Nuclear Ferromagnetism of Two-Dimensional ^{3}He

H. Franco,[1] R. E. Rapp,[1,a] and H. Godfrin[1,2]

[1] *Centre de Recherches sur les Très Basses Températures, Centre National de la Recherche Scientifique, 38042 Grenoble Cedex, France*

[2] *Institut Laue-Langevin, 38042 Grenoble Cedex, France*

(Received 2 June 1986)

The magnetization of ^{3}He adsorbed on Grafoil has been measured as a function of coverage (two to three layers) at millikelvin temperatures. The observation of a well-defined peak substantially above the free-spin (Curie) value proves unambiguously the existence of a surface ferromagnetic effect.

PACS numbers: 67.70.+n, 67.50.−b, 67.65.+z, 67.80.Jd

Surface magnetic phenomena have been observed by several groups in liquid ^{3}He in confined geometries.[1] For different systems the magnetization has a ferromagnetic Curie-Weiss contribution with a characteristic temperature $\theta \sim 0.5$ mK which has its origin in the first adsorbed layers. In these experiments, cooling of the substrate was achieved indirectly, by the liquid ^{3}He in contact with a heat exchanger. Therefore, the effect of the adsorbed layers could not be separated from that of the liquid. Moreover, heterogeneous substrates were often used, making the data analysis ambiguous.

We have developed the techniques necessary to cool down to millikelvin temperatures a well characterized and homogeneous substrate (Grafoil[2]) in order to perform high-sensitivity NMR measurements on monolayers of adsorbed ^{3}He. In a recent Rapid Communication[3] we reported the measurement of a Curie-Weiss constant of 1.8 mK (larger than that observed in confined 3D liquid) at a coverage of 43.9-cm^3 STP (of the order of 2.6 layers). Accurate data in a narrow coverage range were therefore desirable.

We report in this Letter new results obtained with the experimental cell described in Ref. 3. A detailed account of the novel techniques we have developed to obtain good thermal contact between copper and Grafoil and to minimize the heat leaks will be given in a technical publication. The present measurements were made with a new cw NMR spectrometer using lock-in detection. The NMR frequency was 886.0 kHz, the corresponding value of the magnetic field 27.3 mT, and the width of the magnetic field sweep 0.5 mT. The ^{3}He absorption line was integrated numerically by a microcomputer. It was found convenient to calibrate the amplitude of the measured absorption by comparison with that caused by a calibrated attenuator (0.2%). This reference signal was generated immediately after each field sweep. In this way the NMR absorption was measured independently of the gain of the amplifier.

We have checked that the usual method (keeping track of the gain during the measurements) and the present procedure agree within 2%. The area of the normalized absorption signal is proportional to the magnetization, and is expressed here in arbitrary units which are the same throughout the experiment. Magnetization measurements as a function of temperature were performed in the temperature range 3–50 mK. The temperature was given by carbon resistors and a cerium magnesium nitrate mutual inductance calibrated at zero field and at 27.3 mT. The measurements were performed at the following coverages: 22.05, 36.67, 37.95, 39.22, 40.49, 41.76, 43.04, 44.31, 45.58, 46.85, 48.11, 49.38, 50.65, 51.92, 54.48, and 57.03 cm^3 STP.

From neutron measurements,[4] the specific coverage at monolayer completion is 0.108 atoms/Å^2. The monolayer coverage was found to be unchanged from our previous experiment: 22.0 cm^3 STP. These values allow us to calculate specific coverages (n) for the adsorbed volumes (V): $n = (4.9\times10^{-3}$ Å^{-2} cm$^{-3})\,V$ (STP). We first investigated the coverage corresponding to the monolayer, i.e., to a dense 2D solid[4] with exchange frequencies $hf_e \ll k_B T$ even at 3 mK.[5] A Curie law is expected and observed at this coverage. This measurement provides a calibration for the magnetization scale. The coverage range is centered around the value of 43.9 cm^3 STP where the preliminary experiment[3] detected large ferromagnetic effects. Figure 1 shows the evolution of the magnetization at 3, 5, 10, and 20 mK as a function of coverage. The dashed line corresponds to Curie behavior for all spins, scaled to fit the results at monolayer coverage. The most striking feature is that the magnetization is above the Curie value for a large range of coverages: The value at 43.9 cm^3 STP agrees with our preliminary result.[3] The analysis of the experimental results requires a discussion of the structure of the adsorbed layers. According to van Sciver and Vilches,[6] completion and solidification of the second layer should occur at $n = 0.186$ Å^{-2}. However, neutron experiments[7] performed at $n = 0.203$ Å^{-2} did not detect the

