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Finite size effects in ferromagnetic ^3He nano-clusters

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Abstract. ^3He adsorbed on Graphite enables to create model 2D ferromagnetic Heisenberg systems. The exchange energies are of the order of 2 mK, typical sizes on the order of a thousand spins.

By adding ^4He (which is non magnetic) to the system, one can tune the effective size of one ferromagnetic domain. Up to now, the theoretical tools available did not allow a quantitative understanding of the magnetism of these clusters. For the first time, "engineered" ferromagnetic nano-clusters are compared to accurate theoretical models in order to understand the finite size effects. The experimental magnetization of a cluster of about 16 spins is compared to exact diagonalization and Monte-Carlo simulations based on the Heisenberg Hamiltonian.

1. Introduction

^3He atoms adsorbed on graphite are a realization of an *ideal* two dimensional (2D) $S = \frac{1}{2}$ nuclear magnetic system. Indeed, the binding energy is very large (of the order of 100 K), which ensures that at mK temperatures the system is perfectly confined in the substrate's plane. Moreover, the large sizes of the exfoliated graphite platelets ($\approx 500 \text{ \AA}$ for Papyex [1] sheets) enable truly 2D studies.

The 2D solid is described by the multiple spin exchange model (MSE) [2, 3], with exchange constants being a function of the areal density [4]. At low densities, (cyclic) exchange processes up to six bodies are important, leading to a strong quantum frustration. The ground state is a *spin liquid* [5], realized in the commensurate second layer [6], [7, 8, 9, 10]. When this solid ^3He second layer densifies, three body exchange finally dominates and the system becomes an almost perfect 2D Heisenberg ferromagnet [11, 12, 13] with exchange J in the mK range (ferromagnetic region). The solid then forms a triangular Bravais lattice incommensurate with the underlying layer.

^4He atoms are non magnetic and, due to their larger mass they adsorb preferentially in strong binding sites. Adding controlled amounts of ^4He allows to replace ^3He atoms from the first adsorbed layer, to remove atoms trapped in deep potential defects, and to *control the sizes of the ferromagnetic domains*. Indeed close to the so-called *ferromagnetic anomaly* seen in pure 2D- ^3He , adding ^4He replaces ^3He atoms of the second layer and pushes them into the fluid overlayer, without changing notably the density. We thus demonstrated our ability to create *ferromagnetic nano-clusters* [14] using 2D- ^3He .

On 2D ferromagnetic ^3He clusters we performed continuous wave Nuclear Magnetic Resonance (cw-NMR) measurements, down to about $100 \mu\text{K}$. The most relevant experimental details are given in E. Collin *et al.* [14]. The experimental cell was mounted on a demagnetization

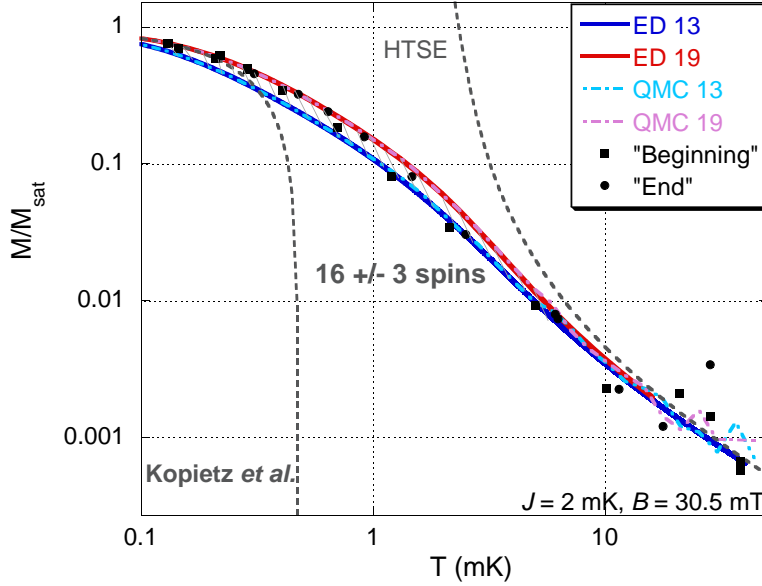


Figure 1. Magnetization of 2D Heisenberg ferromagnetic clusters (at the beginning \blacksquare , and at the end \bullet of the ferromagnetic region). The full and dot-dashed lines are ED and QMC calculations respectively (in perfect agreement), with the two colors distinguishing the sizes N (13 and 19 spins). The dashed lines are at high temperature the HTSE [16], and at low temperatures the Kopietz *et al.* expression [17].

