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Lattice Relaxation of $^4\text{He}$ with $^3\text{He}$ Impurities: NMR Studies

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Abstract.
Measurements of the $^3\text{He}$ nuclear spin relaxation times of dilute $^3\text{He}$ impurities in solid $^4\text{He}$ have been used to explore the unusual dynamics of solid $^4\text{He}$ at low temperatures. The $^3\text{He}$ impurities move through the lattice by quantum mechanical exchange with neighboring $^4\text{He}$ atoms. Because of the larger zero point motion of the $^3\text{He}$ atoms, there is an appreciable lattice distortion that accompanies the tunneling $^3\text{He}$ atom and the tunneling motion depends on the elastic properties of the $^4\text{He}$ lattice. This motion modulates the $^3\text{He}^-^4\text{He}$ nuclear dipole-dipole interactions and thus determines the NMR relaxation rates. We compare the observed temperature dependence of the NMR relaxation rates with that expected from the measurements of the shear modulus by Syshchenko et al. [Phys. Rev. Lett. 104, 195301 (2009)].

1. Introduction
Great interest has been generated by the observation of a ‘giant plasticity’ in solid $^4\text{He}$ by Rojas et al.[1, 2] that has been attributed to a remarkable mobility of dislocations in the solid at low temperatures. It was shown[3] that for very pure single crystals the dislocations can move over large distances and produce large changes in both the elastic shear modulus and the dissipation that is associated with the motion of the dislocations. This observation was preceded by the detection of a significant change in the shear modulus of solid $^4\text{He}$[4–6]. The unusual plasticity has been invoked[3] to explain some of the previous anomalies reported for solid $^4\text{He}$ including the detection of non-classical rotational inertias in the same temperature region[7–13] and unexpected peaks in the sound attenuation[14, 15]. For recent reviews see Balibar[16] and Chan et al.[17].

We have carried our NMR studies of the motion of $^3\text{He}$ impurities in solid $^4\text{He}$ as a new non-invasive means of studying these unusual properties of the $^4\text{He}$ solid[18]. The $^3\text{He}$ impurities tunnel by particle-particle exchange through the lattice[19, 20] but because of the larger zero point motion of the $^3\text{He}$ atom compared to $^4\text{He}$ atoms, the lattice surrounding the impurity is distorted and the lattice must relax as these deformations move. Studies of the microscopic $^3\text{He}$ motion using NMR techniques can therefore provide a sensitive probe of any change in the elastic properties of the lattice.
2. Diffusion of Impurities in Quantum Solids

The atoms in quantum solids such as $^3$He are weakly localized with respect to their lattice sites because of the relatively high zero point kinetic energy and the weak Van der Waals binding forces and can therefore exchange sites with nearest neighbors by quantum tunneling[21]. This effect leads to the multi-particle spin exchange in solid $^3$He[22, 23] and the quantum diffusion of isotopic impurities such as $^4$He in solid $^3$He[24]. The diffusion of $^3$He in solid $^4$He has been studied extensively by Richards and colleagues[19, 20, 25–27] and by the team of Grigoriev[28] who interpreted the results in terms of a gas-like diffusion of the impurities with a tunneling frequency $J_{34}$ and diffusion constant $D \sim zJ_{34}a_0^2/x_3\sigma$ where $z$ is the lattice co-ordination number and $\sigma$ the scattering cross-section for tunneling impurities. A full quantitative understanding for relatively high $^3$He concentrations was not achieved until Landesman and others[29–31] realized that one had to account for the lattice distortion around the $^3$He impurity that results from the larger zero point motion of the $^3$He atoms compared to the $^4$He atoms in the lattice.

The elastic deformation around each impurity leads to a long range interaction $K_{ij} = K_0/r_{ij}^3$ between $^3$He impurities[29]. The range of this interaction, $R_K$, is determined by the distance of closest approach for two impurities which occurs when the elastic energy difference is equal to the change in kinetic energy or $a_0 |\nabla K(r)| = J_{34}$, for which $R_K = (3K_0J_{34})^{1/4} = 8.2a_0$ where $a_0$ is the lattice spacing. The effective tunneling frequency $J_{eff} \sim J_{34}/K_0 \sim kHz$. The mean separation between $^3$He atoms is given by $R_M = x_3^{(3/2)}a_0$, and for $^3$He concentrations $x_3 > 180$ ppm, $R_K > R_M$, and the $^3$He impurities are in constant (liquid-like) interaction. Experimental studies prior to 2010 were limited to concentrations $10^4 < x_3 < 10,000$ ppm and studied this constant interaction regime for which the the characteristic microscopic correlation time is given by $\tau_c^{-1} = 19R_0^2 x_3^{-1/3}$ [30, 32]. The corresponding nuclear spin-lattice relaxation time induced by the tunneling is temperature independent and given by $T_1^{nu} = 0.86x_3^{-1/3}$.

