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On the spin-liquid phase of two-dimensional ^{3}He

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Abstract

Multiple spin exchange processes lead to strong quantum frustration in low density solid phases of ^{3}He. It has been predicted theoretically that the 4/7 phase of the second layer of ^{3}He adsorbed on graphite is a two-dimensional RVB magnet with a spin-liquid ground state. We have measured the nuclear magnetization of the second layer of ^{3}He adsorbed on a Papyex substrate preplated by a monolayer of ^{4}He, using cw-NMR techniques, for a large temperature range (0.1–400 mK). Special attention has been devoted to carefully eliminating from our sample spurious phases and paramagnetic defects. The analysis of the data provides us with information on the presence of a spin gap in the excitation spectrum and on the density of states of the magnetic excitations of this phase.

1. Introduction

Helium-three (^{3}He), which has a nuclear spin of 1/2, allows us to create and investigate magnetic model systems under a large variety of experimental conditions of temperature, magnetic field, density and dimensionality. We have studied in detail [1, 2] two-dimensional solid ^{3}He adsorbed on graphite, showing that it constitutes a model system for almost localized fermions in two dimensions. These solid phases have a simple triangular structure, as seen by neutron diffraction [2]. Confined by their neighbours, the light helium particles experience a large zero-point motion, which favours quantum tunnelling of the atoms and, as a consequence of quantum statistics, large effective magnetic interactions between the nuclear spins. This is now understood well in terms of the Dirac–Thouless multiple spin exchange (MSE) model of quasi-localized magnetism [3–5]: groups of atoms experience coherent cyclic exchange motions and ring exchanges involving from 2–6 particles are found to be the most frequent processes.

The '4/7 phase' of adsorbed ^{3}He is a particularly simple bilayer system which has attracted considerable interest. It consists of a low density solid layer of ^{3}He, commensurate

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in a $4/7$ density ratio with respect to the underlying first solid layer (^{3}He, ^{4}He or HD) adsorbed on graphite. The first layer produces a corrugation potential which keeps the ^{3}He atoms of the second layer sufficiently separated to allow substantial quantum tunnelling motions. Under these conditions, nuclear exchange constants as large as several millikelvins are observed. The competition between different exchange processes (in-plane MSE processes) in the second layer leads to a highly frustrated novel type of magnetism, as pointed out by Roger [6]. The double-peak structure and the large density of states observed by heat capacity measurements at very low temperatures by Fukuyama *et al* [7] led these authors to suggest a spin-liquid ground state for this system.

The magnetization, on the other hand, displays a soft temperature dependence, Curie–Weiss-like, even for temperatures much smaller than the characteristic exchange constants [1, 8–14]. This behaviour is characteristic of frustrated systems [15].

Recent theoretical work on the MSE Hamiltonian has put forward interesting predictions for the ground state of the $4/7$ phase and its excitations [16–18]. A spin-liquid ground state is found to be selected by quantum fluctuations, for a specific range of exchange parameters. Also, a gap of the order of the four-spin exchange constant should separate the lowest singlet and triplet excitations.

Our experimental results on the magnetization of the $4/7$ phase allowed us to determine, using the data taken above 2 mK, the exchange parameters for the $4/7$ phase [13]. An excellent agreement with the theoretical phase diagram (in the exchange parameter space) was found. The objective of the present paper is to concentrate on the very low temperature magnetization of the $4/7$ phase and to examine the constraints that our data can place on theory.

