A FREQUENCY DEPENDENCE STUDY OF He-3
ADSORBED ON EXFOLIATED GRAPHITE

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ABSTRACT

The investigation of ^{3}He adsorbed on exfoliated graphite has been carried out using the conventional pulsed Nuclear Magnetic Resonance (NMR). The measurements were done at coverage of 0.6, 0.605 and 0.61 monolayer at the vicinity of the registered phase. The spin-spin relaxation time T_2 and the spin-lattice relaxation time T_1 were determined as a function of frequency and substrate orientation as well. The frequency expands from 1.3 to 8.3 MHz while the angle of orientation extends from zero to $\pi/2$.

All data were taken at 1.2 K where the spins exhibit quantum motion. It was found that T_2^{-1} depends linearly on Larmor frequency while T_1^{-1} obeys a logarithmic law with frequency. The anisotropy of T_1 was found to be weaker than that of T_2. These results give evidence that the relaxation is not only dipolar, but it is also due to the motion in the graphite local fields.

INTRODUCTION

^{3}He adsorbed on graphite is considered as an excellent candidate to study the physosorption phenomena. This implies a two dimensional system which consists of two substances; the adsorbate which is held to the surface of the adsorber. The adsorption happens when the adsorbate approaches the surface of the adsorber (substrate) and experiences an attractive Van-der-Waals potential. The result is that there will be a big chance for the adsorbate to be trapped on the surface of the substrate (1). In the present investigation, the adsorbate is the Helium atoms while the adsorber is the grafoil substrate.

Grafoil is a special type of graphite which has been exfoliated in a strongly oxidizing medium then rinsed and rapidly heated (2). The expanded particles are pressed together and rolled into binderless flexible sheets which have big surface area. These sheets are composed of graphite crystallites in the form of platelet. The length of the exposed basal plane is reported to be of the order of 100Å While the specific area of grafoil is about 20 m²/gm (3).

The phase diagram of the Helium-grafoil system was first constructed by Bretz et al, in 1973 (2), from the heat capacity measurements. The phase of interest in this work is the registered phase. It was found (4) to be

in the neighborhood of 0.6 monolayer at temperature less than 3 K. This phase is characterized by its regular adatoms arrangement with respect to the substrate lattice. The Helium atoms occupy one third the sites of the grafoil hexagon and form a triangle lattice structure of spacing of 4.2Å (5). Hence the substrate structure affects the behavior of the Helium film.

The NMR investigation of the spin-spin relaxation time T_2 and spin-lattice relaxation time T_1 of ^{3}He adsorbed on grafoil at the registered phase was of interest to a number of scientists. Among these; Richards and his coworkers who worked at 1 MHz (6), Sato and Sugawara who worked at 1 and 10 MHz (7) and Cowan who worked at a range of frequency extending from 1 to 10 MHz (8). The work presented here is a continuation of this investigation. The NMR has been employed to study the mobility of the spins of ^{3}He adsorbed on grafoil at the registered phase. This was done by measuring T_2 and T_1 as a function of Larmor frequency and substrate orientation at constant coverage and temperature as will be described shortly.

EXPERIMENTAL

A conventional dilution ^{4}He refrigerator was used.

The master oscillator used is 5600 Rockland frequency synthesizer with very good stability and low noise. The minimum frequency is 0.1 MHz and the maximum is 160 MHz. Hence it was easy to change Larmor frequency to the desired value over the range of interest.

To monitor the change of relaxation times with the orientation of the substrate at different angles, a rotating mechanism had to be constructed. It was important to rotate the sample chamber without opening the cryostat. Hence a home made rotator was constructed. To indicate the substrate orientation, a disc of paxolin printed circuit board was fitted to the end of sample chamber. This board is etched to leave a set of radially distributed gold plated copper pads electrically connected. Two metal film resistors were fixed to the board; a 1 kΩ resistor is fixed before the zero degree while a resistor of 500 Ω was fixed after the location of the 90° angle to simplify the detection. Between these two values, angles were varied in steps of 9 degrees by monitoring the conducting and the following non conducting position using an Ohm-meter. Hence, a precise orientation angle was obtained by rotating anticlockwise starting from 90-degree position.

