Abstract. We report measurements of the nuclear spin-lattice and spin-spin relaxation times of very dilute \( ^3\text{He} \) in solid \( ^4\text{He} \) in the temperature range \( 0.01 \leq T \leq 0.5 \) K for densities where anomalies have been observed in torsional oscillator and shear modulus measurements. We compare the results with the values of the relaxation times reported by other observers for higher concentrations and the theory of Landesman that takes into account the elastic properties of the \( ^4\text{He} \) lattice. A sharp increase in the magnitude of the nuclear spin-lattice relaxation times compared to the classical Landesman theory is observed close to the temperatures where the torsional and shear modulus anomalies are observed. The NMR results suggest that the tunneling of \( ^3\text{He} \) impurities in the atomic-scale elastic distortion is affected by the same processes that give rise to the macroscopic elastic dissipation anomalies.

1. Introduction

Great interest has been generated by the discovery of non-classical rotational inertia fractions (NCRIFs) in solid \( ^4\text{He} \) by Kim and Chan [1, 2] as this could be evidence of a transition to a state supporting superflow in the solid at low temperatures [3]. In addition measurements of the elastic properties of the crystal have demonstrated the existence of a frequency dependent change in the shear modulus [4] and an enhanced dissipation peak [5] with a temperature dependence that appears to mimic that of the NCRIF anomaly. An anomalous peak is also seen in the sound attenuation [6]. These results suggest that the dynamical properties of the \( ^4\text{He} \) lattice play an important role in these two anomalous responses and we therefore carried out NMR experiments [7, 8] to study the microscopic dynamics for samples at the very low \( ^3\text{He} \) concentrations (\( x_3 \leq 25 \) ppm) for which the NCRIFs are still clearly visible.

Measurements of the nuclear spin-lattice relaxation times (\( T_1 \)) (Fig. 1) revealed a well-defined peak in the relaxation time in a narrow temperature range comparable to that reported for the other two anomalies [7]. A similar but less well defined anomaly is seen in measurements of the nuclear spin-spin relaxation times (\( T_2 \)). These deviations from the expected temperature independent relaxation induced by the tunneling of the \( ^3\text{He} \) atoms through the \( ^4\text{He} \) lattice are distinct from the phase separation into pure \( ^3\text{He} \) droplets that occurs at lower temperatures. In order to better understand the tunneling in the \( ^4\text{He} \) lattice, the results at high temperatures (\( T \geq 0.2 \) K) are compared with the values reported elsewhere for higher concentrations.
(100 ≤ x_3 ≤ 10,000 ppm) for molar volumes 28.9 ≤ V_m ≤ 29.2 cm^3 in the same temperature range 0.2 ≤ T ≤ 0.5 K.

![Figure 1. Temperature dependence of the nuclear spin-lattice relaxation for a 3He concentration of 16 ppm (data from [7]). The sharp drop at 85 mK marks the 3He-4He phase separation, while the narrow peak at 175 mK is the anomalous temperature dependence discussed in Sec. 3.](image1)

![Figure 2. Variation of the function F(x_3) = (\frac{1}{2})T_1T_2x_3^2M_2^2/\omega_L^2 as a function of 3He concentration. For a unique correlation time F(x_3) = 1.0. Experimental data: □ Kim et al. [7], △ Allen et al. [9], ○ Schratter et al. [10], ⊙ Schuster et al. [11], ▽ Hirayoshi et al. [12].](image2)

2. Comparison of Results with Relaxation Models

In order to understand the origin of the prominent peak in the relaxation time at T = 175 mK we will first review the standard models of nuclear spin relaxation expected for 3He impurities in otherwise pure solid 4He and compare the results with published results for 16 ≤ x_3 ≤ 10^4 ppm [7, 9, 10, 11, 12, 13]. The most remarkable result is that the data for T_1 and T_2 cannot be described by a common correlation time. In Fig. 2 we plot the product $F(x_3) = (\frac{1}{2})T_1T_2x_3^2M_2^2/\omega_L^2$ where $M_2$ is the NMR second moment for pure 3He and $\omega_L$ is the Larmor frequency. For a simple Lorentzian spectral density with a unique correlation time, $F(x_3) = 1$, independent of $x_3$. This is clearly not satisfied. Attempts to use exponential spectral densities such as that proposed for $^3$He$_2$ molecular motion [14, 15] also fail. The latter leads to ratios $(T_1/T_2) \sim \exp(\omega_L/2J_{34}) \sim 3$ while the experimental values are $\sim 10^3$. The results lead one to conclude that there are at least two components to the spectral densities, one for the high frequency response and $T_1$, and a second for the low frequency response and $T_2$.

Obvious candidates for the distinct components to the spectral densities are (i) the group velocity of the tunneling 3He impurity, and (ii) the scattering of that motion by the elastic fields of the lattice. Because of the difference in zero-point motion of a 3He atom compared to a 4He atom there is an appreciable lattice distortion surrounding the 3He atoms described by a long range interaction $K(r_{ij}) = K_0/r_{ij}^3$ for 3He atoms at sites $i$ and $j$ [16, 17]. The 3He motion is therefore determined by both the tunneling rate $J_{34}$ and the elastic field constant $K_0$. The relaxation rates are determined by evaluating the expectation values of the correlation functions $\Gamma_{ijkl}(t) = \langle C_{ij}(t)C_{kl}(0) \rangle$ where $C_{ij}(t) = \exp(-iK_it)[D_{ij}, H_{34}] \exp(iK_jt)$ with $D_{ij}$ representing the tensorial components of the dipolar interactions. $H_{34}$ is the 3He-4He tunneling Hamiltonian. Two different approaches have been made to calculate these expectation values: (1) the statistical method of Landesman [16] valid for the high frequency limit of the spectrum and estimations of
Figure 3. Observed concentration dependence of the nuclear spin-lattice relaxation times for dilute ³He in solid ⁴He. [square] Greenberg et al. [13] and for the rest of symbols see the caption for Fig. 2.