2. Multiple spin exchange Hamiltonian

The magnetic properties of a collection of quasi-localized fermions are formally described quantum mechanically in terms of permutation operators [3]: $\mathcal{H} = \sum_2^n (-1)^n J_n \mathcal{P}_n$, where $\mathcal{P}_n$ is the operator for cyclic permutation of n particles. In solid ^{3}He, hard-core correlations lead to a significant probability of coherent exchange of several particles, as was pointed out by Thouless [4]. The MSE model was indeed successful in explaining the unusual magnetic properties of bulk solid ^{3}He [5, 19] and those of 2D ^{3}He [2, 7, 20–22]. Quantum Monte Carlo calculations of the various exchange frequencies have been performed with excellent accuracy for bulk ^{3}He [23]. For 2D ^{3}He adsorbed on graphite, the task is more difficult, and currently available results [24] are only semi-quantitative. However, the exchange constants for 2D systems can be determined from the analysis of thermodynamic measurements [22], using high temperature series expansions of the MSE Hamiltonian [25]. More precisely, we use a truncated Hamiltonian which retains the most significant exchange constants. These are the J_n involving the most symmetric rings of n nearest neighbours with (by decreasing magnitude) $n = 3, 2, 4, 6, 5$:

$$\mathcal{H}_{\mathrm{ex}} = J \sum^{(2)} \mathcal{P}_2 + J_4 \sum^{(4)} \mathcal{P}_4 - J_5 \sum^{(5)} \mathcal{P}_5 + J_6 \sum^{(6)} \mathcal{P}_6. \tag{1}$$

The sums correspond respectively to cyclic two-, four-, five- and six-particle permutation operators acting on spin variables. Cyclic n-particle exchanges, with n odd, can be expressed in terms of $(n-1)$-particle exchanges [4]. Three-particle exchange J_3 is incorporated in an effective pair exchange constant: $J = J_2 - 2J_3$. The model has therefore four exchange constants, which can be determined from the high temperature behaviour of the heat capacity and the susceptibility.

3. Experimental details

We have measured the magnetization of the 4/7 phase of ^{3}He adsorbed on exfoliated graphite by cw-NMR techniques at ultra-low temperatures (down to about 100 μK), in a magnetic field of 30.51 mT. Thermometry is provided below 10 mK by a Pt-wire NMR thermometer measured by the pulse spectrometer and above this temperature by a calibrated carbon resistance (Speer 100 Ω). Our exfoliated graphite substrate is a special Papyex sample, characterized by very large (about 50 nm) atomically flat platelets. The sample has a mass of 0.632 g. It was studied in detail by adsorption isotherms as well as by extensive NMR work at submonolayer and multilayer coverages. From these measurements we determined accurately the densities quoted hereafter for the adsorbed helium layers. The NMR lines (absorption and dispersion signals are recorded) are measured as the magnetic field is swept around the resonance frequency. The magnetic field of 30.51 mT used here is such that $\mu B \ll k_B T$ over all our temperature range.

We first performed a run at a submonolayer coverage of 9.28 atoms nm^{-2} to obtain an absolute calibration of the magnetization measured by the NMR spectrometer. At this coverage, the magnetization of the adsorbed ^{3}He follows the Curie law quite accurately. This run also allowed us to verify that the cell is in thermally good contact with the thermometers for the entire temperature range (0.1–400 mK) and to test the non-resonant radio-frequency heating of the graphite substrate.

In the experiments described here, the substrate was first preplated by an atomic layer of ^{4}He. It is important to note that we then adsorbed a given amount of ^{3}He, which was kept constant throughout the experiment, and that we subsequently added small amounts of ^{4}He, used as a 'pushing gas', to achieve a fine-tuning of the ^{3}He density all across the 4/7 phase existence domain.

We started with a total coverage of ^{4}He of 6.370 ccSTP, and we added an amount of ^{3}He (2.973 ccSTP), which is slightly lower than that needed to form the commensurate solid 4/7 phase for this total coverage. We then added by steps controlled amounts of ^{4}He.