The more interesting low concentration regime for $R_M \gg R_K$, corresponding to $x_3 \ll 100$ppm has only recently been studied in the experiments by Toda et al.[11, 33] and Kim et al.[18, 34, 35]. In this regime the separation of the $^3$He atoms is much larger than the scattering length and collisions are rare. This is the coherent tunneling regime (originally postulated by Richards et al.[19]) where the group velocity of the $^3$He impurity motion is $v_G = za_0J_{34}$. The characteristic time for a $^3$He atom to tunnel between sites is given by $\tau_{coh}^{-1} = (zJ_{34})^{1/4}x_3^{1/3}$ for which the nuclear spin-lattice relaxation time is $T_1^{coh} = 2.410^{-3}x_3^{-1/3}$ for $J_{34}/(2\pi) = 0.6$ MHz in good agreement with experiments above 0.2 K. Below 0.2K we observe an anomalous peak in the nuclear spin-lattice relaxation close to where Beamish et al.[4, 6] observed a peak in the dissipation for the measurements of the shear modulus. Anomalous features in the attenuation of sound have also been observed[14], pointing to changes in the elasticity of the solid in this temperature region.

3. Experimental Considerations

The sample chamber consisted of a polycarbonate cylinder sealed at one end with a silver end piece that is an integral part of a cold finger extending from a nuclear demagnetization refrigerator. As shown in Fig 1. the helium sample is admitted from the opposite end of the cell which includes an in situ pressure gauge. A crossed-coil design was employed for NMR pulsed excitation and reception[34] with the excitation coil thermally isolated from the sample cell. The NMR spectrometer was a super-heterodyne design operating at 2.01 MHz. The latter was chosen to keep the expected nuclear spin-lattice relaxation times below $10^4$ s. To enhance the signal/noise a low temperature RF preamplifier was deployed close to the sample chamber[36]. The amplifier was operated at 0.8K as determined by the bias current and was moved and thermally linked to the still of the dilution refrigerator when operating to reach the lowest temperatures of $\sim 10mK$. 


Gas samples were prepared by mixing known gas volumes of $^3\text{He}$ and $^4\text{He}$ at room temperature and then condensed into the sample volume at the melting pressure corresponding to the desired molar volume. The sample cell was cooled very slowly through the melting curve as monitored by the pressure gauge, and the samples are formed by the blocked capillary method. Part of the $^3\text{He}$ composition is lost to wall adsorption during the condensation and the precise $^3\text{He}$ composition was determined from the NMR amplitudes. Fig. 2 shows typical results for the temperature dependence of the NMR amplitudes for several samples, illustrating the Curie law behavior at high temperatures followed by the formation of Fermi liquid droplets at very low temperatures[35]. Gas samples prepared with 30 ppm and 20 ppm of $^3\text{He}$ were found, for example, to have only 26 ppm and 16 ppm concentrations, respectively, in the solid sample.

4. Results
The results for measurements of both the nuclear spin-lattice times, $T_1$, and spin-spin relaxation times, $T_2$, as a function of temperature are shown in Fig. 3 for a sample with a $^3\text{He}$ concentration of 16 ppm. A sharp change in the value of $T_1$ by a factor of 30 is seen at the phase separation temperature of 90 mK. This transition temperature is in good agreement with the theoretical predictions of Balibar and Edwards[35, 37]. Above the transition temperature we expected a temperature independent relaxation as predicted for the coherent quantum tunneling of the isotopic impurities[29, 30] and as observed at much higher concentrations[20, 27].

