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Pulsed Nuclear-Resonance Investigation of the Susceptibility and Magnetic Interaction in Degenerate He^3 Films*

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We report measurements of the nuclear magnetic susceptibility of absorbed He^3 at temperatures down to 50 mK using pulsed nuclear resonance. In broad agreement with previous measurements at higher temperatures, it exhibits smaller antiferromagnetic deviations from Curie's law than bulk liquid. The deviations are too large to be explained by a statistical layer model which is adequate at higher temperatures, indicating that the adsorbate, or part of it, is more nearly ferromagnetic than the model predicts.

The He^3 film is an extremely convenient system for the investigation of nuclear magnetic properties of assemblies of finite geometry. Continuous-wave nuclear-resonance measurements have been made recently of the susceptibility and spin-lattice relaxation time of such films[1] (of pure He^3 and He^3-He^4 mixtures) in the temperature range between 1.5 K and 0.4 K, for various film thicknesses. In the region investigated, the films either were classical, in the sense that their susceptibility followed a Curie law, or showed only small (up to ~26%) Fermi degeneracy, depending on coverage and temperature. It was shown that the susceptibility was consistent with a statistical layer model in which the first two layers could be considered as magnetically independent of each other and of the rest of the film, and each layer region of the film exhibited the same susceptibility as that of bulk He^3 at the same interatomic spacing. Such an interpretation is not necessarily unique, and it is expected to be an approximation. In the present report, we show that there are large deviations from this model at lower temperatures down to 50 mK, in such a direction as to indicate a lower Fermi temperature in the real film than in the model, or a larger ferromagnetically inclined exchange interaction.

The adsorbed phase is formed on a substrate of Vycor porous glass (Corning 7390), as in numerous other experiments in this laboratory.[2,3] The original form of the Vycor was not the same as that in the previous work, being a sheet instead of a rod, although the disks were cut in the same circular shape about 1.2 mm thick and 7 mm in diameter. Adsorption isotherms showed that their adsorption-geometric characteristics were very similar, with an effective average pore radius of 35.6 Å, and a total void volume of 0.142 cm^3. A schematic diagram of the low-temperature part of the apparatus is shown elsewhere.[4] Ten of the disks, with total surface area 79.7 m^2, were contained in an epoxy resin cell (Epibond 100A) with a minimal dead volume, and measurements were carried out with the chamber full of liquid. The adsorbed He^3 inside the Vycor was therefore always in direct contact with the small quantity of bulk liquid He^3 in which it was immersed. In turn, this liquid was thermally connected by means of a column of liquid, 0.318 cm in diameter and about 4.5 cm long, to a brush of about 5000 copper wires which were embedded in the epoxy and passed into the mixing chamber of a He^3 dilution refrigerator. The sample was also thermally connected by means of a similar liquid column 2 cm long to a smaller chamber situated below.

Mixing chamber temperatures could be measured with a powdered cerium-magnesium-nitrate (CMN) pill immersed in the fluid, using a mutual-inductance bridge method. Speer carbon-resistance thermometers (220 Ω, $\frac{1}{2}$ W) were placed in the mixing chamber and in the sample He^3 above and below the Vycor chamber. In addition, we could measure the spin diffusion coefficient D of the bulk liquid He^3 surrounding the Vycor disks and also of the bulk liquid in the small chamber below the experimental cell.[4] These diffusion coefficient measurements could be used as an additional thermometric parameter by comparing D with recent measurements.[5] A CMN thermometer could not be incorporated into the sample chamber for fear of contaminating the Vycor substrate with adsorbed water. Temperatures quoted below are probably accurate to within about 3 mK at 50 mK.

The NMR receiver coil was embedded in the epoxy and surrounded the Vycor disks. The transmitter coil was a saddle Helmholtz pair located around the glass vacuum jacket, and the experiments were performed in a field of about 500 G (resonance frequency ~1.6 MHz) provided by a copper-wound liquid-nitrogen–cooled sixth-order solenoid located in the nitrogen bath. This magnet provided a field homogeneity of $3{:}10^5$ over the sample volume of about 0.5 cm^3 even without careful adjustment of the compensating coils, and was satisfactory.

The susceptibility χ of the adsorbed He^3 was obtained by measuring the amplitude $A(t)$ of the free induction tail following a single 90° pulse. This amplitude is given by

$$A(t) = A(\Delta t)\exp(-t/T_2), \qquad (1)$$

where T_2 is a measure of the total spin dephasing time which is determined in our case largely by the field inhomogeneity, and Δt is the sum of the receiver recovery time and the length of the 90° pulse itself. If the pulse is narrow enough for the dephasing of the nuclear moments during the time Δt to be negligible, then $A(\Delta t) = A(0) \propto M_0 = \chi H_0$, where M_0 is the magnetic moment in the steady field H_0. In our experiments the width of the 90° pulse was typically 50 μsec and the receiver recovery time was about 15 μsec, whereas T_2 was about 3 msec; consequently, a simple extrapolation to the zero of time to obtain $A(0)$ could be made with negligible error. As usual, the circuit parameters are not sufficiently accurately known for absolute evaluation of χ, and a normalization procedure must be used. In our case, we have normalized our results to the Curie value in the region 1.5–2.0 K, where we find $\chi T = C$, a constant, and where previous measurements[1,3] in this laboratory of χ as a function of coverage and temperature give reason to suppose that the susceptibility has its free-spin value.