4. Experimental results

The raw data and the analysis of the high temperature regime have been presented in a previous publication [13]. We briefly summarize here the main results. For total ^{4}He coverages of 6.557, 6.706, 6.919, 7.131, 7.344 and 7.641 ccSTP we are in the 4/7 phase area of the phase diagram. The onset of ferromagnetism, which corresponds to the formation of an incommensurate solid ^{3}He phase of a very different nature, is reached for a ^{4}He quantity of 8.043 ccSTP. The 4/7 phase is thus present for a large fraction of the phase diagram. However, depending on the coverage, we observe a coexistence of this phase with second or third layer liquid and also, for some coverages, with almost paramagnetic impurities. These phases introduce magnetic contributions that we have subtracted to determine the intrinsic magnetization of the 4/7 phase, shown in figure 1. Two sets of data are shown in this graph: the dots correspond to the total solid-like contribution measured for the coverages 6.919, 7.131, 7.344 and 7.641 ccSTP. For these data points, the solid is in a pure 4/7 phase, the only additional contribution originates from the liquid and it has been subtracted. We call this the 'clean regime'.

The circles shown in figure 1 correspond to the solid magnetization after subtraction of a relatively large Curie–Weiss contribution, characterized by an antiferromagnetic Weiss temperature. We attribute this contribution to ^{3}He spins localized in defects of the two-dimensional helium crystals and in defects of the substrate. These impurities are particularly visible at low densities (data not presented here), when they coexist with 2D fluid; we have shown that, under these conditions, their magnetization follows a Curie–Weiss law with a Weiss

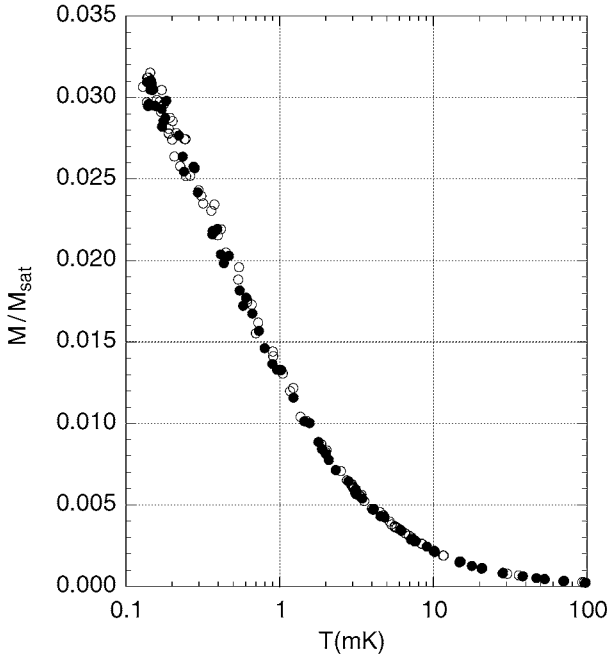

Figure 1. Temperature dependence of the magnetization normalized to the saturation value, for the $4/7$ phase of ^{3}He adsorbed on Papyex preplated by a monolayer of ^{4}He. Dots indicate the results for coverages within the clean regime and circles correspond to coverages where impurities are present (their contribution has been subtracted).

temperature of -105 μK. This is larger in magnitude than that of the -33 μK we observe in the present experiment, where densities are higher. Furthermore, we have analysed data at much higher coverages, in the ferromagnetic regime, and we also find a significant contribution of impurities. This was already pointed out by Schiffer *et al* [26]; we have reanalysed their data and determined that 28% of the second layer ^{3}He spins are almost paramagnetic, with a Weiss temperature of +13 μK. This trend agrees well with the known density dependence of the exchange processes. We shall come back to the impurity problem later on, since their presence is of considerable importance at the lowest temperatures.

The analysis allows us to determine the amount of 2D liquid and that of impurities present at each coverage. The results are shown in figure 2. For the first coverage, a small amount of liquid is present in the second layer. The first increase in ^{4}He coverage causes the second layer to solidify completely (point L). Then, as the amount of ^{4}He is further increased, the number of ^{3}He atoms in the second layer solid is found to decrease, as ^{3}He atoms are promoted as liquid into the third layer. In the regime between the points L and D of figure 2 the ^{3}He defects are very efficiently removed by the added ^{4}He. The preferential adsorption of ^{4}He atoms, due to their larger mass and hence smaller zero point motion, is therefore an excellent tool to characterize and remove defects. For a substantial range of coverages, between points D and F in figure 2, we observe a one-by-one replacement, i.e. one ^{4}He atom enters the second layer, expelling exactly one ^{3}He atom to the third layer liquid. This very specific behaviour can be clearly ascribed to the commensurate nature of the $4/7$ phase.