All the work has been carried out at the same temperature of 1.2 K. The coverage were chosen very close to perfect registry as it thought to be at 0.605 monolayer (9), hence present work was done at 0.6, 0.605 and 0.61 monolayer.

The measurements were classified as follows:

a) The spin-spin relaxation time and spin-lattice relaxation time were determined at frequencies of 1.3, 2.6, 3.9, 5.1, 6.2, 7.5 and 8.3 MHz. The angle of orientation was fixed at 90° at these measurements.

b) T_2 and T_1 were measured as functions of the substrate orientation angle which is the angle between the normal to the substrate plane and the magnetic field. The substrate orientation changes from 0° to 90 in steps of 9°. These data were taken at two frequencies; 2.6 and 5.1 MHz.

To measure the spin-spin relaxation time, the $\pi/2-\tau-\pi$ sequence was used, while the $\pi/2-\tau-\pi/2$ sequence was used to measure the spin-lattice relaxation time.

RESULTS AND DISCUSSION

For the system of He-3 adsorbed on grafoil, the most effective contribution to T_2 would come from different things (10). Mainly; the variation in the diamagnetism of the grafoil, the paramagnetic impurities in or adsorbed on the grafoil and the dipolar field created by other ^{3}He spins. While T_1 mechanism is provided by modulation of the He-3 nuclear dipolar field due to the motion of vacancies or adatoms. Changes in the mobility of the helium atoms would show up as changes in T_2 and T_1 for any of the previous contributions.

The key factor which enables one to relate the relaxation times to the atomic motion is the form of the dipolar autocorrelation function $G_n(t)$ or its Fourier transform, the spectral densities $J_n(\omega)$. The expressions for the reduced spatial correlation functions which describe both short and long time behavior were established by Cowan (8) to be:

$$\frac{g_0(t)}{g(0)} = \frac{P_0}{[1+a_0^2 t^2/\tau^2]^{1/2}} + \frac{Q_0}{[1+b_0^2 t^2/\tau^2]}$$

$$\frac{g_2(t)}{g(0)} = \frac{P_2}{[1+a_2^2 t^2/\tau^2]}$$

where

$P_0 = 0.524$	$a_0 = 0.262$
$Q_0 = 0.476$	$b_0 = 0.928$
$P_2 = 1.0$	$a_2 = 0.944$

and τ is the correlation time which measures the speed of the motion which could be determined by connecting the experimental data with the theoretical assumptions, bearing in mind, according to Cowan (11), that T_1 is given by the following expression;

$$\frac{1}{T_1} = J_1(\omega_0) + 4J_2(2\omega_0)$$

where J_n is the spectral density and ω_0 is Larmor frequency.

Figure 1. Spin-spin relaxation rate as a function of the Larmor frequency at 1.2 K and 90 degree.

