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Quantum Tunneling in ^{3}He Monolayers

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Measurements are reported of NMR linewidth, $1/T_2$, for ^{3}He atoms adsorbed on Grafoil at 1 K for various quantities of gas adsorbed. The data show three well-defined regions corresponding to a two-dimensional fluid, a two-dimensional solid, and a region where a second layer of adsorbed atoms is forming. A sharp minimum in T_2 gives a precise indication of monolayer completion. A theory for tunneling in the two-dimensional solid is presented.

Specific-heat and isotherm data for samples of inert gases adsorbed on graphite give evidence of unusual homogeneity of the adatom-substrate potential.[1,2] Researchers from the University of Washington[2] have developed a phase diagram for helium adsorbed on Grafoil[3]—a convenient form of graphite having a large surface area per unit mass. NMR studies of adsorbed ^{3}He atoms are capable of supplementing thermal data by giving new information about the dynamical state of the adsorbate. Previous[4-7] NMR studies of ^{3}He adsorbed on graphite have shown linewidth changes and susceptibility data consistent with the proposed solid-liquid transition on the phase diagram, although there is disagreement over the evidence for the "registry" phase where ^{3}He atoms are located in positions determined by the carbon atoms in the substrate.

We report here detailed ^{3}He NMR linewidth measurements (i.e., $1/T_2$) taken at 1 K as a func-

tion of the amount of ^{3}He gas adsorbed. The substrate was made up of a multiple sandwich of layers of 0.015-in. Grafoil separated by insulating sheets of Mylar. The Grafoil was heated to 1000°C for 24 h *in vacuo* and transferred to a helium atmosphere where the Mylar sheets were inserted and the sandwich sealed into an epoxy chamber with a sealed filling capillary. The chamber was then transferred to the main apparatus and fixed inside an rf coil whose axis was in the plane of the Grafoil sheets. The superconducting magnet also had its axis in the plane of the Grafoil sheets. With use of pulsed NMR at frequencies between 0.3 and 2.0 MHz, no significant heating by the rf pulses was observed. The data were taken by progressively adding gas at 1 K and then warming the samples to 8 K for about 20 min followed by slow cooling back to 1 K to take data.

The results are shown in Fig. 1 where the transverse relaxation time T_2, measured at 1 MHz, is plotted against x, the fraction of a saturated monolayer. The amount of gas corresponding to $x=1$ is obtained from a 4.2-K isotherm using the point B criterion.[2] The data show three clear-cut regions: (a) $x<0.75$, where T_2 is nearly independent of x but shows a dependence on the applied magnetic field. (b) $0.75<x<0.98$, where T_2 is independent of the magnetic field but is strongly dependent on x so that $T_2=0.18\exp[(0.98-x)/0.06]$ msec. (c) $0.98<x<1.5$, where T_2 now increases as x increases so that $T_2-0.18=3.0(x-0.98)$ msec.

The following models are suggested for the different regions:

(a) Specific-heat data[2] suggest that at 1 K and $x\lesssim 0.7$, ^{3}He particles adsorbed on Grafoil move as a two-dimensional fluid. If the particles are moving with thermal velocities, then the correlation time for dipole-dipole modulation is $\sim 10^{-11}$ sec and this would lead to T_2 values of about 10^3 sec by this process. Since the observed values are $\sim 10^{-2}$ sec, it is likely that some other process is present. Local magnetic fields will be created by the bulk anisotropic susceptibility of the graphite if the crystal planes are not all parallel, and by any paramagnetic impurities such as iron and oxygen. The former process will contribute[8] a term to the linewidth proportional to H_0^2/D, where H_0 is the applied field and D the diffusion coefficient, by assuming that motional narrowing of these fields is occurring. The latter will contribute a field-independent term. We may get information about D for the two-dimensional (2D) fluid by comparing the field dependence of T_2 for these low-coverage samples with that for the sample chambers filled with liquid helium for which D is known. Using

$$D_{2D}=D_{3D}\left[\frac{\partial(1/T_2)}{\partial H_0^2}\right]_{3D}\left\{\left[\frac{\partial(1/T_2)}{\partial H_0^2}\right]_{2D}\right\}^{-1},$$

we find $D_{2D}=1.2\times10^{-4}$ cm^2 sec^{-1} at $x=0.3$. This is of the same order as the kinetic-theory value for a perfect 2D gas of this density and temperature, for which $D\approx v/8na\approx 3\times10^{-4}$, where v is the thermal velocity, a is the atomic radius, and n is the areal number density. (*Note added.*—In the neighborhood of $x=0.6$, specific-heat data[2] suggest that below 3 K the system is in a state which is ordered with respect to the substrate. In data taken since these reported here we have observed small anomalies in both T_1 and T_2 at the experimental coverage and temperature.)

