ACTIVATION ENERGY OF ^{3}HE ADSORBED ON GRAFOIL AT REGISTRY

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ABSTRACT

The pulsed Nuclear Magnetic Resonance (NMR) has been employed to study the properties of ^{3}He adsorbed on grafoil. The work was done at 2.6 and 5.1 MHz, while the substrate orientation was 90 deg. The spin-lattice relaxation time T_1 and the spin-spin relaxation time T_2 were measured as a function of coverage at temperature of 1.2 K. The activation temperature was determined from T_1 and T_2 temperature dependence for a numher of coverage at the perfect registry. At this phase; the Helium atoms are regularly distributed on the grafoil mesh was recorded. The activation temperature was found to have its maximum value at perfect registry which indicates the slowing down of the motion.

INTRODUCTION

During the last two decades [1-4], great interest has been given to the Physics of low dimensional system. ^{3}He adsorbed on grafoil [5] is one of the most attractive two dimensional systems, especially in submonolayer films where a variety of Phases have been found by specific heat measurements [1,3]. These phases are mainly the two dimensional solid, the registered lattice gas and the two dimensional fluid. A series of strong heat capacity peaks was observed around 3 K. These peaks were interpreted [1] as a second order phase transition from disordered two dimensional gas phase to an ordered lattice gas in registry with grafoil structure. The registered phase is $\sqrt{3} \times \sqrt{3}$ epitaxial system where the ^{3}He atoms are in registry with the substrate structure. In such a case, every helium atom occupies one out of each three sites of grafoil hexagon to form a triangular lattice structure with spacing 4.2Å. This has been detected by neutron scattering experiments [6]. Thus the substrate plays an important role in the spins motion. Also one of the frequency dependence relaxation mechanisms is due to the motion in the grafoil local fields.

The aim of the present study is to investigate the coverage and the temperature dependence of both relaxation times in the neighborhood of the perfect registry.

EXPERIMENTAL

Measurements of the spin-lattice relaxation time T_1 and the spin-spin relaxation time T_2 were carried out using the pulse NMR technique. A home made broad band spectrometer, as explained by Cowan et al in reference [7], was used to cover a wide range of measurements between 1 and 10 MHz. The master oscillator used is 5600 Rockland synthesizer with excellent stability and low noise. The minimum frequency which could be achieved is 0.1 MHz and the maximum one is 160 MHz. Two Larmor frequencies were chosen to perform the work; 2.6 and 5.1 MHz. The measurements were classified into two sets:

a- T_1 and T_2 were determined as a function of coverage x at constant temperature $T=1.2$ K, where the spins exhibit quantum motion and the relaxation times are independent of temperature.

b- The relaxation times T_1 and T_2 were measured as a function of temperature in a range extends between 1 and 4.2 K. These data were taken at fraction of monolayer between $x=0.57$ and $x=0.625$ in steps of 0.005 monolayer.

In these measurements the angle of orientation between the external magnetic field and the normal to the substrate was taken to be $\pi/2$. The saturation recovery method was used to perform the data for T_1. While the spin echo technique was applied for measurements of T_2 [8].

RESULTS AND DISCUSSIONS

The change of the spin-lattice relaxation time as a function of coverage at temperature of 1.2 K and two Larmor frequencies 2.6 and 5.1 MHz is illustrated in Figure (1). It is noticed that at low coverage T_1 decreases slowly with the increase of x. Above $x=0.57$ monolayer the decrease of T_1 becomes steeper to have its minimum value at perfect registry, $x \leq 0.61$ of monolayer.

An increase of T_1 is observed for $x > 0.61$ which becomes almost coverage independent for higher values of x. The behavior is similar at the two Larmor frequencies although the data shows that the position and the depth of the dip is slightly frequency dependence.

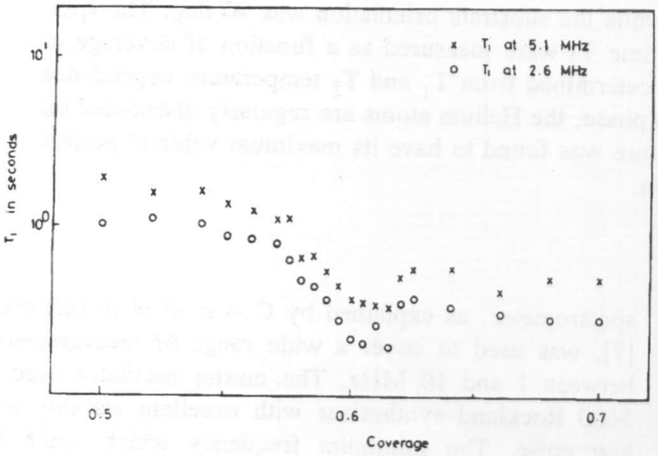

Figure 1. The variation of T_1 as a function of coverage at T = 1.2 K.