The presence of impurities strongly affects the low temperature magnetization data, a feature which is well known in the field of frustrated magnets. This is emphasized in figure 3

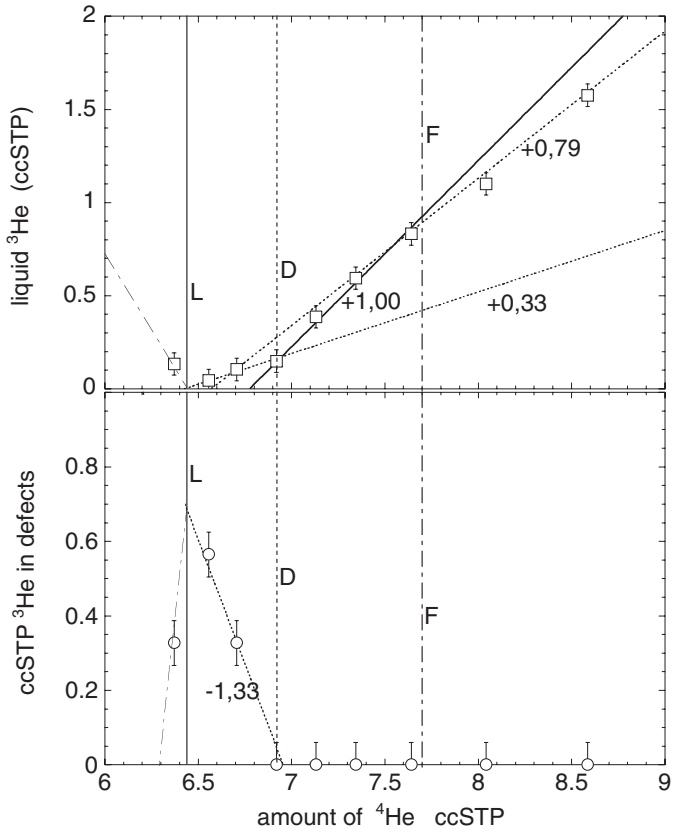

Figure 2. Amount of two-dimensional liquid ^{3}He (upper graph) and amount of ^{3}He localized in substrate defects (lower graph), expressed in ccSTP, as a function of the amount of ^{4}He introduced to compress the second layer solid ^{3}He. The full line (L) indicates the end of the liquid–solid ^{3}He coexistence in the second layer. The maximum number of defects is seen at this point. The broken line (D) marks the end of the regime where defects are present, while (F) shows the beginning of the ferromagnetic phase. The numbers correspond to the slope of the straight lines fitting the data in some regions of this diagram. Note that the slope of 1 between D and F (clean regime) corresponds to a one-by-one displacement (a ^{4}He atom removes exactly one ^{3}He atom).

where the paramagnetic impurities dominate the low temperature magnetization for coverages near and above the $4/7$ phase completion, until a clean regime is reached where these effects disappear.

5. $4/7$ phase magnetization data analysis

The procedure described above provides us with clean data of the intrinsic magnetization of the $4/7$ phase as a function of temperature (figure 1). Note that for the very low field used in this experiment, the magnetization is simply proportional to the susceptibility. As seen on figure 1, even at the lowest temperatures the magnetization is well below the saturation value.

The data analysis is done in two steps. First, we analyse quantitatively our high temperature data (down to 2 mK) using Padé approximants of the high temperature series expansions (HTSE) of the MSE Hamiltonian [27].

