Measurements of the Nuclear Spin-Spin Relaxation Times for Commensurate $^3$He-Ne Films Adsorbed on Hexagonal Boron Nitride

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Abstract.

Measurements of the $^3$He nuclear spin-spin relaxation time, $T_2$, have been carried out for commensurate layers of $^3$He-Ne mixtures adsorbed on hexagonal boron nitride for temperatures $0.2 < T < 10$ K. A temperature independent relaxation is observed at low temperatures and is interpreted in terms of the effective exchange frequencies for $^3$He particle exchange on the surface. The results show a strong dependence on the fraction of neon in the adsorbed layer. This variation is discussed in terms of a multiple spin exchange model for $^3$He in a monolayer. The contributions to $T_2$ from different components of the exchange, 2-spin exchange ($J_2$), 3-spin exchange ($J_3$), 4-spin exchange ($J_4$) and higher exchange permutations depend on the $^3$He coverage and thus permit the separation of the amplitudes of the different exchange rates, and in particular allow one to deduce the relative strengths of 2-atom and 3-atom exchange where other methods are sensitive only to the effective two-particle term $J_{\text{eff}} = J_2 - 2J_3$.

1. Introduction

Numerous studies, experimental[1, 2, 3, 4, 5, 6] and theoretical[7, 8, 9, 10, 11, 12, 13, 14], of the nuclear magnetic properties of solid helium three in both bulk and two-dimensional films have been successfully interpreted in terms of the multispin exchange model. In this model the spin exchange Hamiltonian is given by

$$H = -\hbar \sum_n (-)^n J_n P_n$$

where $J_n$ is the strength of the exchange for an $n$-particle cyclic permutation described by the spin permutation operator $P_n$. The most striking feature of the exchange, particularly in monolayer films, is that the $J_n$ are comparable in strength, and exchanges up to the sixth order are needed to obtain a good description of the experimental studies of $^3$He on graphite[4, 5, 8]. The experimental parameters used to infer values for the $J_n$ are: (i) the Curie constant $\Theta = 3J_{\text{eff}}^X$ with $J_{\text{eff}}^X = -(J_2-2J_3+3J_4-\frac{5}{8}J_6,...)$, and (ii) the nuclear heat capacity, $C^N = \frac{1}{2}(k_BT)^2J_{\text{eff}}^C$ with $J_{\text{eff}}^C = (J_2-2J_3+\frac{5}{8}J_4+...).$ Although relatively large values of $J_2$, $J_3$ and $J_4$ are predicted, the competing signs of $J_n$ result in small values for the susceptibility and $J_{\text{eff}}^C$. Experiments reported
to date have been sensitive only to the combination $J_2 - 2J_3$ and we sought a more direct means to
determine the relative strengths of $J_2$ and $J_3$. In order to do this we prepared samples of $^3$He and
neon (with a random distribution of neon) to reduce the number of three-particle permutations
(and higher orders) by dilution. Preliminary results have been previously reported[15] and
this study compares the results for three concentrations studied at low temperatures where
temperature independent exchange-induced relaxation dominates the relaxation rates.

2. Experimental considerations
Commercially available hexagonal boron nitride (BN)[16] was used for the studies and after
washing with methanol to remove impurities was baked at 400 C in high vacuum before loading
into the NMR cell. Micrograph studies showed that the BN consisted of thin platelets measuring
typical 100–200 microns across with a thickness of approximately 10–30 microns. The BN was
tightly compressed around a brush of copper wire that was part of a cold finger extending from a
dilution refrigerator. The gas samples were admitted as a hot gas via a heated capillary (∼12 K)
to a sample cell held at 1 K using $^4$He exchange gas exterior to the cell. This technique optimized
the random distribution of $^3$He and Ne on the cold surface and minimized clustering of the neon.
The nuclear spin-spin relaxation studies were carried out using pulsed NMR techniques at high
Larmor frequency (203 MHz) in order to maximize the signal to noise ratio and to simplify
the interpretation of the NMR results. The precise coverage was determined by measuring the
spin-spin relaxation time, $T_2$, as a function of gas admitted to the cell for a fixed temperature
(0.2 K). The results shown in Figure 1 exhibit a sharp minimum as a function of coverage with
the minimum identified as the completion of a commensurate monolayer.[15, 17] The variation
for a 33% mixture shows an appreciable broadening of the minimum (compared to that observed
for pure $^3$He[15]) as expected for a random distribution of the neon.