monitor the spins motion and showed that it is not purely dipolar, but it is also a result of the motion in the grafoil local fields.
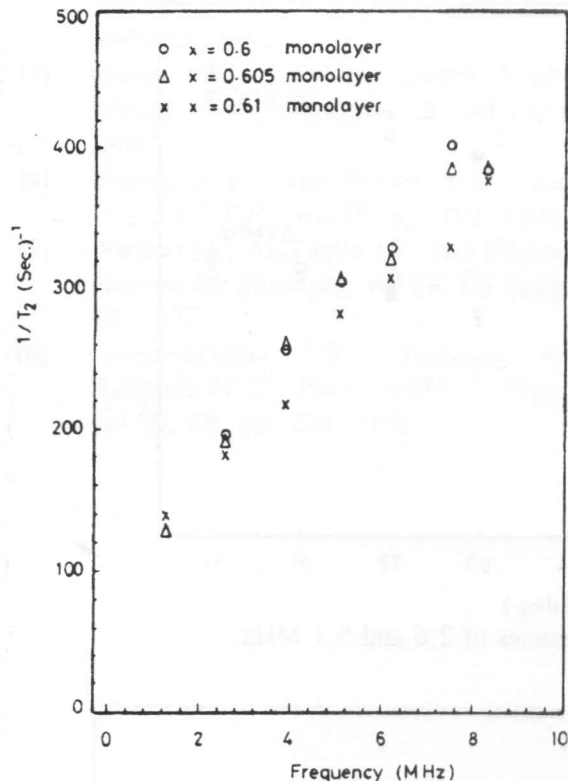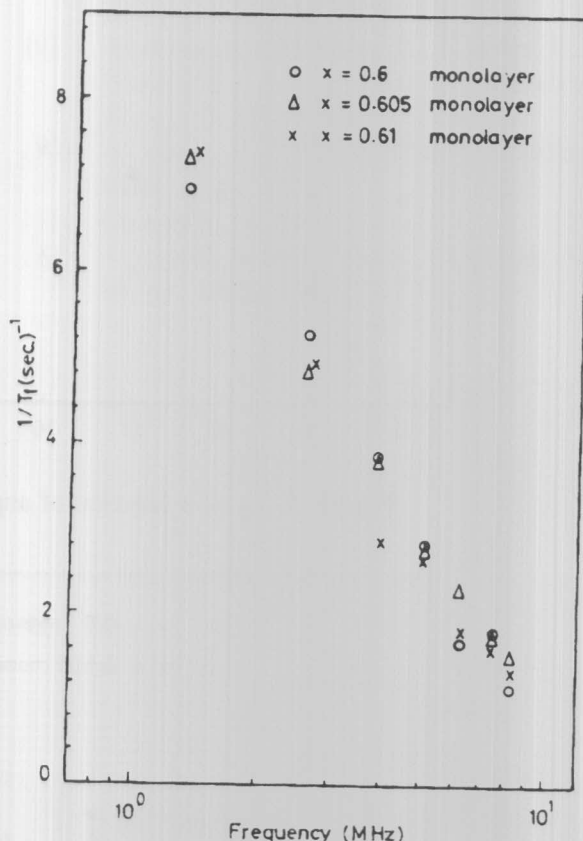

Figure 2. Spin-lattice relaxation rate as a function of the Larmor frequency at 1.2 K and 90 degree.

Figures (1) and (2) display the experimental data for T_2 and T_1 as functions of the Larmor frequency at coverage range of 0.6, 0.605 and 0.61 monolayer. Temperature was 1.2 K and the angle of orientation was 90°. The frequency range extended from 1.3 to 8.3 MHz. From Figure (1), we notice that T_2 is of the order of milli seconds and $1/T_2$ changes linearly with frequency obeying the following relation (10);

$$\frac{1}{T_2} = A + B\omega_0$$

where the frequency independent part, A, is clearly representing the dipolar part of the relaxation, while B is the relaxation due to the motion in the grafoil local fields which is not fully understood yet. On the other hand, Figure (2) shows the logarithmic dependence of $1/T_1$ on Larmor frequency, this as was predicted by Cowan (11) and Avogadro and Villa (12). However, the frequency dependence of both relaxation times