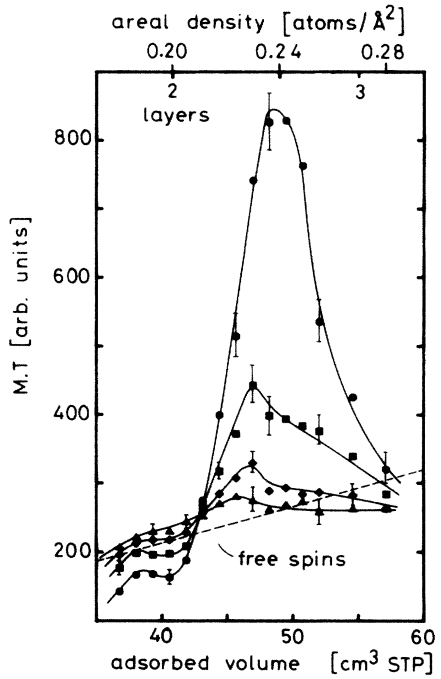

FIG. 1. The product of magnetization and temperature vs coverage for (circles) 3 mK, (squares) 5 mK, (lozenges) 10 mK, and (triangles) 20 mK. Solid lines are guides to the eye. Dashed line is the free-spin behavior.

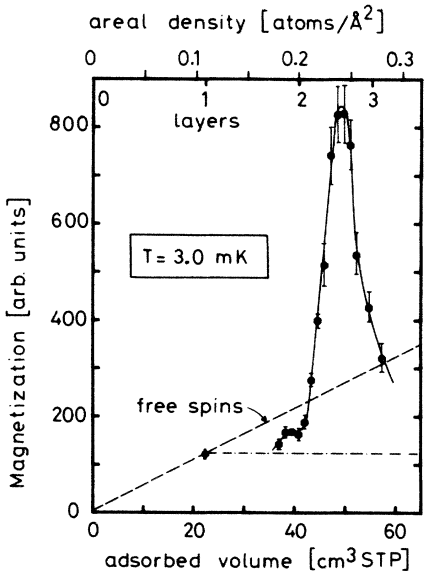

FIG. 2. Magnetization at 3.0 mK vs coverage. Dashed line, free-spin behavior; dot-dashed line, first-layer contribution.

presence of solid in the second layer. In the case of ^{4}He,[4] the density of the compressed first layer is 0.117 Å^{-2} (0.111 Å^{-2} for ^{3}He) and the total density for compressed first and second layers is 0.210 Å^{-2}; scaling to the ^{4}He data suggests that for ^{3}He the corresponding value is about 0.20 Å^{-2}. A rough estimation of the density of the third layer based on the density of bulk liquid ^{3}He gives 0.07 Å^{-2}; third-layer completion should occur at a total density of the order of 0.27 Å^{-2}. Therefore, the coverages investigated in this work are in the range of two to three adsorbed layers (see coverage scale on the figures).

Our data at low temperatures (Fig. 2) show that a small peak in the magnetization occurs precisely at the coverage of 0.186 Å^{-2} where specific-heat measurements suggested a possible second-layer completion and solidification.[6] However, the magnetization for this coverage is substantially lower than the Curie value. The same behavior is followed for neighboring coverages. After subtraction of the first-layer contribution, the magnetization has a Fermi-liquid–like behavior similar to that reported[8] at submonolayer coverages. In this case, however, the degeneracy temperature is very low, of the order of 10 mK. No substantial difference is seen for coverages below 0.186 Å^{-2} where the second layer is certainly liquid, and above 0.186 Å^{-2}, where solidification was reported.[6] We believe that this coverage corresponds to the beginning of third-layer promotion, but not necessarily to solidification. The small peak would indicate an increase in the liquid mobility as atoms are added in the third layer. At higher temperatures (Fig. 1) the magnetization for this coverage region follows the Curie law corresponding to all the ^{3}He spins.

The largest effects arise for coverages between 0.2 and 0.27 Å^{-2}. According to the preceding discussion, this corresponds to a situation where 2 to 3 adsorbed layers are present. At low temperatures the magnetization varies linearly with coverage, becomes substantially larger than the Curie value for all the ^{3}He spins in the system (note that this was not verified in the experiments[1] with confined liquid ^{3}He), and then decreases. The linear (low coverage) side of the peak is characterized by a magnetization which diverges at about 2.8 mK (at a coverage of 0.24 Å^{-2} we observe a magnetization larger than 6 times the Curie-law value, at 2.9 mK, the minimum temperature reached by the dilution refrigerator) but is not well described by a Curie-Weiss law. On the other hand, the high-coverage side of the peak is characterized by a magnetization which is well described by a Curie-Weiss law (after subtraction of the first-layer contribution). The Curie-Weiss temperature θ which is of the order of 2.8 mK at the peak decreases towards 0.5 mK as the coverage increases. This agrees with the value observed[1] for the surface Curie-Weiss contribution in liquid ^{3}He in confined geometries, which is then a surface effect attenuated in the thick-film limit. These results are

