Quantum Frustration in the "Spin Liquid" Phase of Two-Dimensional ^{3}He

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We have measured the ultralow temperature and low field magnetic susceptibility of the $\frac{4}{7}$ phase of two-dimensional ^{3}He adsorbed on graphite preplated by one layer of ^{4}He. The experiments are performed by progressively adding ^{4}He to the system, thus suppressing in a controlled way the ^{3}He atoms trapped in substrate heterogeneities. This procedure enables us to determine the intrinsic properties of this spin $\frac{1}{2}$ model magnet in the zero field limit. The results show quantitatively that the system is strongly frustrated by multiple spin exchange interactions. A characteristic gapped spin liquid behavior is observed at ultralow temperature.

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Strongly frustrated magnets are of fundamental interest due to the possible existence of disordered ground states. Recent studies have concentrated on spin chains and ladders, Mott insulators with orbital degeneracy, disordered systems, and also geometrically frustrated systems where the incompatibility of the local antiferromagnetic interactions with the global symmetry of the lattice leads to unconventional magnetic properties at low temperatures [1,2]. The main features of frustrated magnets are the existence of a large density of low energy states evidenced by heat capacity experiments and the extremely weak departure from the Curie-Weiss susceptibility even at very low temperatures [3]. The expectation that highly frustrated systems may have a disordered *spin liquid* ground state, characterized by a finite correlation length at zero temperature and a singlet-triplet spin gap in the excitation spectrum has motivated an active search of suitable experimental model magnets.

A unique example of a strongly frustrated two-dimensional $S = \frac{1}{2}$ nuclear magnet is provided by a solid layer of ^{3}He adsorbed on a graphite substrate [4–9]. In this quasilocalized quantum system, the large zero-point motion favors cyclic permutations of the ^{3}He atoms. Such exchange processes lead to an effective magnetic interaction between the nuclear spins formally described by the Hamiltonian $\mathcal{H} = \sum_2^n (-1)^n J_n \mathcal{P}_n$ where $\mathcal{P}_n$ is the operator for cyclic permutation of n particles. High order spin exchanges are particularly important in a hard-core solid like ^{3}He; indeed, a quantitative description of the magnetic properties involves several cyclic processes up to six particle exchange [9].

The cyclic exchange of an even number of particles contributes to an antiferromagnetic Heisenberg coupling, while that of an odd number of particles has the opposite (ferromagnetic) sign [10]. In addition, multiple spin exchange processes involving four or more particles introduce new physics due to higher order effective coupling terms in the Hamiltonian [5]. The impossibility to satisfy simultaneously the spin configurations favored by different multispin exchange processes gives rise to a sophisticated *quantum frustration* in addition to the geometrical frustration associated with the triangular lattice structure.

We will concentrate in this Letter on the properties of the second layer of adsorbed ^{3}He which forms a low density solid commensurate with respect to the first layer triangular solid in a $\frac{4}{7}$ density ratio [11]. The second layer forms strictly speaking a triangular Bravais lattice with a basis. However, quantum Monte Carlo calculations show that the exchange constants are essentially site independent, and it can be considered as a simple triangular lattice [12].

Ishida *et al.* [8] have shown that the second layer heat capacity displays a double peak structure and an unusual temperature dependence at the lowest temperatures ($T \approx 90\ \mu$K). This feature led the authors to the suggestion of a disordered ground state for the $\frac{4}{7}$ phase.

Recently, we have been able to determine experimentally the multispin exchange constants up to six particle exchange [9] as a function of the second layer areal density, thus completely determining the Hamiltonian of this system. High order ring exchanges, in particular four spin exchange, are found to be of the order of several millikelvin. According to exact diagonalizations of finite clusters for the multiple spin exchange (MSE) model, a gapped spin liquid ground state is expected for the experimentally determined exchange parameters [13].