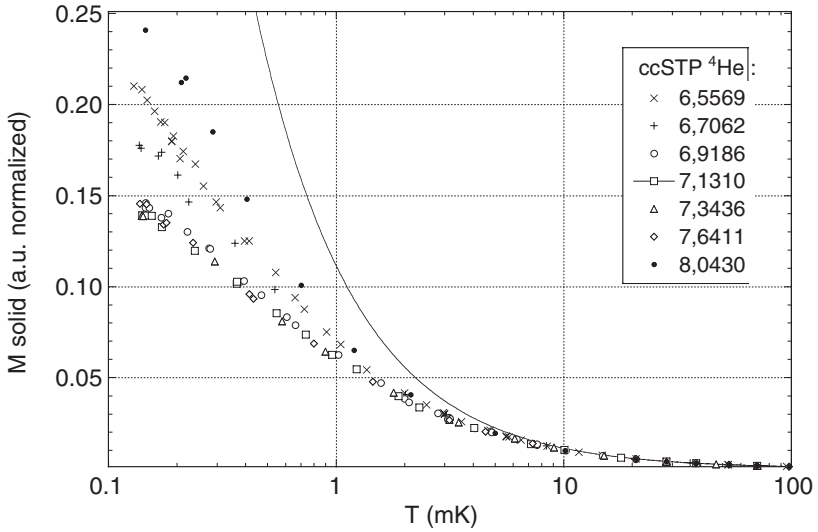

Figure 3. Nuclear magnetization of the adsorbed solid ^{3}He (the liquid contribution has been subtracted) normalized to the amount of solid, for different total coverages. Note the large influence of quasi-paramagnetic defects at low coverages and the good superposition of the data at higher coverages (clean regime). The dots correspond to a coverage slightly above the transition to the ferromagnetic regime. The Curie law associated with the amount of solid ^{3}He, extrapolated from high temperatures, is shown as a full curve.

5.1. Multiple spin exchange analysis

We assume the hierarchy $J_2^{\text{eff}} > J_4 > J_6 > J_5$ predicted by theory and verified for pure ^{3}He films [22]. We also limit the parameter space to a physically sound range, consistent with our pure ^{3}He films' data. Within these hypotheses, we determine the main exchange constants with an accuracy of 0.1 mK: $J_2^{\text{eff}} = -2.8$ mK, $J_4 = 1.4$ mK, $J_5 = 0.4^5$ mK and $J_6 = 1.2^5$ mK. The leading term J_χ of the susceptibility is found to be very small and slightly ferromagnetic: $J_\chi = 0.07 \pm 0.1$ mK. This is a signature of a strongly frustrated system. The Curie–Weiss temperature ($\Theta_{\text{CW}} = 3J_\chi = +0.2$ mK) is quite different from the value obtained by a simple empirical Curie–Weiss fit and has the *opposite* sign ($\Theta_{\text{CW}} = -0.9$ mK), as a result of the strong cancellation of the Heisenberg term due to multiple spin exchange. The exchange constants deduced from this analysis place the system in a region of the multiple spin exchange parameters space where theory predicts a spin-liquid ground state [16–18]. Nevertheless, it lies very close to the border between the spin-liquid and the ferromagnetic ground states. As a consequence, the spin gap predicted for the triplet excitations in the spin-liquid phase is expected to be strongly reduced, i.e. its magnitude can be much smaller than J_4.

5.2. Spin wave analysis

The spin-liquid ground state predicted theoretically has $S = 0$. Its magnetization at small fields should display an exponential behaviour associated with the presence of a spin gap. Unfortunately, calculations based on the MSE model, which could describe the low temperature susceptibility, are not yet available. Small systems (up to 24 spins) have been solved exactly, but their low temperature behaviour is dominated by finite size effects below 1 mK. These calculations, however, give a qualitative picture of the low-lying excitations of the 4/7 phase.

