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Condensation of ^{3}He in $2\frac{1}{2}$ dimensions and indirect exchange in adsorbed films

H. Godfrin
Centre de Recherches sur les Très Basses Températures, Centre National de la Recherche Scientifique,
Laboratoire associé a l'Université J. Fourier, Boîte Postale 166, 38042 Grenoble Cedex 9, France

R. E. Rapp
Centre de Recherches sur les Très Basses Températures, Centre National de la Recherche Scientifique,
Laboratoire associé a l'Université J. Fourier, Boîte Postale 166, 38042 Grenoble Cedex 9, France
and Instituto de Física, Universidade Federal do Rio de Janeiro, Bloco A 4° andar,
Cidade Universitaria, Caixa Postal 68528, Rio de Janeiro RJ 21945, Brazil

K.-D. Morhard, J. Bossy, and Ch. Bäuerle
Centre de Recherches sur les Très Basses Températures, Centre National de la Recherche Scientifique,
Laboratoire associé a l'Université J. Fourier, Boîte Postale 166, 38042 Grenoble Cedex 9, France

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The experimental properties of multilayer ^{3}He films adsorbed on graphite are presently described by a model assuming the existence of several structural phases in the second layer and a continuous growth of uniform liquid beyond second-layer coverages. We propose an alternative model, where second-layer ferromagnetism is induced by a condensed third-layer liquid, which provides a simple description of experimental data.

The observation of two-dimensional nuclear magnetism in ^{3}He films adsorbed on graphite[1] has motivated detailed studies of these systems.[2–5] The magnetic behavior of the second atomic layer is of particular interest: large antiferromagnetic exchange interactions are found at low coverages where this layer has probably a commensurate solid structure, whereas large ferromagnetic exchange is found at high coverages. In this last case, the solid is thought to have a solid incommensurate structure, and fluid ^{3}He is present above the second layer.

The nature of exchange processes in these systems is not well understood. In-plane multiple spin exchange[6] and indirect exchange[7] are able to describe qualitatively the experimental results, but some difficulties arise due to the incomplete knowledge of the second-layer phase diagram. Greywall proposed a possible phase diagram[2] compatible with heat-capacity data; according to this interpretation there would exist two commensurate phases, characterized according to NMR data[1,4] by antiferromagnetic exchange constants J of similar magnitude, and an incommensurate phase shown to be ferromagnetic by NMR experiments.

The observed[1,4] gradual transition from antiferromagnetism to ferromagnetism is ascribed to a coexistence of structural phases. In recent ultralow temperature experiments[3] the saturation magnetization of the second layer was found to have a linear variation as a function of second-layer coverage, providing support to a model of coexisting antiferromagnetic and ferromagnetic structural phases in the second layer.

We discuss in this article the possibility to interpret existing data using different assumptions for the phase diagram of multilayer films, motivated by recent theoretical work on two-dimensional liquid films.[8] We focus on the coverage region above 0.18 Å^{-2} where the second layer is solid. Following Greywall, liquid in the third layer is observed for coverages above 0.184 Å^{-2}, and fourth-layer promotion at about 0.24 Å^{-2}. This last effect occurs at the ferromagnetic peak coverage,[1] but this is fortuitous in the presently accepted interpretation.

Linear coverage dependences of the heat capacity and nuclear magnetization have been used to identify phase coexistence regimes.[1,2] However, helium layers are highly compressible and substantial density increase of a given layer is observed as a layer immediately on top begins to form. The density of a given layer is therefore nonlinear in total coverage, particularly at the beginning of layer formation and around layer completion. In the coverage range we are discussing, the second layer is reaching gradually its maximum density and the third-layer density grows initially slowly and then rises almost linearly. Therefore, a measured property cannot depend linearly both on total density and in second-layer density in the coverage range discussed here; the magnetization isotherms at low temperatures, however, were found to be linear as a function of total coverage in this range, and a coexistence regime was proposed initially between two phases of different magnetizations.[1] According to the model presently used, one would expect a linearity in second-layer density. This contradiction motivated us to investigate existing experimental data to determine the relevant variable for coverage dependences.

We find that several properties discussed below vary linearly with the third-layer density n_3 in a substantial range of total coverages. We conclude that the third-layer fluid cannot be uniform (contrarily to what is observed[2] for the first- and second-layer fluids). We are led then to the hypothesis that the third-layer fluid condenses in islands, and check the consequences of puddling on the interpretation of experimental results.