The first measurement of the nuclear susceptibility of the $\frac{4}{7}$ phase at temperatures well below the exchange energies showed a substantial deviation from the Curie-Weiss law at temperatures on the order of 100 μK [14]. It was not clear, however, whether the very peculiar temperature dependence observed in this experiment was an intrinsic property of this strongly frustrated system, or an artifact

due to the presence of substrate heterogeneities or to the large field used in this experiment, where k_BT was on the order of μB at the lowest temperatures.

In this Letter we report the determination of the intrinsic nuclear magnetic susceptibility of the $\frac{4}{7}$ phase of ^{3}He down to temperatures of 100 μK in low magnetic fields. The measurements are done by preplating a Papyex exfoliated graphite substrate by one layer of ^{4}He and introducing a quantity of ^{3}He slightly lower than that needed to form the commensurate solid $\frac{4}{7}$ phase. Then, and this is an essential feature of this work, ^{4}He is added to the system while keeping the ^{3}He quantity fixed at 2.97 ccSTP (cm^3 of gas in standard temperature and pressure conditions).

This procedure allows us to finely tune the formation of the $\frac{4}{7}$ phase taking advantage of the preferential adsorption of ^{4}He to suppress progressively the magnetic signal of the ^{3}He atoms trapped at heterogeneities [15].

A run at a submonolayer coverage (9.28 atoms/nm^2) where the magnetic susceptibility follows a Curie law enables us to calibrate the NMR spectrometer and to verify that the cell is in good contact with the thermometers in the entire temperature range (0.1–400 mK). The temperature is measured using a pulsed Pt-NMR thermometer and a calibrated carbon resistance thermometer. A magnetic field of 30.5 mT was used in order to ensure that $\mu B \ll k_BT$ for all temperatures. Further experimental details are given elsewhere [14].

The evolution of the nuclear susceptibility in the vicinity of the $\frac{4}{7}$ phase as we add ^{4}He is shown in Fig. 1. At high temperatures all curves display the Curie behavior corresponding to the constant ^{3}He amount. At intermediate temperatures one observes a decrease in magnetization as we increase the ^{4}He quantity. This is due to the promotion of ^{3}He atoms into the third layer liquid which has a smaller magnetization than the solid. The low temperature magnetization is emphasized in the inset. In this plot we subtracted the contribution of the liquid and normalized the magnetization to the quantity of solid ^{3}He remaining in the second layer. We observe initially (for amounts of ^{4}He equal to 6.56, 6.71, and 6.92 ccSTP) a rapid decrease of the magnetization. We then reach a regime where the magnetization of the second layer solid remains remarkably unaffected for a large range of ^{4}He coverages (open symbols: 6.92, 7.13, 7.34, 7.64 ccSTP; see also Fig. 2). Finally, at a ^{4}He quantity of 8.04 ccSTP (filled circles) the system changes its nature, the sudden increase of the susceptibility indicating the onset of ferromagnetism.

This evolution can be understood by considering the rearrangement of the second layer as ^{4}He atoms are added. The second layer contains initially atoms in the $\frac{4}{7}$ phase coexisting with some ^{3}He atoms located at substrate heterogeneities. These strongly bound atoms are preferentially replaced by ^{4}He atoms, giving rise to the initial decrease of the low temperature susceptibility. When these defect atoms have been removed, the effect of the addition of ^{4}He is to promote atoms from the $\frac{4}{7}$ phase to the third layer liquid. In this regime, the magnetization normalized to the amount of solid ^{3}He does not change; only the amount of $\frac{4}{7}$ phase varies.

