore expected that Δ_{theo} calculated for the MR5 potential should be in better agreement with Δ_{expt} than is the same quantity calculated for the LJ1 potential. The results shown in Table I are in agreement with this expectation but cannot be taken too seriously because of the error limits for Δ_{expt}. The situation can be described in another way by stating that the calculated values of χ/χ_C obtained for either potential differ from the experimentally determined values by less than the $\pm0.5\%$ experimental error.

The preceding results have indicated that the perfect-Fermi-gas theory ($\Delta=1$) does not describe the behavior of the susceptibility of He^3 vapor at 2 and 3.1°K. In an attempt to determine the conditions required for the applicability of this theory, Δ_{theo} for the MR5 potential was calculated for the temperature range $0°K<T<3.1°K$. The general features of the behavior of Δ_{theo} as a function of temperature can be inferred from Table I. The condition that $\Delta=1$ is satisfied for some temperature between 0.1 and 1°K; however, it is obvious that this temperature has no significance as a condition for the general applicability of the perfect-gas theory. The quantity Δ_{theo} has a maximum value at $T\approx0.1°K$ and then monotonically decreases to $\Delta_{theo}=1$ as the temperature decreases to 0°K. Again from Table I it is seen that temperatures less than 0.002°K are required to assure that Δ_{theo} differs from unity by less than 20%. Therefore in general the difference between the susceptibility of He^3 vapor and the Curie susceptibility is given by the perfect-gas theory to within $\pm20\%$ of the difference only at temperatures less than 0.002°K. The values of Δ_{theo} can be used to calculate the fractional difference between the susceptibility of He^3 vapor at the saturated vapor pressure and the Curie susceptibility as a function of temperature. It can be demonstrated that this fractional difference has a relative maximum of about 1% at a temperature near 1.2°K. Therefore, it is expected that the susceptibility of the vapor differs from the Curie susceptibility by no more than 1% at any temperature below about 2°K or at temperatures above 2°K whenever the density is sufficiently low that only binary collisions are important.

VI. CONCLUSIONS

The approximate theoretical expression derived for the susceptibility of He^3 vapor at low densities gives an adequate explanation of the results of susceptibility measurements made in this laboratory at 2.0 and 3.1°K. Both the experimental and theoretical results demonstrate that deviations from Curie's law are much smaller than expected from the perfect-Fermi-gas theory at these temperatures. The observed deviations are too small compared to the experimental error to constitute

[16] J. E. Kilpatrick, W. E. Keller, and E. F. Hammel, Phys. Rev. 97, 9 (1955).

unambiguous evidence of departure from Curie's law. The theoretical results show that deviations from Curie's law are proportional to the difference between the virial coefficient calculated for He^3 and the virial coefficient calculated for a hypothetical He^3 with zero spin; therefore, the relatively small observed deviations from Curie's law are experimental evidence of a relatively small difference between the two calculated virial coefficients. For both quantities the standard of comparison is the corresponding quantity calculated for a perfect gas. From the theoretical results it is concluded that much lower temperatures ($T<0.002$°K) are required to assure that the perfect-Fermi-gas theory gives a good approximation to the deviations from Curie's law. Moreover, because of the limited range of vapor density any deviations from Curie's law are expected to be less than 1% in He^3 vapor at any temperature and density whenever only binary collisions need to be considered.

The theoretical results presented in this paper are in agreement with the results of the previous measurements[4] made at 3°K in this laboratory. The discrepancy between Romer's results[3] and our results remains unexplained.

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Angular-Momentum Experiments with Liquid Helium*

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Angular-momentum measurements on helium I and helium II contained in a cylindrical vessel suspended from a virtually frictionless magnetic bearing are reported. Helium I behaves as a classical viscous fluid but the results for helium II are quite different. The latter shows two distinct types of behavior, either assuming an equilibrium state of solid-body-type rotation or else forming a metastable state wherein the liquid refuses to be excited into rotation. Both types of behavior are shown to be in accordance with predictions of the Onsager-Feynman hypothesis of quantized circulation in the superfluid. Data are presented which seem to imply that macroscopic turbulence already present in the superfluid is a necessary condition for the generation of quantized vortex lines in this type of geometry.

INTRODUCTION

LIQUID helium, according to current theory, consists of a superfluid phase of zero viscosity and entropy plus excited states (phonons and rotons) at all temperatures greater than 0°K and less than about 2.19°K. These quantum excitations possess both entropy and viscosity and are called the "normal" fluid component. Formally their density can be represented by a number ρ_n and, if the ordinary density of the liquid be ρ, the density of the unexcited (superfluid) states is $\rho_s=\rho-\rho_n$. The ratio ρ_n/ρ is a function of the temperature in the above range.

The ρ_s component, in addition to having zero viscosity and entropy, is thought to satisfy another criterion, namely, $\nabla\times\mathbf{v}_s=0$, where $\mathbf{v}_s$ is the flow velocity of the ρ_s fraction. This condition implies some interesting consequences. For a cylindrical geometry, the only solutions with cylindrical symmetry are

$$v_s=0 \quad \text{and} \quad v_s=A/r,$$

where A is a constant and r is the radial distance in the cylindrical vessel. In a simply connected geometry the only possibility for the second type of velocity field is a vortex with a hollow core in the region $r=0$. If pure superfluid were placed in a cylindrical vessel (bucket) and rotated about its vertical axis of symmetry, then either the superfluid would not be excited into rotation or it would form a hollow-core vortex. The free energy of the vortex mode, however, is rather high, so the first alternative would appear to be the preferred one.

The experiment when tried some years ago[1–3] failed to confirm this expectation. The superfluid either rotated as a solid body or, less commonly, produced a hollow-core vortex.

The reasons for this behavior became clearer when Onsager[4] and, independently, Feynman[5] showed that circulation in the superfluid should be quantized in units of $h/m\cong10^{-3}$ cm²/sec. (h=Planck's constant,

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