Nuclear Spin-Spin Relaxation in ^{3}He-Ne Films

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Abstract

NMR measurements of the nuclear spin-spin relaxation times are reported for commensurate monolayers of ^{3}He and ^{3}He-Ne films on boron nitride for temperatures 0.1 <T< 3.5 K. The results are analyzed in terms of particle-particle exchange motions of the adsorbed ^{3}He atoms. The effective exchange rates were observed to change significantly on replacing a fraction of the helium atoms with relatively immobile neon atoms. This is understood if there is a significant 3-particle term in the exchange Hamiltonian that is of opposite sign to that of the 2-spin exchange term. Values of the vacancy formation energy for the monolayer coverage and the atom-vacancy exchange rate are also obtained from the experimental observations.

Key words: helium3; NMR; quantum tunneling

1. Introduction

Adsorbed films of ^{3}He on clean well-characterized substrates such as exfoliated graphite or hexagonal boron nitride represent one of the most ideal two-dimensional quantum systems. The atoms are weakly localized with respect to quantum zero-point motions, and as a result the atomic wave functions can overlap significantly, leading to a high probability of atom-atom exchange. The exchange is not limited to 2-spin exchange, and higher order cyclical 3-spin, 4-spin and higher order exchanges also occur.

The multi-spin exchange (MSE) Hamiltonian is given by

$$H = -\hbar \sum_n (-)^n J_n P_n \quad (1)$$

where J_n and P_n are the exchange frequency and permutation operator, respectively, for cyclic permutations of n atoms. The nuclear magnetism at very low temperatures is dominated by the MSE. In the 2D helium films there is a strong dependence on local density and on the particular geometry of the center of mass lattice structure[1], *i.e.* whether commensurate $\sqrt{3} \times \sqrt{3}$ or higher order structures at higher surface densities. The lowest coverages tend to be ferromagnetic because of the suppression of 2-spin exchange by the corrugation of the absorption potential. The behavior becomes more antiferromagnetic on approaching perfect $\sqrt{3} \times \sqrt{3}$ coverage, and ferromagnetic on completing the full monolayer coverage.

The observed temperature dependence of the nuclear spin susceptibility, $\chi^N = C/(T - \Theta)$, and the heat capacity, $C^N = \frac{9}{4}(J^C_{eff}/(k_B T)^2)$ can be understood in terms of the MSE model with $\Theta = 3J^{\chi}_{eff}$, where $J^{\chi}_{eff} = -(J_2 - 2J_3 + 3J_4 + ...)$ and $J^C_{eff} = J_2 - 2J_3 + 5J_4/2 + ...)$.

The purpose of this report is to show that the effective exchange frequencies can be determined from measurements of the NMR relaxation rates. NMR is sensitive to the motion of atoms because of the modulation of the nuclear dipole-dipole interactions by the atom-atom exchanges. The relaxation rates are determined by the spectral densities $G_n(n\omega)$ given by the Fourier transforms of the autocorrelation functions $< Y^{kl}_{2n}(0) Y^{kl}_{2n}(t) >$ where the Y^{kl}_{2n} are the spherical harmonics associated with the orientation of the inter-

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atomic vector $\hat{r}_{kl}$. ω is the nuclear Larmor frequency.

The transverse nuclear spin relaxation rate (T_2^{-1}) is given by

$$T_2{}^{-1} = \frac{3}{2}G_0(0) + \frac{5}{2}G_1(\omega) + G_2(2\omega) \qquad (2)$$

At high magnetic fields, $\omega >> J_{eff}$. $G_1(\omega)$ and $G_2(2\omega)$ are therefore negligible, and

$$T_2{}^{-1} = \frac{3}{2}G_0(0) = (\frac{3}{2}M_2)/J_{eff}^{NMR}. \qquad (3)$$

$J_{eff}^{NMR} = \sqrt{(2M_4/(\pi M_2))}$ where M_2 and M_4 are the second and fourth moments of the NMR lineshape. For the triangular lattice, we find

$$J_{eff}^{NMR} = 1.4|(J_2 - 2xJ_3 + \frac{3}{4}x^2J_4 + ...)| \qquad (4)$$

where x is the probability that a given lattice site is occupied by a ^{3}He atom.

2. Experimental Measurements

In order to study the nuclear spin relaxation at high magnetic fields, we used powdered hexagonal boron nitride rather than exfoliated graphite because the latter has a relatively high electrical conductivity and a high diamagnetism that can distort the NMR line shapes and relaxation. The NMR relaxation rates were measured from the decay of echoes, following a $90° - 180°$ RF pulse sequence. The sample cell consisted of a cake of loosely packed boron nitride powder interspersed with fine copper wires. The surface area was characterized by two methods: first using adsorption isotherm measurements[2], and second, by measuring the relaxation at fixed temperature as a function of surface coverage. A sharp minimum was observed in the relaxation time for a complete monolayer coverage.[3] This reliable characterization of the commensurate coverage for the boron nitride surface was carried out for both pure ^{3}He and 50% ^{3}He:Ne.

The relaxation times observed as a function of temperature for a fixed coverage, corresponding to the $\sqrt{3} \times \sqrt{3}$ coverage, are shown in Fig. 1. An exponential temperature dependence attributed to the thermal activation of vacancies was observed at high temperatures(solid line A in Fig. 1). The temperature independent regions (B and C of Fig. 1) result from the quantum mechanical exchange motions. The values in this plateau region can be analysed using the expressions given above for J_{eff}^{NMR}. Neglecting J_4, J_5 etc., we find $J_2 = 3.25.10^5$ Hz, and $J_3 = 10.4.10^5$ Hz.

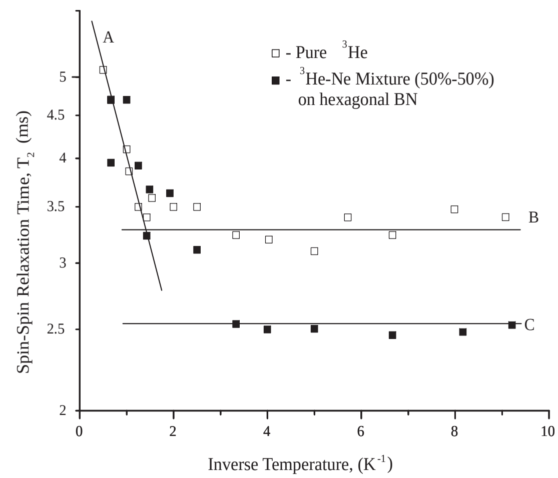

Fig. 1. Temperature dependence of the nuclear spin-spin relaxation times for commensurate $\sqrt{3} \times \sqrt{3}$ monolayers on BN for: (i) pure ^{3}He, and (ii) a 50% ^{3}He:Ne mixture. The solid lines B and C represent the quantum tunelling regimes determined by the effective exchange frequencies for the pure and diluted samples, respectively.

3. Conclusions

The temperature independent nuclear transverse relaxation times observed at low temperatures for monolayer ^{3}He films have been interpreted in terms of an effective exchange frequency that changes significantly with replacement of the ^{3}He atoms with immobile Ne atoms. The relaxations times are observed to be reduced on diluting the ^{3}He coverage whch is opposite to the behavior expected for a classical rigid lattice. Numerical analysis of the observed values for the relaxation rates yield values for the two exchange frequencies, assuming higher spin exhanges can be neglected.

Acknowledgements

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