Very low frequency SQUID NMR measurements of two dimensional solid ^{3}He films

A Casey, C Lusher, B Cowan, J Saunders [1]

Dept. of Physics, Royal Holloway University of London, Egham, Surrey, TW20 0EX, U.K.

Abstract

Pulsed NMR measurements have been performed on ^{3}He films adsorbed on an exfoliated graphite sample of area 2m^2. The spectrometer is constructed using a DC SQUID amplifier, with additional positive feedback, and bandwidth 3.4 MHz. The input circuit is a superconducting flux transformer and so is intrinsically broadband. The static magnetic field is provided by a superconducting solenoid enclosed in a niobium shield. We report measurements at the "ferromagnetic anomaly" at frequencies below 50 kHz. A narrow ^{13}C line is also observed, which can be used to directly measure the temperature of the substrate.

Key words:
helium films; SQUID NMR; low dimensional magnetism

There is considerable interest in the study of solid ^{3}He films adsorbed on graphite as models for two dimensional magnetism. The second layer has been extensively studied experimentally; it is magnetically frustrated because of competing cyclic ring exchanges. This frustration, and the magnetic ground state, can be tuned by varying the total coverage. In the absence of a fluid overlayer, the ground state is believed to be a quantum spin liquid, while at coverages near 24 nm^{-2}, in the presence of a fluid overlayer, the film has ferromagnetic exchange, with an effective exchange constant $J \sim 2$ mK. Here the magnetism is reasonably well described by the Heisenberg nearest neighbour model, and in this case a ferromagnetically ordered state is expected at $T = 0$. According to the Mermin-Wagner theorem, in two dimensions this order is destroyed at finite temperatures by low frequency spin waves. However earlier studies at very low fields of the ferromagnetic solid boundary layer on graphite in contact bulk liquid, where the effective exchange constant $J \sim 0.1$ - 0.2 mK, were interpreted as evidence of spontaneous order at finite temperatures. Low field NMR studies of the second layer of ^{3}He on graphite, where the ferromagnetic exchange constants can be much greater, have also been recently reported and interpreted as evidence for spontaneous magnetization at finite temperature [1].

For our investigation of these systems over a wide range of magnetic field we have developed a pulsed NMR spectrometer, which uses a DC SQUID with additional positive feedback as a first stage amplifier. An important feature of our spectrometer is the relatively large bandwidth, 3.4 MHz in this case, which is achieved by a direct coupling of the SQUID to the room temperature amplifier, and by operating the SQUID in flux locked loop mode using the direct offset integration technique [2]. The large bandwidth is important in studies of samples with ferromagnetic exchange, since at low temperatures NMR signals are broad due to the demagnetizing field arising from high sample polarisation. In addition the spectrometers high slew rate provides a wide dynamic range, important since the signal size ranges over a factor of more than 10^4.

The graphite substrate for this cell consists of 0.125 mm sheets of exfoliated graphite with 0.025 mm thick silver foil diffusion bonded to each side. The sample