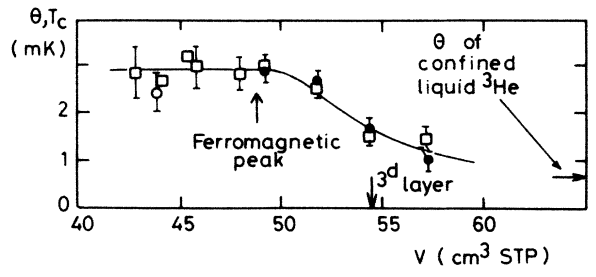

FIG. 3. The Curie-Weiss temperature θ (solid circles), and the temperature T_c at which the magnetization diverges (squares) as functions of coverage. (T_c is defined as the Curie-Weiss temperature determined only with data between 2.8 and 5 mK.) Open circle is the Curie-Weiss temperature θ from Ref. 3. Note that the first-layer contribution is subtracted in the present analysis.

shown in Fig. 3.

The measurements at 20 mK (Fig. 1) show that the magnetization at high coverages does not follow the dashed line (Curie behavior for all atoms): It saturates with increasing coverage at a value which corresponds to about two layers of paramagnetic atoms. The ferromagnetic effect arises during third-layer formation and there is no reason to believe that the magnetization of the first layer could be affected in this coverage range. We suppose therefore that the contribution of the first layer is paramagnetic and focus discussion on the behavior of the second layer. An obvious explanation is to suppose that the second layer is solid at high coverages. However, a Curie-Weiss contribution to the magnetic susceptibility of adsorbed liquid ^{3}He has been theoretically predicted.[9] We are thus led to two different interpretations of the data until further experiments (neutron scattering) provide information on the nature of the second layer.

If the second layer of ^{3}He on Grafoil does not solidify, our data necessarily imply that adsorbed liquid ^{3}He is strongly polarized: Its magnetization diverges at a temperature which is coverage dependent and has a maximum as a function of coverage for a constant temperature with a characteristic thickness of the order of 1 interatomic distance. The paramagnon theory of liquid ^{3}He predicts that an attracting potential well gives rise to magnetization oscillations as a function of distance with a characteristic length of 1 interatomic distance. The oscillations are attenuated within distances of the same order of magnitude and therefore a magnetization peak as a function of distance appears as the main feature of the theoretical curves.[9] Even though the calculations have not been carried out in the case of a finite thickness, but for liquid near a wall, the effects we observe can be considered as a natural consequence of the model.[9]

Another interpretation can be given to the data if we suppose that the second layer solidifies. The linear increase of the low-coverage side of the ferromagnetic peak suggests a coexistence of two phases: a 2D solid of low density characterized by a large ferromagnetic tendency ($\theta \sim 2.8$ mK) and a liquid with a small magnetization below 10 mK. As the coverage is increased, the proportion of solid increases; at the peak coverage, the second layer is completely solidified. A further increase in coverage causes a compression of the solid second layer and a corresponding reduction of the exchange and of the Curie-Weiss temperature. Increasing the coverage beyond three layers does not affect the second-layer density[4]; this explains why surface effects are still observable in liquid ^{3}He in confined geometries. Two ferromagnetic exchange mechanisms can be considered within this interpretation. In the case of a 2D triangular lattice three-spin exchange has been predicted[10] to be dominant, leading to a ferromagnetic interaction with an order of magnitude of 1 mK, in agreement with our experimental results: The reduction of the Curie-Weiss temperature at high coverages is in fair agreement with the theoretical prediction. An indirect exchange of two particles in a solid layer via a third particle in the liquid has been proposed by Jichu and Kuroda.[11] The increase of the number of atoms in the third layer will also give a linear variation of the magnetization on the low-coverage side of the ferromagnetic peak. The high-coverage side of the peak is explained by the compression of the second layer (reduction of the localization length β in the model) and by the reduction of the interlayer exchange due to the increase of the kinetic energy of the atoms in the third layer as the coverage increases, a motional narrowing effect described by Mullin and Landesmann.[12] The range of this exchange interaction is restricted in practice to 1 atomic distance. Therefore, it is a particular kind of cyclic three-particle exchange and this explains its ferromagnetic nature.

An experiment on ^{3}He films has been analyzed recently[13] in terms of surface superfluidity with high critical temperature.[14] The interpretation of our data as an effect in the liquid suggests that this extraordinary superfluidity could be due to the high polarization of the liquid surface layers, equivalent to the effect of a high magnetic field.