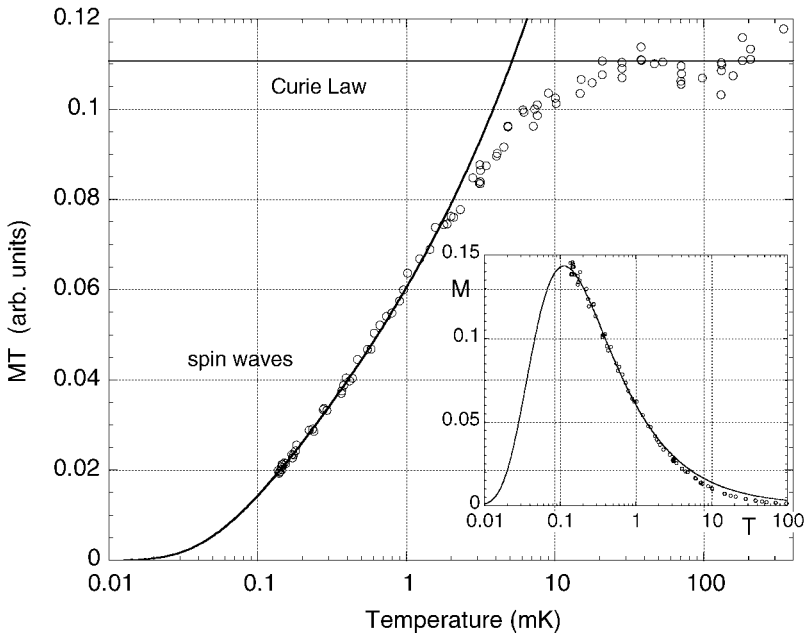

Figure 4. Low temperature behaviour of the magnetization of the 4/7 phase of ^{3}He on ^{4}He adsorbed on graphite. The main graph shows the product of the magnetization and temperature, and the inset the magnetization. The full curve is a fit to the data based on a model of gapped spin-wave-like excitations, valid at low temperatures (see text).

Following a method we have used earlier [2] to deal with the low energy excitations of the ferromagnetic phase of adsorbed ^{3}He, we propose to describe the system thermodynamics by assuming spin-wave-like excitations. This is clearly an approximation, since the present system is not magnetically ordered. We assume that the low lying excitations are $S = 1$ bosons characterized by a wavevector k and a dispersion relation given by

$$\mathcal{E}(\vec{k})_{\sigma=\pm1} = \Delta + J\, k_B S|(|\vec{k}| - k_0)d|^n - g_N \mu_N \sigma |\vec{B}|.$$

The parameter n allows modelling the density of states of these excitations and J is an effective interaction which, as the spin gap Δ, depends on the exchange constants.

The very low temperature magnetization can then be calculated using techniques described, for instance, by Troyer *et al* [28]. This leads to the following expression:

$$M(T) = \mathcal{A}\frac{C}{T}\left(\frac{T}{JS}\right)^{2/n} \exp\left(-\frac{\Delta}{T}\right).$$

It appears that such an expression fits adequately our data, as shown in figure 4. We find, within about 10% for both figures, $\Delta = 75\ \mu$K and $n = 6$.

The value $n = 6$ is quite unusual. However, such a magnon dispersion relation has been found by Momoi *et al* [29] for the *uuud* phase of 2D ^{3}He, which is one of the possible phases within the MSE model. The magnitude of the gap depends somewhat on the temperature dependence of the prefactor of the exponential, but its existence is clearly seen in our data. This can be emphasized in a different plot, suggested by Troyer *et al* [28]. The logarithmic derivative of the magnetization as a function of the inverse temperature is simply given by the expression

$$-\frac{\partial \ln M(T)}{\partial 1/T} = \Delta - \left(1 - \frac{2}{n}\right)T.$$