Figure 4. Comparison of the concentration dependence of the nuclear spin-spin relaxation times reported in the literature for dilute ³He in solid ⁴He. For symbols see the caption for Fig. 2.

$T_1$, and (2) the method of Huang et al. [17] valid for $T_2$ calculations. Landesman [16] evaluates the correlation function

$$\Gamma(t) = 48J_{34}^2 \sum_{i,j} (a_0/r_{ij})^6 \prod_{p \neq (i,j)} \exp[it(K_{ip} - K_{pj})]$$

and, restricting the terms $K_{ip} - K_{pj}$ to values close to zero, finds a correlation time $\tau_c^{-1} = 23(J_{34}^2/K_0 x_3^{1/3})$. For $\omega_L > \tau_c^{-1}$, the calculated relaxation time is

$$T_1^L = \frac{\omega_L^2 K_0}{M_2 46J_{34}} = 0.86 \times 10^{-3} x_3^{-2/3}$$

for $J_{34}/(2\pi) = 1.8$ MHz and $J_{34}/K_0 = 1.0 \times 10^{-3}$ chosen to provide the best fit as shown by the solid green line in Fig. 3. (The values of $T_1$ have been normalized to a Larmor frequency of 2 MHz, assuming a Lorentzian spectral density.) For very dilute ³He concentrations, $x_3 \leq 200$ ppm, the ³He-³He collisions are too rare to be effective and the characteristic frequency that determines the relaxation is given by the time, $\tau_{ch}$, for the impurity to travel the mean distance between sites, $\tau_{ch}^{-1} = x_3^{1/3} z J_{34}$ where $z$ is the lattice coordination number. This yields a relaxation time $T_1 = 2.8 \times 10^3 x_3^{-4/3}$ s, shown by the solid red line in Fig. 3.

If one uses the calculation of Landesman to estimate the spin-spin relaxation, the result is off by an order of magnitude and predicts the incorrect concentration dependence as shown by the green line in Fig. 4. Huang et al. [17] addressed this discrepancy by noting that because of the $r^{-6}$ dependence in the correlation functions, only atoms very close together can contribute and that for an atom at site $r_i$ tunneling to $r_j$ with a spectator at $r_k$, only pairs for which $K(r_i) - K(r_j) = 2J_{34}$ are to be considered. This energy conservation occurs for separations $r \sim a_0(K_0/2J_{34})^{1/3}$ which becomes greater than the mean ³He separation for $x_3 \leq 500$ ppm. Weighting the $r^{-6}$ sum in Eq. 1 by the number of sites satisfying the conservation requirement, one estimates an effective tunneling frequency $\tau_H^{-1} = 8.8J_{34}^2/K_0 = 9.6 \times 10^4$ rad/s, independent
of $x_3$, using the same values of $J_{34}$ and $K_0$ as in Landesman’s calculation. The fit to the data shown by the solid red line of Fig. 4 is remarkably good given the approximations that have been made. Huang et al. [17] carried out detailed Monte Carlo calculations and showed that for $x_3 \leq 400$ ppm $\tau_H^{-1}$ develops a linear dependence on $x_3$, which leads to a concentration and temperature independent $T_2$ as shown by the dotted red line of Fig. 4.

3. The Anomalous Nuclear Spin Relaxation

The approaches to the calculations of the nuclear spin relaxation times of $^3$He impurities in solid $^4$He treat the distortion field around the impurity as temperature independent. The experiments of Beamish et al. [4, 5] indicate that there is “in addition” a dynamical component that has a prominent temperature dependence. Including a simple static thermal excitation does not fit the data as it leads only to a smooth step in the temperature dependence of $T_1$. Beamish et al. [4, 5] introduced a dynamical relaxation $\Gamma(t) = \gamma_0 \exp(E/T)$ to describe the frequency dependent shear modulus. If we assume that the nuclear spin relaxation occurs via this dynamical relaxation of the lattice in series with the process described in section 2, then because the effective frequency of the rapidly oscillating term in the relaxation function $\Gamma(t)$ is $\omega_e = x_3^{4/3}K_0 \approx 100$ Hz, the additional relaxation would be expected to be proportional to the Lorentzian form $\omega_e \tau(T) /[1 + (\omega_e \tau(T))^2]$ analogous to the anomalous term observed for the shear modulus. While this temperature dependence does provide a qualitative fit to the data (with an appropriate scale factor), a detailed microscopic theory of the lattice relaxation is needed to justify the introduction of this additional process for the relaxation.

4. Conclusion

The detailed concentration dependence of the nuclear spin relaxation of $^3$He impurities in solid $^4$He for $10 \leq T \leq 400$ mK can be understood if we assume that the tunneling of the $^3$He atoms is modified by the same process that leads to the macroscopic dynamical relaxation observed in studies of the shear modulus. The temperature dependence of this additional term fits a model of a thermally activated relaxation for the lattice in response to the effective motion of the tunneling in the local crystal fields.

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