cryostat, and temperature was recorded with a pulsed NMR platinum thermometer at low temperatures. At high temperatures, a calibrated carbon resistor was used. Further experimental details are given elsewhere [15].

2. Experiment compared to theory

The experiments are realized in low magnetic fields (30.5 mT), covering more than 3 orders of magnitude in temperature (from $T \ll J$ to $T \gg J$).

At high temperatures, the theoretical tool which was used up to now to fit the data is High Temperature Series Expansions (HTSE) [16]. Although they permit an extraction of a very good estimation of J in standard-size (thousands of atoms) experiments, they are not suited for small clusters. Indeed, the deviation of the fit value of J as a function of the size N seen in E. Collin *et al.* [14] is questionable, and certainly due to the fit procedure.

At low temperatures, the tool available up to now was an analytical formula proposed by Kopietz *et al.* [17], and foreseen by Godfrin *et al.* [12]. This expression appears to be obtained under assumptions which *are not* compatible with a quantitative analysis of the data.

In the intermediate temperature range $T \approx J$, the situation is even worse: the exponential growth of the zero-field susceptibility with decreasing temperature [18] has no analytical expression at finite fields and sizes.

We report in the present article a *direct comparison* between a finite size experiment and *exact theoretical tools* available nowadays for the whole temperature range: Exact Diagonalization (ED) and Quantum Monte-Carlo simulation (QMC).

In Figure 1 we present the measured magnetization of ^3He clusters obtained at the beginning of the ferromagnetic region (\blacksquare), and at the end (\bullet), the ^3He atoms removed by ^4He being continuously pushed in the fluid overlayer [14]. The standard mathematical tools are shown with dashed lines: a J of 2 mK was used for both high and low temperature expressions, with

a cluster size N of 19 spins for the low temperature expression (and a 30.5 mT field). The colored lines (thick and dot-dashed respectively) are ED and QMC, and are undistinguishable within the adopted scale (for $T < 10$ mK). Both are obtained with the Heisenberg Hamiltonian in the presence of a magnetic field:

$$\mathcal{H} = -2Jk_B \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j - g_N \mu_N B_z \cdot \sum_{i=1}^N S_{z,i}, \quad (1)$$

where $\langle i, j \rangle$ means a summation over nearest neighbors within the N spins of the cluster. $S_{\lambda,i}$ with $\lambda = x, y, z$ is measured in units of \hbar , and μ_N , g_N are the nuclear magnetic moment and the ^3He nuclear Landé factor, respectively. The (superimposed) lower curves in Figure 1 are obtained for a 13 spin hexagonal cluster (blue), and the (superimposed) upper curves for 19 spins (red). For all of them, the exchange was 2 mK and the field 30.51 mT. The ED simulations were performed on a cluster of PC, using the lapack library [19]. Taking into account the largest abelian subgroup of the group of symmetry of the cluster (i.e. $2\pi/6$ rotations), the Hamiltonian is split into 6 diagonal blocks, each of it being extremely sparse. The QMC simulations were run on a PC using the ALPS libraries [20].

Within the shaded region, the agreement between theory and experiment is excellent, leading to a mean cluster size of 16 ± 3 . The small discrepancy is believed to be due to the distribution of sizes actually present in the sample, and to their exact shapes. Note however that *the size is the only free parameter*, the exchange being known for this areal density and the field being measured.

3. Conclusion

In this article we presented nuclear magnetization data measured on 2D- ^3He nano-clusters, and compared them in the whole experimental temperature range to exact theoretical calculations (ED and QMC) of the magnetization of Heisenberg ferromagnets of only a few spins. The agreement is excellent, with as the only fitting parameter the size (and exact shape) of the clusters.

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