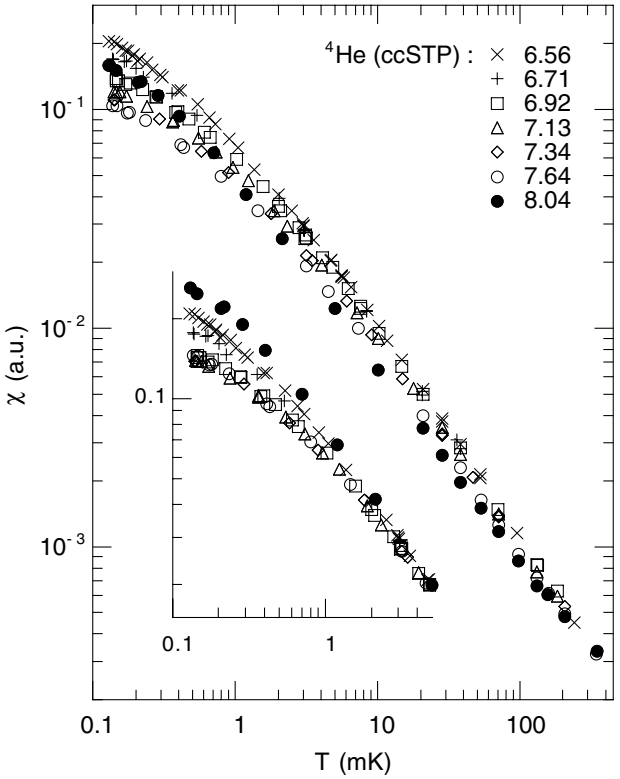

FIG. 1. Nuclear susceptibility of 2D ^{3}He in the vicinity of the $\frac{4}{7}$ phase as ^{4}He is added to the system. Inset: the liquid contribution has been subtracted and the magnetization normalized to the quantity of ^{3}He solid in the second layer.

This scenario is supported by a quantitative analysis. By fitting the magnetization at temperatures $T \geq 2$ mK we are able to determine the quantity of ^{3}He atoms pushed into the liquid third layer (Fig. 2a). We can then extract the quantity of ^{3}He atoms trapped in the substrate heterogeneities (Fig. 2b) by fitting the low temperature data as a coexistence between the $\frac{4}{7}$ phase and defects having a temperature dependence close to Curie law [16].

At point B in Fig. 2, all the trapped ^{3}He atoms have been replaced. It is interesting to note that both graphs show a discontinuity at this point.

The total amount of atoms trapped in substrate heterogeneities when the second layer has completely solidified is of the order of 16% of the second layer. This is consistent with heat capacity measurements of Ishida *et al.* [8].

The system then enters a region (from points B to E) where all magnetization curves of the second layer solid are identical (see open symbols in the inset of Fig. 1). All defects have been replaced by ^{4}He atoms, and we thus measure the intrinsic magnetic properties of the $\frac{4}{7}$ phase. Here one ^{4}He atom replaces one ^{3}He atom. In this plateau region, which is approximately 3 times wider in density than for the pure ^{3}He system [17,18], the amount

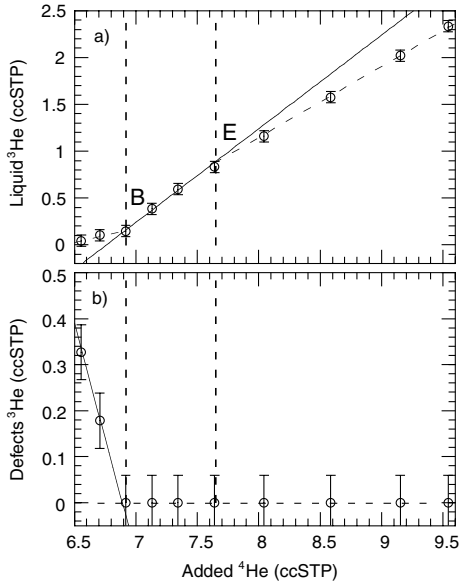

FIG. 2. Evolution of different properties as we add ^{4}He. (a) Amount of ^{3}He atoms promoted to the third (fluid) layer. A slope of one (solid line) corresponds to a situation where one ^{4}He atom replaces one ^{3}He atom in the second layer. (b) Quantity of second layer paramagnetic defects. The points B (beginning) and E (end) define the region where adding ^{4}He does not change the magnetic behavior of the solid layer. The error bars represent 2% of the ^{3}He quantity in the second layer.

of ^{4}He in the second layer varies from 20% to 40%. This demonstrates that the $\frac{4}{7}$ phase is very stable.