In the following we define n_3 as the difference between the total coverage n and that of the first and second layers ($n_3 = n - n_1 - n_2$); it includes therefore all fluid contributions above the two solid layers, and is equivalent to third-layer coverage below a density presently thought to be about 0.045 Å^{-2}.[2]

Let us discuss first the properties of the third-layer fluid. As shown in Fig. 1, the coefficient γ of the contribution linear in temperature in the heat capacity displays a characteristic increase as a function of density above the density-independent value expected for a noninteracting Fermi gas.[2] This variation reflects the increase of the effective mass, and is similar for the first and the second layer (the low γ value of the point at $n = 0.01$ Å^{-2} is due to the fact that the real liquid density is much lower, since a small amount of solid is formed at substrate defects; the liquid is not in the degenerate regime in the temperature range of the measurements). The variation observed in the case of the third layer is clearly different: it is linear in third-layer coverage for $n_3 \leq 0.04$ Å^{-2} (Fig. 2), as expected for a condensed liquid phase.

Let us examine the consequences of third-layer puddling on the magnetic properties of the second layer. The variation of the effective exchange constant of the second layer[1,4,5] as a function of third-layer density (Fig. 3) is linear from the antiferromagnetic to the ferromagnetic regime. This suggests an antiferromagnetic exchange in the naked second-layer solid and a ferromagnetic exchange mechanism in the fraction of the second-layer solid covered by liquid islands. The process changes radically when all the second layer is covered (the islands of liquid merge); the corresponding coverage (which was associated to fourth-layer promotion in Ref. 2) coincides naturally with the ferromagnetic peak in this model. The islands may induce ferromagnetic regions either by a "compression" (cage effect) of the underlying solid, or by the indirect exchange mechanism.[7] Given the larger magnitude of the ferromagnetic exchange constant, the latter mechanism is probably the dominant one. This provides a possible explanation to the fact that data obtained on preplated substrates[4] are identical to the data for pure ^{3}He films[1,5] in the strongly ferromagnetic regime (Fig. 3), despite the difference in adsorption potentials.[9]

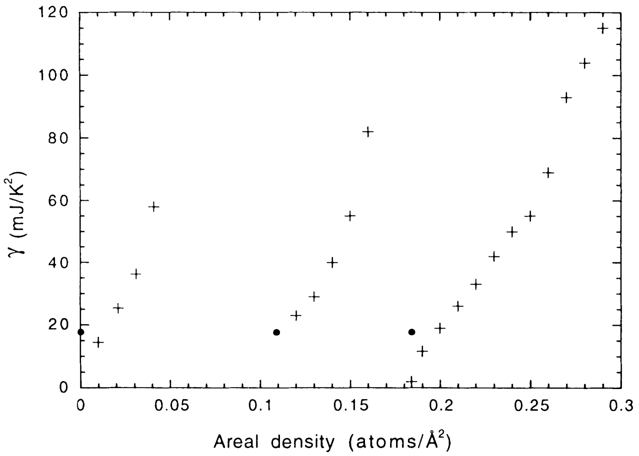

FIG. 1. Coefficient γ of the term linear in temperature of the heat capacity of ^{3}He films as a function of total coverage. This Fermi-liquid contribution is shown for the first, second, and third and upper layers. The dots represent the density-independent value expected for a two-dimensional uniform Fermi liquid (data from Ref. 2).

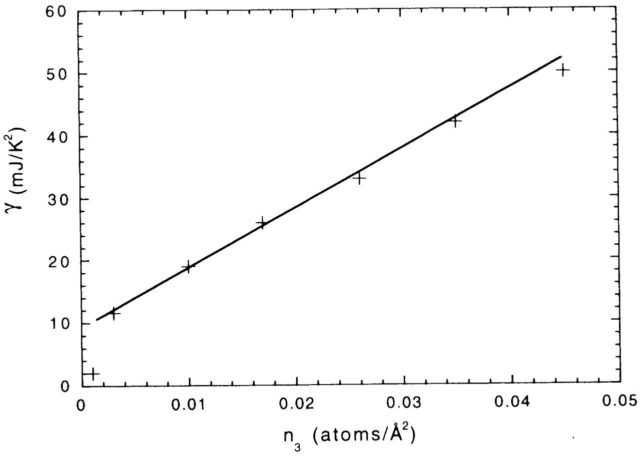

FIG. 2. Third-layer data for γ (see Fig. 1), represented as a function of the third-layer coverage n_3 (data from Ref. 2).

