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J. Owers-Bradley, A. Thomson, M. Richards. N. M. R. MEASUREMENTS ON  $^3\text{He}$  ADSORBED ON GRAPHITE. Journal de Physique Colloques, 1978, 39 (C6), pp.C6-298-C6-299. 10.1051/jphyscol:19786131 . jpa-00217537

**HAL Id: jpa-00217537**

**<https://hal.science/jpa-00217537>**

Submitted on 4 Feb 2008

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N. M. R. MEASUREMENTS ON  $^3\text{He}$  ADSORBED ON GRAPHITE

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Résumé.- On rapporte les temps de relaxation R.M.N. de  $^3\text{He}$  adsorbé sur les substrats graphite, grafoil et UCAR ZYX. On observe les phases du 2D solide, 2D fluide et de l'enregistrement, avec anisotropie forte pour le deuxième substrat, ce qui indique un meilleur alignement des cristaux.

Abstract.- N.M.R. relaxation times are reported for  $^3\text{He}$  adsorbed on the graphite substrates, grafoil and UCAR ZYX. 2D solid, 2D fluid and registry phases are observed with marked anisotropy for the latter substrate indicating improved crystallite alignment.

Specific heat measurements for helium monolayers adsorbed on the surface of graphite crystals show [1] that with the exception of the registry phenomena, the behaviour of the adsorbate is dominated by the lateral interactions between adatoms. N.M.R. on  $^3\text{He}$  adatoms provides a useful probe for studying the dynamics of the monolayers and the observation of quantum tunnelling in the 2D solid [2] and of 2D gas like behaviour [3] in low coverage films has already been reported. The graphite substrate used in these studies has usually been grafoil [4], an exfoliated form of graphite offering a specific area of about  $20 \text{ m}^2/\text{g}$ . From the point of view of N.M.R., it suffers from having the adsorption planes neither all approximately parallel nor randomly oriented [5]. In addition, graphite is strongly diamagnetic and this leads [6] to N.M.R. relaxation processes that compete with the dipole-dipole process which usually dominates relaxation in  $^3\text{He}$ . A material with improved crystallite alignment, known as UCAR ZYX has recently become available [4] and a specific heat study [7] of the registry phase transition has shown a large peak associated with larger ordered domains in the registered phase. The lower specific area of about  $2.5 \text{ m}^2/\text{g}$  presents signal sensitivity problems for both specific heat and N.M.R. work.

In the work reported here pulsed N.M.R. was used, the substrates consisting of about 8 g of graphite in the form of sheets about 0.04 cm thick which had previously been heat treated at  $1000^\circ\text{C}$  under vacuum for 24 hours.

Figure 1 shows some measurements made at 1 MHz ( $\sim 300$  gauss) using the grafoil substrate. The data are taken by adding small amounts of gas at 1 K warming to 12 K to facilitate the spreading of gas throughout the sample, then slowly cooling back to 1 K.

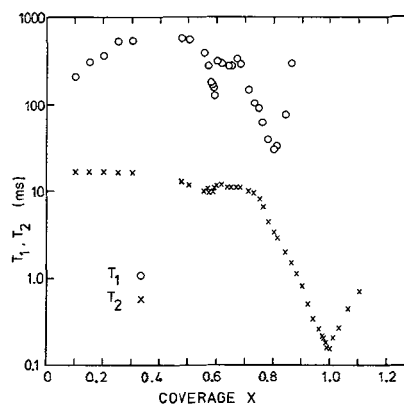


Fig. 1 :  $T_1$  and  $T_2$  vs. fractional monolayer coverage<sup>2</sup> for  $^3\text{He}$  on grafoil.

The ordinate X represents the fraction of a completed monolayer, completion being determined from a 4.2 K isotherm using the point B criterion [1]. The interesting features are :

- (i) A coverage independent  $T_2$  in the 2D fluid phase ( $X < 0.7$ ).
- (ii) A rapidly decreasing  $T_2$  as X increases in the 2D solid phase. This is associated with decreasing quantum tunnelling as the areal density increases.
- (iii) A very sharp minimum in  $T_2$  at monolayer completion followed by rapidly increasing  $T_2$ .

probably due to first-second layer tunnelling.

(iv) A  $T_1$  minimum in the 2D solid when  $\omega_0 \tau_c \approx 1$  ( $\omega_0/2\pi$  = Larmor frequency,  $\tau_c$  = correlation time for tunnelling motion).

(v) A sharp dip in  $T_1$  at a coverage corresponding to registry  $|1|$  of  $^3\text{He}$  atoms with the underlying substrate lattice.

The data were all taken with  $\beta$ , the angle between the normal to the grafoil sheets and the static field  $B_0$  equal to  $\pi/2$ . However similar data taken at  $\beta = 0$  and  $\pi/4$  display little anisotropy.

Figure 2 shows similar data taken at 1.5 MHz using the UCAR ZYX substrate at three values different of  $\beta$ .

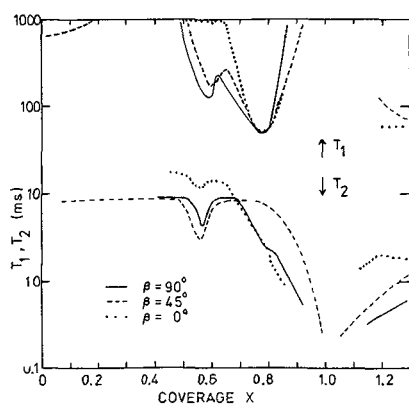


Fig. 2 :  $T_1$  and  $T_2$  vs. fractional monolayer coverage  $X$  for  $^3\text{He}$  on UCAR ZYX.

The new features that appear are :

(i) A marked anisotropy in both  $T_1$  and  $T_2$ . This occurs because  $|8|$  motion parallel to and perpendicular to the direction of  $B_0$  make very different contributions to the various matrix elements of the two spin dipole Hamiltonian which determines  $T_1$  and  $T_2$ . Even if relaxation is dominated by spatially varying fields of the graphite, this too will be anisotropic since the diamagnetic susceptibility of the graphite is itself highly anisotropic.

(ii) Anomalies in  $T_1$  and  $T_2$  at coverages corresponding to registry. The marked similarity of the data for the 2D solid (i.e.  $1 > X > 0.7$ ) on the two substrates (apart from the expected anisotropy) is further evidence for relaxation in this regime being primarily due to dipolar fields created by the adatoms themselves. The value of  $T_1$  at the

minimum (30 ms for grafoil at 1 MHz; 50 ms for UCAR ZYX at 1.5 MHz) is in good agreement with the expression  $(T_1)_{\min} \approx \omega_0/M_2$  where  $M_2$  is the second moment calculated by assuming a triangular 2D lattice with dipolar coupling. The failure of  $T_2$  to become approximately equal to  $T_1$  on the fast (low  $\tau_c$ ) side of the  $T_1$  minimum must represent the onset of some mechanism in this region contributing to  $T_2$  but not  $T_1$ . The mechanism for relaxation in the fluid phase ( $X < 0.7$ ) is not understood. Both  $T_1$  and  $T_2$  increase with temperature in this region which indicates that  $T_1$  is on the fast side of the minimum. That the graphite diamagnetism plays a part is suggested by the field dependence of  $T_2$ ,  $T_2$  decreasing as the field is increased.

The  $T_1$  and  $T_2$  anomalies observed in the registered phase suggest that N.M.R. will be a useful probe of that phase provided we can fully understand the mechanisms responsible for relaxation at low coverages.

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