![Figure 1](image)

**Figure 1.** Variation of the spin-spin relaxation time ($T_2(x)$) with the fraction of coverage for
monolayers of $^3$He-Ne mixtures on boron nitride for fixed temperature (T=0.4 K) for a 50%
mixture of $^3$He-Ne.
3. Results
Two different temperature regions are observed for the relaxation times[15]. At low temperatures \((T < 0.5 \text{ K})\) the relaxation is independent of temperature which is the signature of exchange dominated relaxation, while at high temperature a strong increase of \(T_2\) is observed with increasing temperature and has been interpreted in terms of vacancy formation[15, 17]. The high temperature data is poorly described by the expected exponential variation, \(\exp(-\Phi/k_B T)\), for an activation energy \(\Phi\) and may be better described in terms of a 2D lattice gas melting[18]. The values of the low temperature relaxation are used for the analysis below.

4. Discussion
At high Larmor frequencies the transverse nuclear spin relaxation rate \(T_2^{-1}\) is given by

\[
T_2^{-1} = \frac{3}{2} G_0(0) = \left(\frac{3}{2} x M_{2p}\right)/J_{\text{NMR}}^{\text{eff}}
\]

where \(G_0(\omega)\) is the Fourier transform of the temporal autocorrelation function \(\langle Y_{20}^{jk}(t)Y_{20}^{jk}(0)\rangle\) for the spherical harmonic \(Y_{20}^{jk}\) associated with the orientation of the internuclear vector \(r_{jk}\).

\[ J_{\text{NMR}}^{\text{eff}} = 1.4\left[ J_2 - 2xJ_3 + \frac{3}{4}x^2J_4 - \frac{5}{8}J_5 + \ldots \right], \]

\(M_{2p}\) is the NMR second moment for pure \(^3\text{He}\) on BN, and \(x\) is the probability of occupation of a lattice site by a \(^3\text{He}\) atom. (Matsumoto et al.[19] have given an explicit calculation of the fourth NMR moment for bulk \(^3\text{He}\).)

![Figure 2. Observed dependence of the low temperature \((T < 0.5 \text{ K})\) nuclear spin-spin relaxation time, \(T_2(x)\), on the fraction \(x\) of \(^3\text{He}\) coverage for monolayers of \(^3\text{He}-\text{Ne}\) mixtures adsorbed on hexagonal boron nitride.](image)

![Figure 3. Comparison of the dependence of the normalized nuclear spin-spin relaxation time, \(xT_2(x)/T_2(1)\), on the fraction \(x\) of \(^3\text{He}\) coverage for monolayers of \(^3\text{He}-\text{Ne}\) on boron nitride with the variation calculated using the multispin exchange model described in the text.](image)

The observed variation of the low temperature values of \(T_2\) with \(^3\text{He}\) fraction are shown in Figure 2. (The dotted line is only a guide to the eye.) A distinct minimum is observed near \(x=65\%\) which is indicative of the competition between \(J_2\) and \(J_3\). In order to compare the observed values with theoretical predictions for the values of \(J_n\) we have plotted the normalized relaxation times \(xT_2(x)/T_2(1) = J_{\text{eff}}^{\text{NMR}}(x)/J_{\text{eff}}^{\text{NMR}}(1)\), shown by the solid line in Figure 3. For the calculated variation we used \(J_3 = 0.8J_2, J_4 = J_5 = 0.5J_2\) (comparable to the values of Ikegami et
al.[4]). The experimental results do not follow the sharp dip seen for the calculated values and this is attributed to the random nature of the occupation of the neon sites that would lead to a broadening of the site distribution functions and a smoothing of the theoretical calculation. The fit to the absolute values at \( x = 1 \) yields a value for \( J_2/2\pi = 1.1 \text{ MHz} \).

5. Conclusion
Measurements of the temperature independent transverse nuclear spin relaxation times observed at low temperatures for mixtures of \(^3\)He and neon on hexagonal boron nitride show an unusual dependence on the fraction of \(^3\)He in the monolayer. Instead of the conventional \( T_2(x) \sim 1/x \) expected for a classical system, a minimum is observed near \( x = 0.65 \) and is consistent with the multiple spin exchange model for two-dimensional \(^3\)He, the minimum being associated with the lowest order variation of \( T_2 \) with \(^3\)He fraction as determined by the effective exchange frequency for the NMR relaxation given by \( J_{\text{NMR}}^{\text{eff}} = |J_2 - 2xJ_3 + \ldots| \). The higher order terms are also significant in determining the absolute value of \( J_{\text{NMR}}^{\text{eff}} \).

A remarkable prediction of these studies is that for very dilute systems, \( x < 0.3 \), a strong increase in \( T_2 \) is expected with a predominant antiferromagnetic exchange that would exhibit the combined effects of frustration and disorder and could lead to the formation of a nuclear spin glass at very low temperatures.

6. References
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