Low-temperature-heat capacity isotherms,[2] dominated by exchange in the second layer, display a similar behavior when shown as a function of the third-layer density (Fig. 4). It should be pointed out that the behavior as a function of the second-layer density n_2 is more complicated, and led to postulating the existence of a second

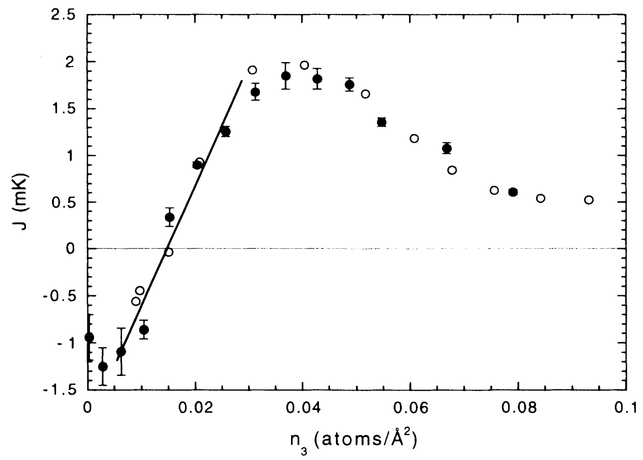

FIG. 3. Effective exchange constant J as a function of the third- and upper-layer coverage n_3 (see text). Filled circles correspond to pure ^{3}He films (Refs. 1 and 5), open circles to ^{4}He preplated films (Ref. 4).

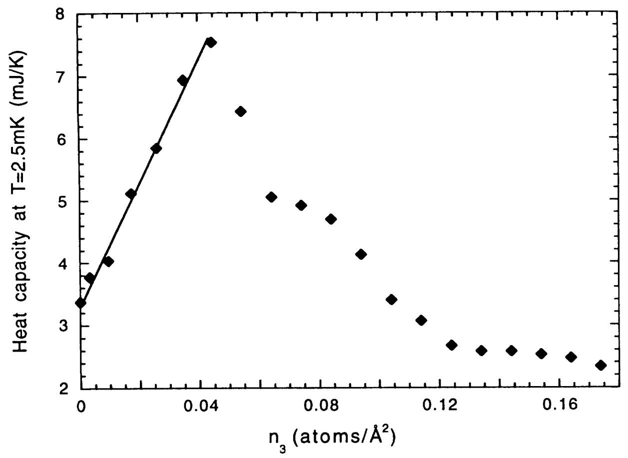

FIG. 4. Heat-capacity isotherm at 2.5 mK as a function of third- and upper-layer coverage (data from Ref. 2). Note that the linear region corresponds to two different coexistence regimes in the phase diagram of Ref. 2.

commensurate phase in the second layer,[2] a fact hardly compatible with the NMR data. The puddling hypothesis only requires solidification of the second layer.

Low-temperature magnetization data[3] are shown in Fig. 5 as a function of n_3. Again, linear behavior is observed, as expected for ferromagnetic domains controlled by the liquid islands. Note that the second-layer density is increasing in this coverage range; at the highest coverages, one obviously expects within the model presented here that this magnitude will become proportional to the *second*-layer coverage,[3] since all second-layer atoms are affected by the ferromagnetic exchange mechanism when the second layer is completely covered by liquid. The exchange constants determined at low temperatures[3] are larger than those found in the high-temperature regime;[1,4,5] the low-temperature behavior is dominated by the ferromagnetic contributions, whereas the higher-temperature exchange constants are effective constants resulting from the combination of ferromagnetic and antiferromagnetic exchange terms weighted by the relative amounts of both phases.[3] The puddling model explains naturally the ferromagnetic and antiferromagnetic phase coexistence and the evolution of the magnetization as a function of temperature and coverage, including the coverage dependence of the coherence length in the spin-wave regime.