In a recent study of ^{3}He adsorbed on sintered silver,[15] a ferromagnetic tendency has been detected but no ferromagnetic peak is observed. The effects are shifted to higher coverages, in a way which does not seem to be simply related to the heterogeneous nature of this substrate.

The latest results[16] on the magnetization of liquid ^{3}He confined in Grafoil suggest the occurrence of a surface ferromagnetic transition below 1 mK in very low magnetic fields ($\lesssim 6$ G).

Further progress in this study of surface physics at millikelvin temperatures requires a neutron-scattering investigation of the structure of the second layer, and NMR measurements for submonolayer coverages, where three-particle exchange should produce ferromagnetic effects at melting densities, and at monolayer completion where indirect exchange should be observable. The experiments, already extremely difficult because of the very poor thermal conductivity of Grafoil,[17] will require extreme care; the understanding of exchange in solid ^{3}He and of the nature of liquid ^{3}He (quasisolid versus quasimagnetic picture) makes this effort worthwhile.

Two of us (H.F. and R.E.R.) thank the Conselho Nacional de Desenvolvimento Científico e Tecnológico (Brazil) for financial support during their stay in Grenoble. We thank B. Castaing, M. T. Béal-Monod, H. J. Lauter, M. G. Richards, and M. Roger for useful discussions.

---

[a]Permanent address: Instituto de Fisica, Universidade Federal de Rio de Janeiro, Rio de Janeiro, Brazil.

[1]A. I. Ahonen, T. Kodama, M. Krusius, M. A. Paalanen, R. C. Richardson, W. Schoepe, and Y. Takano, J. Phys. C **9**, 1665 (1976); A. I. Ahonen, T. A. Alvesalo, T. Haavasoja, and M. C. Veuro, Phys. Rev. Lett. **41**, 494 (1978); H. Godfrin, G. Frossati, D. Thoulouze, M. Chapellier, and W. G. Clark, J. Phys. (Paris), Colloq. **39**, C6-287 (1978); H. M. Bozler, T. Bartolac, K. Luey, and A. L. Thomson, Phys. Rev. Lett. **41**, 490 (1978); H. M. Bozler, D. M. Bates, and A. L. Thomson, Phys. Rev. B **27**, 6992 (1983); R. C. Richardson, Physica (Amsterdam) **126B**, 298 (1984).

[2]Grafoil is an exfoliated graphite manufactured by Union Carbide.

[3]H. Franco, H. Godfrin, and D. Thoulouze, Phys. Rev. B **31**, 1699 (1985).

[4]H. J. Lauter, H. Wiechert, and R. Feile, in *Ordering in Two Dimensions,* Proceedings of the International Conference on Ordering in Two Dimensions, Lake Geneva, Wisconsin, 1980, edited by S. K. Sinha (North-Holland, New York, 1980); K. Carneiro, L. Passell, W. Thomlinson, and H. Taub, Phys. Rev. B **24**, 1170 (1981).

[5]M. G. Richards, in *Phase Transitions in Adsorbed Films,* edited by J. G. Dash and J. Ruvalds (Plenum, New York, 1980).

[6]S. W. van Sciver and O. E. Vilches, Phys. Rev. B **18**, 285 (1978).

[7]C. Tiby, H. Wiechert, H. J. Lauter, and H. Godfrin, Physica (Amsterdam) **107B+C**, 209 (1981).

[8]J. R. Owers-Bradley, B. P. Cowan, M. G. Richards, and A. L. Thomson, Phys. Lett. **65A**, 424 (1978).

[9]M. T. Béal-Monod and A. Theumann, in *Ordering in Two Dimensions* (see Ref. 4), and references therein.

[10]M. Roger, Phys. Rev. B **30**, 6432 (1984).

[11]H. Jichu and Y. Kuroda, Prog. Theor. Phys. **67**, 715 (1982), and **69**, 1358 (1983).

[12]W. J. Mullin and A. Landesmann, J. Low Temp. Phys. **38**, 571 (1980).

[13]A. Sachrajda, F. R. Harris-Lowe, J. P. Harrison, R. R. Turkington, and J. G. Daunt, Phys. Rev. Lett. **55**, 1602 (1985).

[14]Other interpretations have been proposed (Knudsen flow of quasiparticles).

[15]Y. Okuda, A. J. Ikushima, and H. Kojima, Phys. Rev. Lett. **54**, 130 (1985).

[16]L. J. Friedman, S. N. Ytterboe, A. L. Thomson, and H. M. Bozler, Bull. Am. Chem. Soc. **31**, 282 (1986).

[17]L. D. Dillon, R. E. Rapp, and O. E. Vilches, J. Low Temp. Phys. **59**, 35 (1985).