Nuclear Magnetic Resonance of $^3$He Adsorbed Submonolayers on Grafoil and Argon-Coated Grafoil. I. Low Coverages*

S. G. Hegde and J. G. Daunt

Cryogenics Center, Physics Department, Stevens Institute of Technology, Hoboken, New Jersey

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Continuous-wave (cw) nuclear magnetic resonance measurements have been made at 5.5 MHz of $^3$He adsorbed on Grafoil and on argon (monolayer)-coated Grafoil at coverages $x < 0.40$ ($x = 1$ corresponds to one monolayer), in the temperature range 0.35–4.21 K. To determine the fractional coverages, the adsorption area of the Grafoil and the monolayer coverages were determined from observations of argon and $^3$He adsorption isotherms. The results indicate that the adsorbed $^3$He is highly mobile and behaves as a two-dimensional nonideal Fermi gas. Relative magnetic susceptibilities determined from the NMR lines indicate that the onset of Fermi degeneracy is delayed by spin-independent $^3$He–$^3$He interactions, as shown by the fit to the data of the quantum second virial coefficient expansion for the magnetic susceptibility. It is established that the residual (coverage-independent) linewidth is largely determined by spin-diffusion through local field gradients in voids in the Grafoil.

1. INTRODUCTION

The study of physisorbed states of gases, particularly $^3$He and $^4$He, has been the subject of considerable experimental and theoretical interest$^{1,2}$ in recent years. Adsorbed layers of $^3$He or $^4$He on a plane surface of substrate material are expected to exhibit the physical behavior of matter of reduced dimensionality. In order to investigate phase transitions in two dimensions, extensive thermodynamic studies have been made of adsorbed $^3$He and $^4$He systems on such substrates as Zeolite, Vycor porous glass, Grafoil$^\dagger$ (a commercially available form of exfoliated graphite), sintered copper, and many other materials.

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Although thermodynamic measurements are powerful tools in the study of phase transitions, they do not provide a time scale for atomic events, which occur more rapidly than time-averaged thermodynamic experiments can detect. NMR measurements, both pulsed and continuous-wave (cw), are powerful tools for the study of the dynamic state of the adsorbate, and, as such, complement the thermodynamic measurements.

Previous NMR measurements of adsorbed $^3$He have been done on various substrates: e.g., Zeolite,$^{3,4}$ Vycor porous glass,$^{5,6}$ graphitized carbon black,$^7$ and Grafoil,$^{8-10}$ and, where applicable, the results of these measurements will be discussed in this paper and in one to follow.

This study, a brief report of which has been previously published,$^{10-12}$ deals with the measurement of NMR linewidths and magnetic susceptibilities of $^3$He adsorbed on Grafoil and on argon-coated Grafoil and the results that derive from it. These substrates were chosen because of their known high homogeneity. A continuous-wave (cw) NMR technique at relatively low frequencies was employed, because the lower the frequency used, the fewer are the problems with spurious broadening due to the substrate and the fewer are the problems due to rf shielding, and because cw gives minimal rf heating of the system. In this paper we confine ourselves to the results obtained at low $^3$He coverages, $x \leq 0.40$ ($x = 1$ corresponds to one monolayer). At low coverages the adsorbed $^3$He is highly mobile and has a two-dimensional (2D) gas behavior, having at high temperatures (~4 K) a specific heat $C$ approaching the classical 2D gas value of $1k_B$/atom. Moreover, at temperatures below ~1 K, the specific heat of $^3$He on Grafoil does not appear to show any signs of condensation into a liquid phase.$^{13-17}$

2. EXPERIMENTAL ARRANGEMENTS

A continuous-wave (cw) NMR system was used, employing a Robinson oscillator$^{18,*}$ with a magnetic field modulation at 100 Hz of 0.01 G rms and a linear frequency sweep. The signal was detected with a phase-sensitive detector and the frequency sweep through resonance was done slowly (typically 10 Hz/sec). Where signal-to-noise was poor, signal averaging over up to ten sweeps was done using a Northern Scientific Model NS570 (Tracor Northern Inc., Middleton, Wisconsin) signal averager. The static magnetic field $H_0$ was maintained by a superconducting solenoid, with a 2.5-in.-diameter bore, immersed in liquid helium at 4.2 K. This solenoid had a measured inhomogeneity throughout the volume (3.7 cm$^3$) of the specimen less than 4 parts in $10^5$.

*We use a modified Robinson design developed and constructed by Mr. P. Kanepa, University of Florida, Gainesville, Florida.