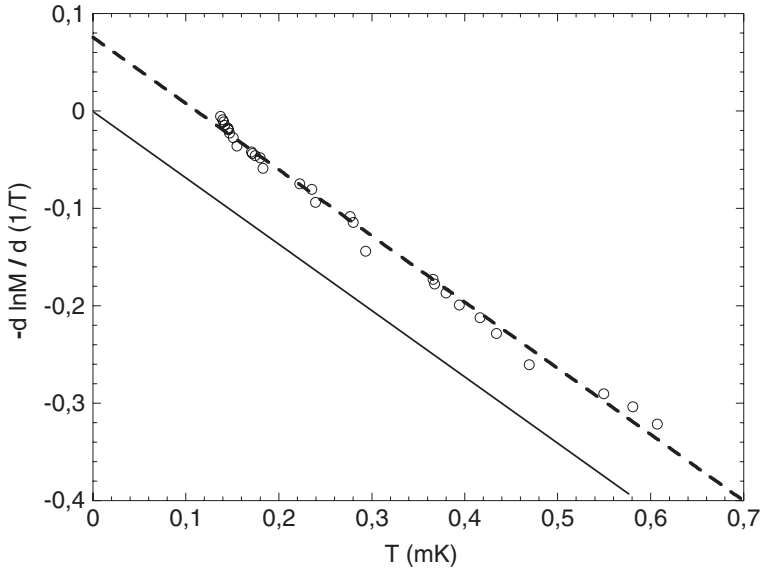

Figure 5. Derivative of the logarithm of the magnetization of the 4/7 phase with respect to the inverse temperature, as a function of temperature. The full line indicates the behaviour expected for a gapless system. The intercept of the broken line with the vertical axis yields the gap value, of about 0.075 mK.

Our data are shown in figure 5. The derivative of the $M(T)$ curve has been calculated numerically around each measured data point. The intercept of the straight line obtained gives the gap value, quoted before. Note that our temperature range is sufficiently low to allow a clear observation of the gap, with a minimum interference of the impurity spins. As seen in the inset of figure 4, the magnetization of the 4/7 phase should rapidly decrease below 100 μK and thus any small amount of localized spins would eventually dominate at lower temperatures.

6. Conclusions

We have studied the magnetic properties of the 4/7 phase of ^{3}He adsorbed on ^{4}He preplated graphite. We have investigated in detail the role of impurities, essentially ^{3}He atoms trapped in defects. This has allowed us to determine the intrinsic magnetization of the 4/7 phase for a very large temperature range. The data above 2 mK have been used to determine the main multi-spin exchange constants, thus entirely characterizing the Hamiltonian of the system. Our experimental values confirm quantitatively the phase diagram of Misguich *et al*, who predicted theoretically a spin-liquid ground state for this exchange constant range. At very low temperatures, well below the typical exchange constants, the magnetization is still small (about 3% of the saturation magnetization at 100 μK in the applied field of 30.51 mT). It displays a slow increase as the temperature is decreased, a typical feature of frustrated systems.

The very low temperature region has been analysed in the framework of a simple effective model. The model fits the data well, showing the presence of a gap for the triplet excitations of the order of 75 μK. We find that the density of states is unusually high at low energies and is described by a k^6 term in the dispersion relation similar to that found by Momoi *et al* [29] in the *uuud* phase. We believe that this could be a general behaviour of energy band narrowing associated with frustration. In this picture, frustration does not allow us to find spin arrangements which take full advantage of the interactions, in contrast to the case of ordered states, where some (usually simple) spin states can reach very small energies.

The system investigated here, that we denote in short by ^{3}He/^{4}He/graphite, displays unusual magnetic characteristics which agree well with the theoretical prediction of a resonating valence bond spin liquid. Other systems of the same type, obtained using different first layer atoms (^{3}He/^{3}He/graphite [30] or ^{3}He/HD/graphite [8, 11, 12, 14]) show a similar behaviour at high temperatures. ^{3}He/^{3}He/graphite is not very useful for ultra-low temperature studies, since the signal of the first layer masks that of the second layer. Experiments performed on ^{3}He/HD/graphite show that the exchange constants are very large, which is convenient from the experimental point of view. However, little is known about the amount of defects in HD coated graphite. It would be particularly interesting to perform the systematic measurements described here on such a system. Due to the first-principles nature of its theoretical description and the simplicity of its experimental properties, the $4/7$ phase of ^{3}He adsorbed on graphite can be considered as one of the purest realizations of frustrated magnetism.

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