For a planar system, the NMR relaxation times depend on the orientation of the surface with respect to the applied static magnetic field (11). This is believed to be due to the anisotropy of the internuclear dipolar interactions. Figures (3), (4) demonstrate the variation of T_2 in milli seconds and T_1 in seconds with the change of the substrate orientation. T_2 and T_1 were determined at two frequencies; 2.6 and 5.1 MHz at coverage of 0.6 and 0.605 monolayer. The change of T_1 is weaker than that of T_2 for the change of angles from zero through 90°. Both relaxation times show strong frequency dependence while there was no appreciable change with coverage for different orientation. This is a further evidence that the relaxation is affected by the grafoil demagnetizing local fields.
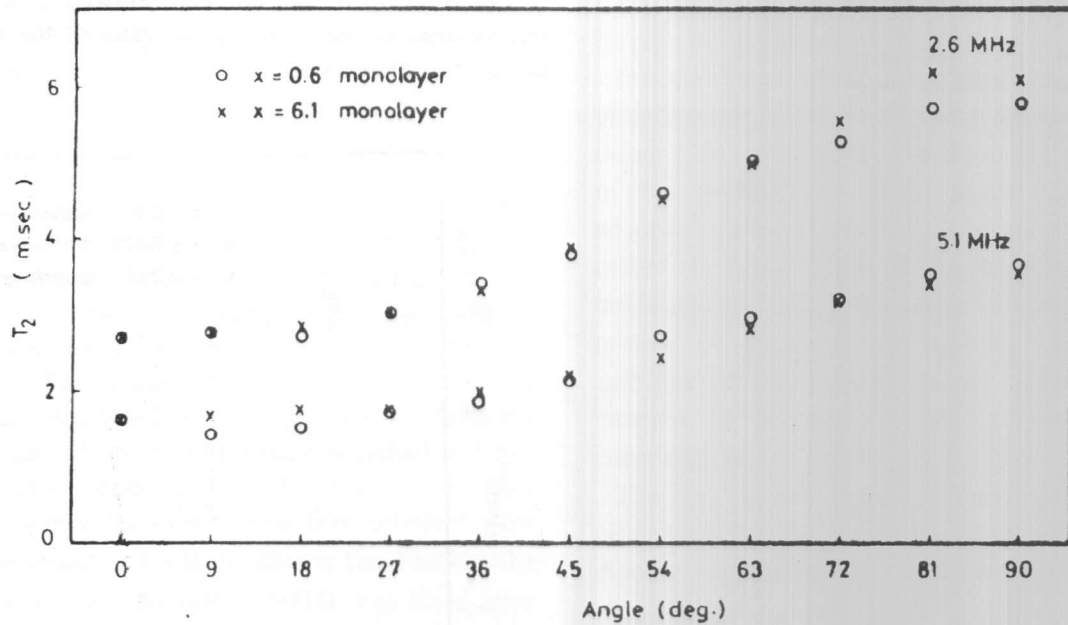
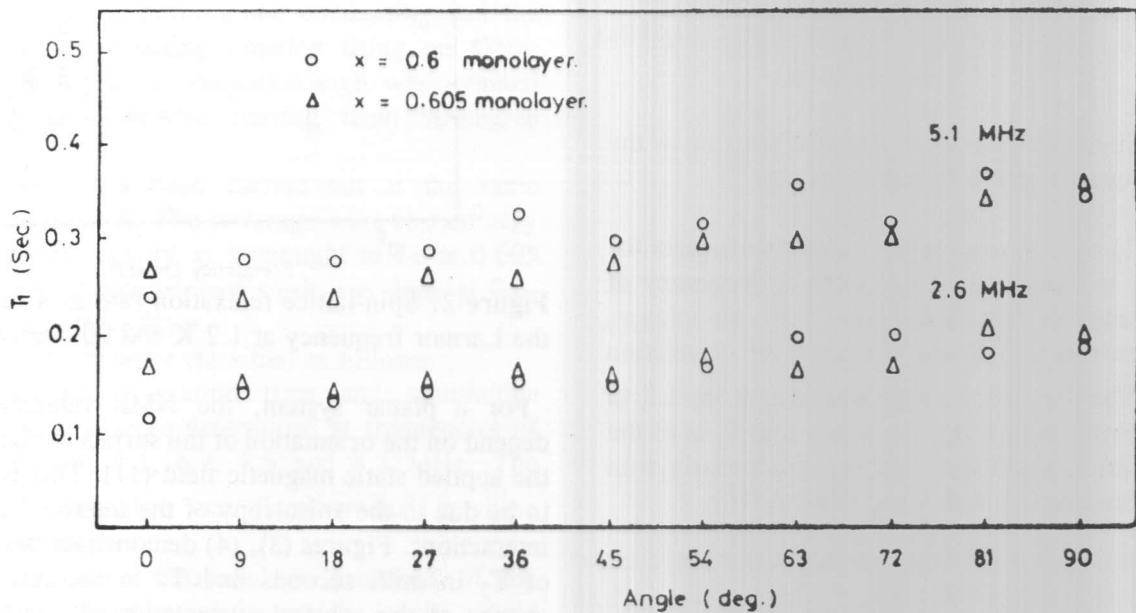**Figure 3**. T_2 as a function of angle at frequencies of 2.6 and 5.1 MHz.

Figure 4. T_1 as a function of angle at frequencies of 2.6 and 5.1 MHz.

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