Our values of $\chi T/C$ versus T are shown in Fig. 1 together with the most recent bulk-liquid measurements[6] at the saturated vapor pressure. These results were obtained from four experimental runs, and the scatter indicates that the reproducibility is of the same order as the precision. It is clear from the linear plot of Fig. 1 that in this system Curie's law is followed down to an appreciably lower temperature than in bulk liquid He^3 and that the total susceptibility is always significantly higher. Furthermore, the curve through the experimental points does not extrapolate readily to the absolute zero.

We now attempt to interpret these results in terms of the statistical layer model already mentioned. The assumptions of this model are that (i) the first layer has an interparticle spacing corresponding to that of solid He^3 at a molar volume of 15.5 cm^3, (ii) the second layer corresponds to bulk liquid at a molar volume of about 26 cm^3, (iii) higher layers have the density of bulk liquid at the saturated vapor pressure, (iv) the first two layers behave independently of each other and of the rest of the film, and these three regions have the properties of the bulk phase with the same interparticle spacing. Although crude, the model is useful for the interpretation of a number of properties[3] of helium, and in particular accounts for the susceptibility[1] down to 0.4 K. In the present case, we write, for a film containing n_t atoms having an average magnetic moment M per atom,

$$n_t M = n_1 M_1 + n_2 M_2 + \sum_{i>2} n_i M_i(l), \qquad (2)$$

where i labels the layer with magnetic moment M_i per atom containing n_i atoms, and $M(l)$ is the magnetic moment per atom of bulk liquid at the saturated vapor pressure. At temperatures below 0.4 K our results show an increasing divergence from the predictions of this model, the observed susceptibility being considerably higher than the predictions. (The divergence occurs only below 0.15 K if the second layer is taken to have Curie behavior.) This is shown most clear-

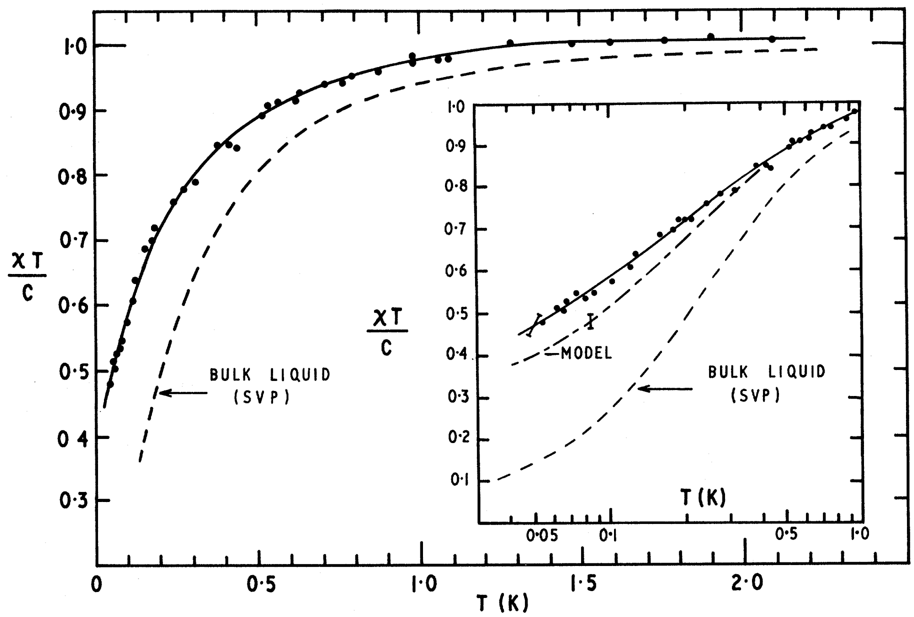

FIG. 1. Main diagram, linear plot of $\chi T/C$ (see text) versus T; the solid line through the points represents the experimental results, the dashed curve is for bulk liquid (Ref. 6). Note that the origin of coordinates is the point (0, 0.2). Inset, the same data plotted semilogarithmically to bring out the low-temperature part. The lower dashed curve is for bulk liquid; the upper dashed curve represents the prediction of the statistical layer model; the points are the experimental results.

ly in the inset to Fig. 1 in which the data are plotted logarithmically to bring out the low-temperature region where the deviations are greatest. The quantities n_i in Eq. (2) were obtained from the absorption characteristics of the Vycor glass as deduced from the usual adsorption isotherms. The error bars shown in Fig. 1 (inset) indicate the uncertainties associated with these measurements.