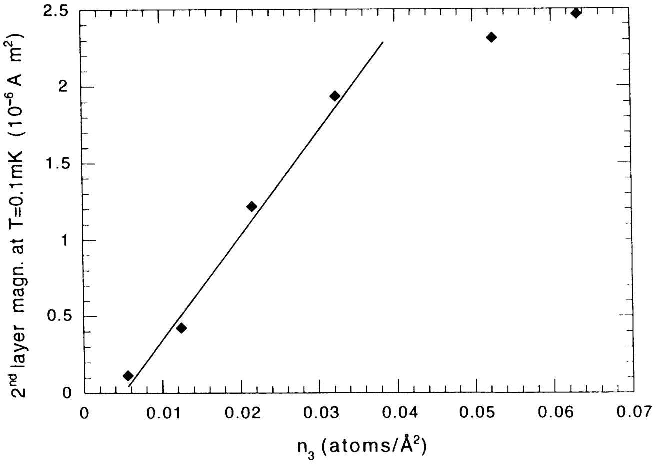

FIG. 5. Second-layer magnetization at low temperatures (corresponding to saturated ferromagnetic regions) as a function of the third- and further-layer coverage n_3 (data from Ref. 3). A linear behavior in n_3 is observed at low coverages. The small offset in the abscissa is probably due to differences in coverage scales with respect to Ref. 2 (see also Fig. 3). At the highest coverages the magnetization is expected to be proportional to the *second*-layer coverage (see text).

The regime linear in n_3, interpreted here as due to condensed liquid, extends approximately up to $n_3 = 0.04$ Å^{-2} in all these experiments, a reasonable value for a two-dimensional liquid. The work of Brami, Joly, and Lhuillier[8] clearly shows that delocalization of the ^{3}He atoms in the third dimension may lead to a self-bound liquid state. We believe that the softer potential normal to the substrate experienced by the third-layer liquid (in comparison with that of the first and second layers) makes condensation possible; liquids in the first, second, and third layer are particularly useful model systems for theoretical calculations.

The interpretation of the nuclear magnetism of ^{3}He multilayer films given here leads to a new interpretation of the second-layer phase diagram. In particular, the existence of the second commensurate phase is not needed to describe the low-temperature heat-capacity data. Also, fourth- and fifth-layer promotions may occur in this model at higher values of total coverage (compared to those assumed in Ref. 2): 0.26 and 0.32 Å^{-2}, respectively, are coherent with the data of Figs. 3 and 4, closer to the values expected from the bulk ^{3}He density, and probably better related to the theoretical predictions of Guyer for indirect exchange in multilayer films.[7]

A detailed analysis is presently difficult due to the uncertainty in the values of n_3; we have used the coverage values given by Greywall,[2] even though severe discrepancies exist with NMR and neutron-scattering data. Other difficulties are related to heterogeneity of the graphite substrate. In real samples the "islands" are probably located along the edges of the graphite platelets, and fill progressively the platelets area. At low liquid coverages heterogeneity effects may dominate, and a strict proportionality to third-layer coverage cannot be expected. The indirect exchange mechanism, for instance, will be affected when the liquid presence is restricted to the edges of the platelets.

The puddling hypothesis does not seem to contradict existing data, despite some weaknesses: the sudden increase of the coefficient γ of the term linear in temperature of the heat capacity (Fig. 2) at the lowest n_3 values cannot only be explained by an inaccurate n_3, except if one accepts some degree of surface reconstruction or heterogeneity (edge) effects (the presently accepted model also suffers from similar difficulties: for instance, a liquid-like heat capacity is observed[2] at a density $n = 0.178$ Å^{-2} where no liquid should be present). Also, heat-capacity data at temperatures on the order of tens of mK are not proportional to third-layer coverage; this may not be a serious criticism, since condensation should occur only at very low temperatures.[8] Another obvious oversimplification of this model is the assumption that the properties of the antiferromagnetic second-layer solid do not change as the total coverage is increased; certainly the second-layer density increase may even make the in-plane interactions become ferromagnetic;[6] this would lead to small deviations from the linear behavior in third-layer density. Present data are not precise enough to allow a more detailed discussion.

The third-layer liquid puddling model provides a new possible explanation of the physics of ^{3}He films. More work is needed to establish with precision the phase diagram and the density of the individual layers of adsorbed ^{3}He multilayer systems in order to understand their extraordinary two-dimensional nuclear magnetism.

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