(b) In this region, specific heat data[2] suggest that ^{3}He adsorbed on Grafoil behaves like a 2D solid. The rapid decrease in T_2 as the density increases is similar to that observed[9] in bulk solid ^{3}He where the quantum tunneling of atoms motionally narrows the dipolar fields, and the tunneling rate J_{3D} decreases rapidly as the density increases.

To deduce the tunneling rate in 2D, J_{2D}, from the T_2 data, we need to make the following assumptions:

(i) The second and fourth moments of the NMR line are calculated[10] for the 2D solid assuming a triangular lattice with the applied field on the plane of the lattice so that

$$M_2=4.94\hbar^2\gamma^4/R^6,\quad M_4=111.8J_{2D}^2\hbar^2\gamma^4/R^6,$$

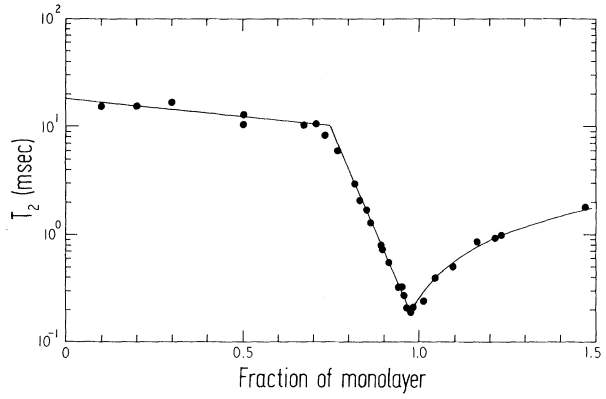

FIG. 1. Measurements of T_2, the NMR transverse relaxation time, for ^{3}He adsorbed on Grafoil at 1 K as a function of the fractional coverage x. The measuring frequency is 1 MHz, and $x=1$ corresponds to a complete monolayer as defined by a 4.2-K isotherm.

TABLE I. Values of the NMR linewidth, $1/T_2$, and the tunneling frequency, J_{2D}, for ^{3}He at various fractional coverages, x.

x	R (Å)	T_2 (msec)	$\lvert J_{2D}\rvert$ (10^5 sec^{-1})
0.770	3.70	6.0	7.74
0.820	3.58	3.0	4.72
0.830	3.56	2.1	3.25
0.853	3.51	1.7	3.02
0.862	3.49	1.3	2.38
0.892	3.43	0.78	1.56
0.896	3.42	0.73	1.57
0.916	3.39	0.55	1.20
0.943	3.34	0.33	0.787
0.954	3.33	0.32	0.777
0.960	3.31	0.27	0.680
0.968	3.29	0.21	0.548
0.976	3.28	0.20	0.532

where γ is the gyromagnetic ratio and R the nearest-neighbor lattice spacing.

(ii) The Gaussian approximation[11] for the correlation function for modulation of the dipolar fields can be used to obtain[10]

$$1/T_2 = 1.30 J_{2D}^{-1} \hbar^2 \gamma^4 / R^6$$

for the adiabatic linewidth. Since we observe that $T_1 \gg T_2$, the nonadiabatic contribution will be small.

(iii) $R = 3.24/\sqrt{x}$ Å. This follows from the assumption[12] that the areal density of atoms is 0.110 Å^{-2} at $x = 1$. The results of this interpretation are shown in Table I and Fig. 2, where data from the 3D phases are also given.