[1] Corresponding author. E-mail: j.saunders@rhul.ac.uk

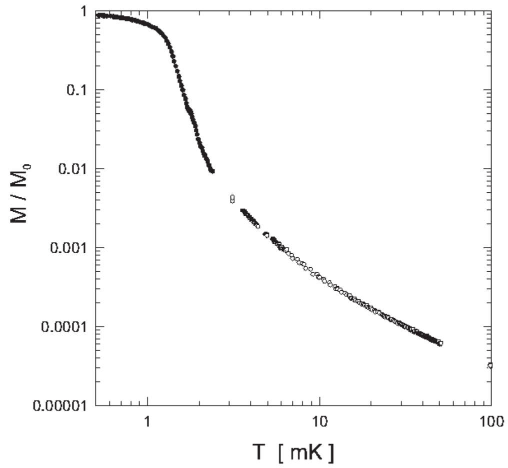

Fig. 1. Reduced magnetization as a function of temperature

consists of a 5 mm cube of 16 such graphite/silver sandwiches, separated by Kapton foil. The total surface area is 1.94 m^2, based on a ^{3}He vapour pressure isotherm at 4.2K, and taking the first layer density at promotion as 10.9 nm^{-2}. The substrate is sealed within a stycast 1266 chamber onto which is wound a solenoidal receiver coil, which forms part of a superconducting flux transformer with the input coil of the SQUID. The saddle shaped receiver coil, wound on a Macor former, is mounted on a platform at mixing chamber temperature. The static magnetic field is produced by a small superconducting solenoid, also mounted on this platform. It is equipped with a persistent switch; an overlapping niobium shield inside the brass former eliminates transient signals from eddy currents in the magnet former.

The temperature dependence of the sample magnetization, at an NMR frequency of 50 kHz, expressed as a fraction of the saturation magnetization, is shown in Fig 1. A series of NMR signals, the Fourier transform of the free induction decay, is displayed in Fig. 2. Below 2 mK a tipping angle of order 1° is used. The magnetization is determined by a numerical integration of the area under the Fourier transform. $M/M_0 = (M/C)(h\nu_0/k_B T)$, where ν_0 is the Larmor frequency and C is the Curie constant for the second layer spins. It can be seen that the magnetization reaches 90% of its saturation value. The large frequency shifts arise from dipolar shifts due to the large sample polarisation, which are negative in this static field orientation.

In this static field we can also probe the substrate temperature by ^{13}C NMR, illustrating the power of the broadband SQUID NMR technique. This simply involves setting the frequency of pulses to 16.5 kHz, the ^{13}C Larmor frequency. The narrow ^{13}C signal is compared with the ^{3}He signal from the paramagnetic first layer at a temperature of 0.9 mK in Fig 3. These

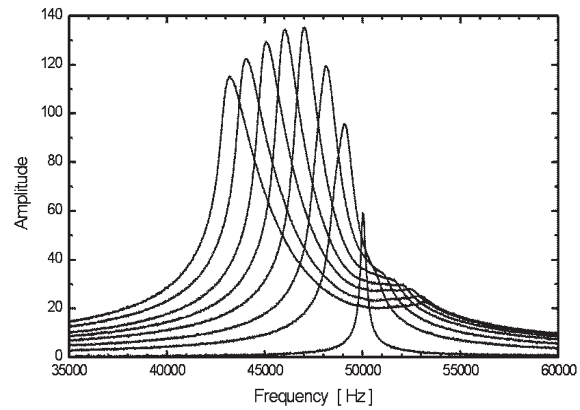

Fig. 2. NMR signals at 1.81, 1.41, 1.27, 1.09, 0.95, 0.81, 0.66, 0.55 mK, showing dipolar frequency shift

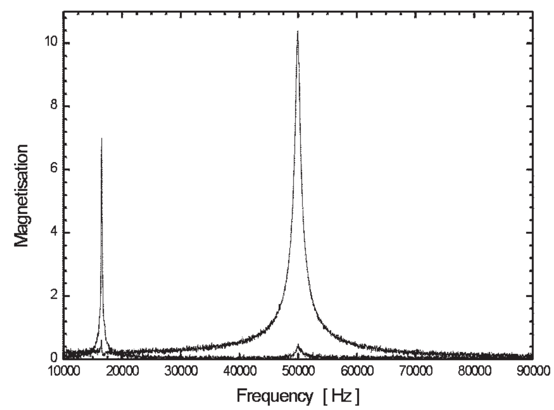

Fig. 3. Carbon-13 and helium-3 signals

are for comparable tipping angles (a 5 cycle pulse for ^{13}C and a 16 cycle pulse for ^{3}He). The ^{13}C line has $T_2^* = 2.3$ ms.

Acknowledgements

This work is part of a collaboration with D Drung and T Schurig, PTB, Berlin. We thank EPSRC (U.K) and the Royal Society for their support.

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