On the other hand, a display of the variation of the spin-spin relaxation time with coverage is shown in Figure (2) under the same conditions of temperature and frequency as mentioned above. A similar behavior as that of T_1 is repeated with the exception that instead of the sharp minimum; there is only a shallow one at perfect registry.

It has to be mentioned that T_1 drawn in units of seconds while T_2 is in m.seconds. The existence of the minima in both T_1 and T_2 around perfect registry is attributed to the slowing down of the spins motion at registry. The shallow minima observed in T_2 compared to T_1 is interpreted as due to the contribution of the grafoil local fields towards the motion of the spins. A similar behavior was reported by Owers-Bradly et al [9] and Richards et al [10] at frequency of 1 MHz.

To estimate a value for the activation temperature (activation energy), the set (b) of data was performed. T_1 and T_2 were measured as a function of inverse temperature for Larmor frequencies of 2.6 and 5.1 MHz. These measurements were taken at a number of coverage from below the perfect registry; x=0.59; to above of it; x=0.62; in steps of 0.005 monolayer. Figures (3) and (4) demonstrate the temperature dependence of T_1 and T_2 at the most probable perfect registry coverage; namely x=0.6 and x=0.61 monolayer.

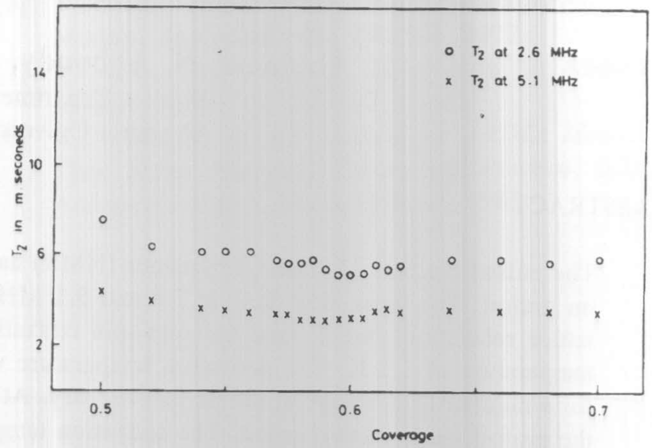

Figure 2. The variation of T_2 as a function of coverage at T = 1.2 K.

It is convenient to classify the data into three regions. Region I, above 3 K, there is only a slight change in both T_1 and T_2 with decreasing of the temperature. Region II, from 2-3 K, both relaxation times are decreasing exponentially with lowering the temperature. The decrease in T_1 is faster than that of T_2. Region III below 2 K, where the values of T_1 and T_2 are independent of temperature.

We assume that the activation energy of the atom is given by E_a [11], then:

$$E_a = E_f + E_m$$

where E_f is the formation energy of vacancies and E_m is the migration energy necessary to overcome the potential barrier due to repulsion between the Helium atoms.

In solid Helium, motion is thought to be quantum tunneling [11], then $E_m = 0$. If we assume the same behavior is true for two dimensions [12], then E_a will be given by E_f only. A direct result of vacancies is the modulation of the internuclear magnetic dipolar interactions [12]. Therefore, the mobility of vacancies implies the mobility of adatoms. At registry, the density of vacancies were found by Sato and Sugawara [12] to be 4.2 %.

However, the activation temperature of vacancies or adatoms was evaluated from the second region of Figures (3) and (4) where the data were found to obey the Arrhenius relation;

$$T_{1,2} = T_0 \exp(-E_a/kT)$$

where T_0 is a constant which could be evaluated from the graphs and k is Boltzman constant while E_a/k is the activation temperature.