Although the statistical layer model as outlined above is not successful, it is instructive to retain the basic idea of noninteracting layers, which is borne out in the higher-temperature region, and in place of assumption (ii), to calculate the second-layer susceptibility necessary to give agreement. This layer might be expected, in the model, to behave like a two-dimensional Fermi system with an unknown degeneracy temperature; such behavior has recently been observed in the specific heat of submonolayer films,[7,8] in the surface tension[9] of ^{3}He adsorbed on the surface of ^{4}He, and in the susceptibility of a ^{3}He-^{4}He film.[10] The result of this calculation is shown in Fig. 2, together with the measured susceptibility of bulk liquid ^{3}He at the same interatomic spacing,[6] with calculated values of the Curie-law susceptibility, and with the susceptibility of a two-dimensional Fermi gas of appropriate surface number density. The last of these was calculated from the equation

$$\frac{\chi}{(\chi T)_{T\to\infty}}=\frac{\chi}{C}=\frac{1}{T_D^{**}}\left[1-\exp\left(-\frac{T_D^{**}}{T}\right)\right], \qquad (3)$$

where T_D^{**}, the two-dimensional magnetic degeneracy temperature, replaces T_D, the corresponding ideal-gas degeneracy temperature.[11] A reasonable fit of this equation is obtained for $T_D^{**}=0.05$ K, whereas the bulk liquid and the Curie-law values differ by a factor of 3 or more at 50 mK. A true test of this idea can only be made at much lower temperatures than are accessible in our apparatus ($T \ll T_D^{**}$), where χ should level off to a constant value.

An alternative interpretation in terms of the layer model, more difficult to test, can be made by assuming that part of the second layer is localized and obeys Curie's law, while the rest behaves like bulk liquid. However, it is doubtful whether such analyses can be profitably pursued further at present, and there is a clear need for a more sophisticated theoretical approach. In this connection, it is worth pointing out that the possibility of treating adsorbed ^{3}He as a uniform magnetic system is further discounted by these experiments: It is impossible to find a unique value of the magnetic degeneracy temperature, T_F^{**}, that will fit the universal curve of Gold-

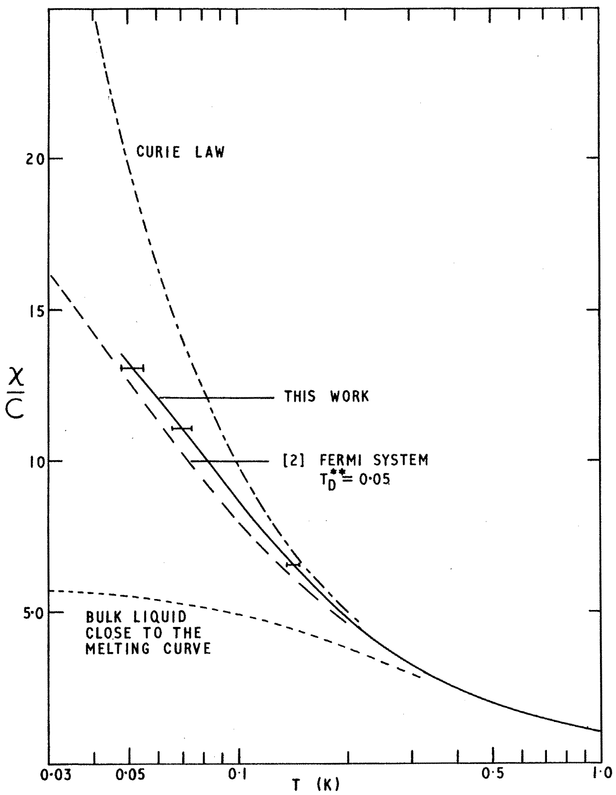

FIG. 2. Plot of χ/C versus T for the second layer (see text), showing large deviations from bulk liquid and from Curie behavior at the lower temperatures, and approximate agreement with a two-dimensional Fermi gas.

stein's theory[11] to our results, where $\chi T/C$ was plotted against the reduced temperature T/T_F^{**}. This result is not surprising because its success depends on a uniform effective mass throughout the film, which in turn presumably depends on a uniform density which is not the case for these adsorbed films. Similarly, the anomalies observed by Hickernell, McLean, and Vilches[8] in submonolayer ^{3}He films adsorbed on graphite, which disappeared when monolayer coverages were approached in this temperature region, do not seem relevant to the interpretation of our results.

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[11]Application of Eq. (3) was suggested by L. Goldstein's phenomenological approach for the bulk liquid [Phys. Rev. 133, A52 (1964)], where a similar expression is quite successful. In the present case, the equation is exact because the two-dimensional Fermi integrals can be solved analytically.