The theoretical curve for J_{2D} was obtained by methods established for quantum solids.[13-15] A correlated wave function is chosen to be

$$\Psi = \prod_i \varphi(\vec{r}_i - \vec{R}_i) \prod_{k<l} f(r_{kl}),$$

where

$$\varphi(\vec{r}_i - \vec{R}_i) \equiv \varphi_i(\vec{r}_i) = \left(\frac{A}{\pi}\right)^{1/2} \exp[-\tfrac{1}{2} A(\vec{r}_i - \vec{R}_i)^2]$$

and

$$f(r) = \exp\{-K[(\sigma/r)^{12} - (\sigma/r)^6]\}.$$

The energy is calculated using a cluster expansion[13] in which only the single-particle and pair terms are considered, and A and K are variational parameters. Values of A and K and the energy are given in Table II. Also given are the J_{2D} calculated by antisymmetrizing the wave function and by making use of a two-dimensional version of the surface-integral formulation of McMahan.[15]

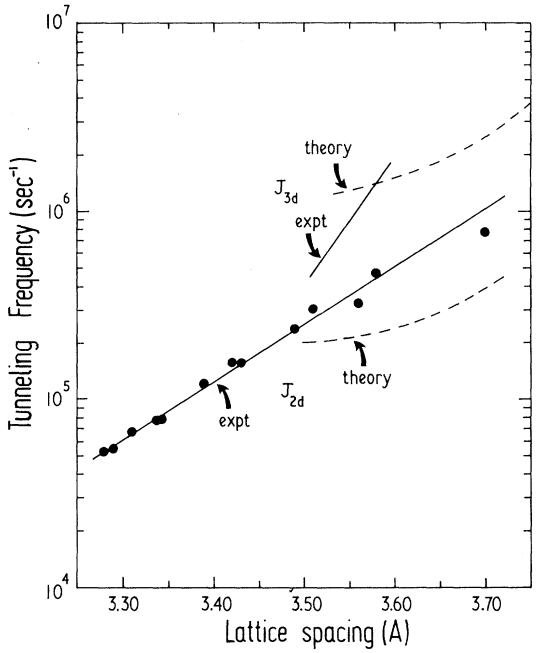

FIG. 2. Values of the quantum tunneling frequency for ^{3}He atoms in the hcp solid (J_{3D}) and adsorbed on Grafoil (J_{2D}) as a function of nearest-neighbor lattice spacing. The dashed lines represent calculated values from Ref. 14 (J_{3D}) and this work (J_{2D}). The full lines represent values based on experimental work from Ref. 16 (J_{3D}) and this work (J_{2D}).

The results are similar in two and three dimensions, the main differences being that J is smaller and varies less rapidly with atomic spacing in two dimensions. These differences are thought to be due to the way in which the motion of an individual ^{3}He atom in two dimensions is more confined by the six nearest neighbors in the plane than by the eight or twelve nearest neighbors in a bcc or hcp three-dimensional lattice. Below $R = 3.50$ Å the energy can no longer be computed, presumably because of the inadequacies in the truncated cluster expansion when the assumed

TABLE II. Calculated values of the optimal variational parameters, the energy, and the tunneling frequency.

R (Å)	A (Å^{-2})	$\ln K$	E (K)	J_{2D} (10^5 sec^{-1})
3.75	1.56	−0.78	10.25	−5.6
3.70	1.63	−0.79	11.05	−3.9
3.65	1.70	−0.79	11.94	−3.0
3.60	1.76	−0.79	12.95	−2.4
3.55	1.81	−0.79	14.08	−2.1
3.50	1.86	−0.79	15.36	−2.0

form of $f(r)$ is used. Precisely the same effect also occurs in the three-dimensional case.[13] The curvature of all the theoretical curves at low R may also be an artifact related to this impending breakdown.

(c) The sharp minimum in T_2 at $x \approx 0.98 \pm 0.01$ is particularly interesting. There is no other phenomenon in adsorbed helium which displays a sharp effect at the completion of a monolayer although the specific heat[2] passes through a shallow minimum at this coverage. The obvious interpretation is that, for $x > 0.98$, atoms begin to form a second layer and can then exchange with first-layer particles. This motion narrows the resonance line, leading to an increase in T_2. We are in the process of constructing a theoretical treatment of this effect.

It is interesting to note that the continual decrease in the value of T_2 on Grafoil up to a coverage of almost a completed monolayer is in marked contrast to the results obtained on other systems. Data on Vycor,[17] for instance, indicate that for submonolayer films T_2 increases as the coverage is increased and the completion of a monolayer is not indicated by any abrupt change in the behavior of the linewidth.

In conclusion the data reported seem to confirm the two-dimensional fluid-solid transition proposed, the areal density dependence of the tunneling rate is obtained experimentally and theoretically, and the existence of a well-defined saturated monolayer is demonstrated by a sharp minimum in T_2. The temperature dependence of T_2 and measurements of the spin-lattice relaxation time will be reported shortly.

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