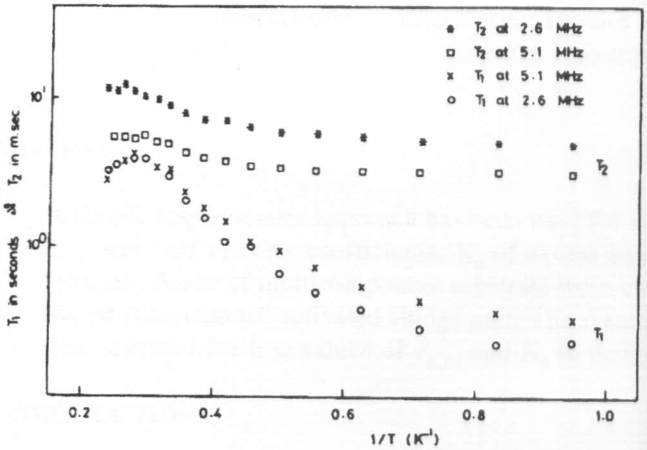

Figure 3. Temperature dependence of T_1 and T_2 at $x=0.6$ monolayer.

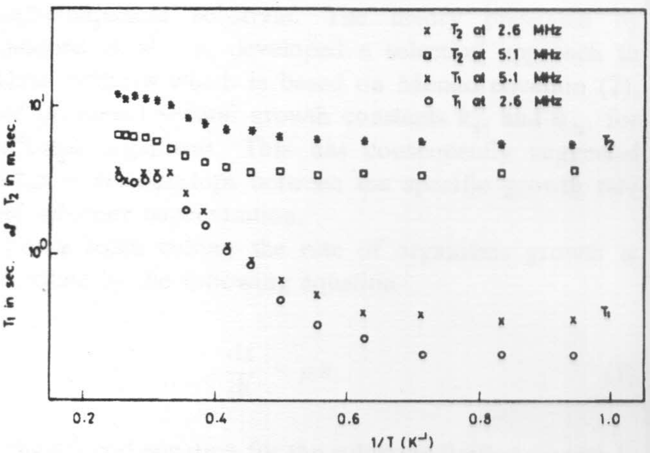

Figure 4. Temperature dependence of T_1 and T_2 at $x = 0.61$.

The activation temperature is performed as a function of coverage in Figure (5). The activation temperature has a maximum around perfect registry whether it is obtained from T_1 or T_2 data at both frequencies. The E_a/k obtained from T_1 shows a maximum value of 8.8 while that obtained from T_2 has a lower value of 4.8. This difference is attributed to the effect of the substrate local fields on T_2. The same discrepancy was claimed by Owers-Bradly [9] and Richards [10] for Larmor frequency of 1 MHz. On the other hand, Sato and Sugawara [12] did not see any change of E_a/k with increase of coverage for Larmor frequency of 10 MHz. This seems to be due to different treatment of the substrate and hence different surface nature which might affect the mobility of the Helium atoms.

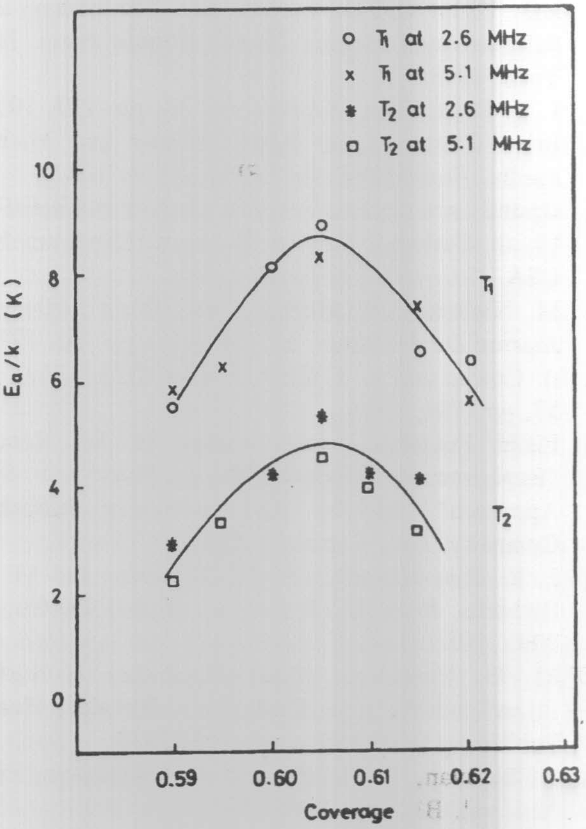

Figure 5. The change of the activation temperature with coverage as obtained from T_1 and T_2 temperature dependence.

As it can be seen from Figure (5), the perfect registry is most probable to be around $x=0.605$. Therefore one may suggest that a more reliable identification of the perfect registry can be obtained from the variation of activation temperature with coverage.

From the above analysis we may conclude that around perfect registry the motion of the spins slows down and has its lowest value at perfect registry. This motion is affected to some extent by the substrate local fields.

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