Another important conclusion can be drawn from the fact that in the plateau region the amount of ^{3}He present in the third layer grows substantially as we add ^{4}He, whereas the magnetism of the second layer remains unchanged. This provides a direct proof that the third layer liquid does not affect the second layer exchange. The same conclusion was reached for films of much higher density in our previous work [9]. The evolution of the magnetism of ^{3}He films as a function of density can therefore be described consistently by the *in-plane* MSE model.

At a proportion of roughly 40% of ^{4}He atoms in the second layer (point E) the system becomes ferromagnetic. Here the number of ^{3}He atoms promoted into the third layer is smaller than the number of ^{4}He atoms added to the second layer, which is a signature of the compression of the second layer solid. The system then passes a ferromagnetic anomaly as already seen in the pure ^{3}He system [19] and other preplated systems [20,21]. In Fig. 3 we show the magnetization of the $\frac{4}{7}$ phase obtained after subtraction of the liquid and defect contributions and normalization to the same quantity of ^{3}He solid. All our data collapse onto a single curve, showing that the measured magnetization is an intrinsic property of this strongly frustrated system.

For comparison we also display previous measurements in higher magnetic fields [14], to which we apply the same analysis. We observe no significant difference in the temperature dependence for the two different magnetic fields which indicates that the measurements are performed in the zero field limit of the MSE Hamiltonian and confirms our previous findings. In recent measurements on a different system, the $\frac{4}{7}$ phase of ^{3}He on a Grafoil substrate preplated by HD, a field independent susceptibility was also observed [22].

The susceptibility follows a Curie-Weiss law down to very low temperatures, in agreement with previous results. Indeed, the system almost acts like a collection of free spins in spite of the strongly interacting environment due to multiple spin exchange. We are able to analyze quantitatively our data using a Padé approximant of the high temperature series expansions (HTSE) of the MSE Hamiltonian [23] down to temperatures of the order of 2 mK. With this approach, we determine the different exchange constants with an accuracy of 0.1 mK. We assume the hierarchy $-J_2^{\text{eff}} > J_4 > J_6 > J_5$ predicted by theory and verified for pure ^{3}He films. We also limit the parameter space to a physically sound range, consistent with pure ^{3}He films data [9]. Within these hypotheses we find reasonable exchange constants: $J_2^{\text{eff}} = -2.8$ mK, $J_4 = 1.4$ mK, $J_5 = 0.4^5$ mK, $J_6 = 1.2^5$ mK. The leading term J_χ of the MSE-HTSE is found to be very small, and even slightly ferromagnetic: $J_\chi = 0.07 \pm 0.1$ mK. Such a small effective exchange constant is a signature of a strongly frustrated system. The corresponding Curie-Weiss temperature which can be deduced ($\Theta_{\text{CW}} = 3J_\chi = +0.2$ 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). This clearly demonstrates the strong

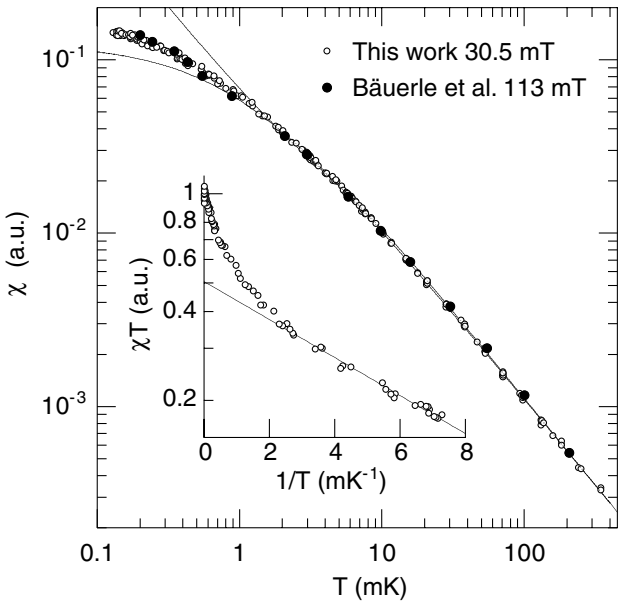

FIG. 3. Magnetic susceptibility of the pure $\frac{4}{7}$ phase. The solid lines correspond to the [2,3] Padé approximant of the HTSE of the MSE Hamiltonian [23] (upper curve), and to a Curie-Weiss fit (lower curve). The inset emphasizes the low temperature behavior of the susceptibility multiplied by temperature. The solid line shows an exponential decrease.

cancellation of the Heisenberg term due to multispin exchange.

The leading term of the heat capacity series expansion deduced from the present susceptibility data is $J_C = 1.6$ mK. The exchange parameters are all slightly smaller than those found in pure ^{3}He films [9] as would be expected since the latter have a slightly lower density.

We do not observe any feature in the susceptibility down to temperatures of 100 μK which could be interpreted as a finite temperature phase transition [5,8,13]. Our susceptibility results suggest that the system displays a very pure two-dimensional behavior [24]. Note, however, that here we use a small magnetic field, and that susceptibility and heat capacity are not equally sensitive to a possible phase transition.

The magnetization is large at very low temperatures, a signature of the large density of low energy magnetic excited states typically found in strongly frustrated systems [2,3]. However, we observe that χT decreases exponentially at the lowest temperatures as emphasized in the inset of Fig. 3. Such a behavior is expected for a spin liquid ground state having a magnetic gap.

Exact diagonalizations predict two different ground states for the MSE model [13]. In the region where predominantly ferromagnetic (FM) interactions are of importance the ground state is simply a FM $S = N/2$ ordered state. However, in a parameter range where antiferromagnetic (AF) interactions are dominant ($S = 0$), no long range correlations are observed even at $T = 0$ and a quantum spin liquid ground state is proposed.

For reasonable exchange parameters an excitation gap of the order of J_4 is predicted. Approaching the AF/FM transition line the gap is expected [13] to become much smaller than J_4.

From our measurements, we can deduce a magnetic gap of approximately 100 μK. Such a small value is consistent with the theoretical predictions, since our experimentally determined exchange parameters fall close to the AF/FM transition line; clearly, more detailed calculations would be desirable.

It is important to note that our experiment rules out the possibility that the observed gap is due to trivial size effects. As already mentioned, while the size of the solid ^{3}He domains is reduced by approximately 30% in the plateau region, the normalized magnetization remains unaffected.

Valuable information on the nature of the spin gap should be obtained by susceptibility measurements at lower temperatures while complementary heat capacity measurements would provide an independent determination of the exchange constants of this very pure model system.

In conclusion, we have measured the intrinsic magnetic susceptibility of the $\frac{4}{7}$ phase of two-dimensional ^{3}He by progressively suppressing the magnetization of the ^{3}He atoms trapped in substrate heterogeneities. We have clear evidence that in this system, the third layer liquid is not participating in the magnetic exchange and hence in-plane multispin exchange determines the magnetic properties of this 2D system. The unusual magnetization curve obtained here corresponds to a unique strongly frustrated two-dimensional $S = \frac{1}{2}$ quantum magnet whose properties are quantitatively described by the MSE model. Our results are consistent with a gapped spin liquid ground state. The gap is found to be 1 order of magnitude smaller than the exchange constants. The nature of the AF/FM transition as well as the value of the excitation gap at the transition is still an open and challenging question.

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