The surprising result was a large peak (factor of 2 increase) in the spin-lattice relaxation time near 180 mK and accompanied by a less clear dip in the magnitude of the spin-spin lattice relaxation time in the same temperature range. We also show results for the spin-lattice relaxation time for 500 ppm for which no strong evidence of a peak is seen, consistent with the results of Schratter et al.[20]. The observations for the dilute sample with 16 ppm $^3\text{He}$ imply an increase at $T \sim 180$ mK in the low frequency component of the spectral densities, $J(\omega)$, of the fluctuations in the nuclear spin-spin interactions that are responsible for the relaxation. The
increase in $J(0)$ would result in a decrease in the spectral density at the Larmor frequency as $J(\omega)$ is normalized. Since $T_2^{-1} \propto J(0)$ and $T_1^{-1} \propto J(\omega_L)$ this change in the spectral densities would lead to a peak in the value of $T_1$ with a concomitant decrease in the value of $T_2$. The value of $T_2$ at low temperatures is higher than the values for $T > 0.2K$, suggesting that some of the quantum tunneling that narrows the line width is reduced by scattering or other mechanisms at high temperatures. The origin of this feature is not understood.

It is worth noting that because of the very long relaxation times it takes about one day to measure each single point in Fig. 3 and for that reason the number of data points for a given sample is limited. In Fig. 4 we also show the results for a 24 ppm sample that was never cooled below the phase transition temperature. The peak in $T_1$ is only seen for low $^3\text{He}$ concentrations when the mean separation is larger than the range of the elastic deformation surrounding each impurity.

![Figure 3. Temperature variation of the nuclear spin-lattice and spin-spin relaxation times for samples with helium concentrations of 16 and 500 ppm (closed circles for cooling, open circles for warming). The dashed lines show the temperature independent predictions for quantum tunneling of the $^3\text{He}$ impurities. The dotted line marks the phase separation.](image)

5. Phenomenological model
The nuclear spin-lattice and spin-spin relaxation times are determined by the fluctuations in the $^3\text{He}$ nuclear spin dipole-dipole interactions that result from the tunneling of the $^3\text{He}$ atoms. Because of the lattice deformation surrounding a $^3\text{He}$ atom, the relaxation of the lattice deformation as the $^3\text{He}$ atom moves and thus the elasticity of the solid, play an important role in the relaxation process. The interaction between the $^3\text{He}$ atoms at sites $i$ and $j$ due to the elastic deformation has been given by Landesman[29] and Huang et al.[31] as

$$K_{ij}(r) = K_0(1 - 3 \cos^2 \theta_{ij}) r_{ij}^{-3}$$

where $K_0/(2\pi) \sim 1200$ MHz[29]. If we designate $\tau_j$ as an operator for the creation of a deformed site $j$ occupied by a $^3\text{He}$ atom, we need to consider the correlation function

$$\langle \tau_j(t) \tau_k(0) \rangle = \left\langle j \left| \exp[-i \sum_p (K_{ip} - K_{pk}) t] \right| k \right\rangle = \prod_p \langle j | \cos[(K_{jp} - K_{pk}) t] | k \rangle$$

where the sum over $p$ will only be significant for near neighbors. The frequency scale for this correlation function is set by the difference $K_{ip} - K_{pj}$ which for nearest neighbors is given by $\omega_p = 3\left(\pi^2 x_3^4\right) K_0$, the root mean square of the gradient of $K(r_{ij})$. For $K_0/(2\pi) = 1200$MHz we find $\omega_p/(2\pi) = 2.4$kHz.

The relevant correlation time for determining the nuclear spin-lattice relaxation times is evaluated by completing the spatial average of Eq. 2 and integrating over time. Because of the rapid oscillation of the terms $\cos(K_{ip} - K_{pj})t$ the terms cancel except for very small $t$. To
carry out the evaluation, the cosine terms are therefore expanded in \( t \) and the average taken over spatial co-ordinates using Anderson’s method[38] before integrating. This method provides a reasonable evaluation of the high frequency component of the spectral densities but it does not account for the low frequency component such as that responsible for the dissipative term of the shear modulus[4, 6]. It is this missing component of the spectral density that we believe is responsible for the anomalous peak seen in the nuclear spin-lattice relaxation near 0.2 K.

In the absence of a microscopic theoretical model for the lattice behavior we use a phenomenological model by separating the time domain of the integration of Eq. 2 into two components: (i) the short time regime considered above, and (ii) a new long term (low frequency) additional component. We approximate this additional term by replacing the oscillatory terms by the average \( \cos(\omega_p t) \) and adding a phenomenological decay term \( \tau_L = \tau_0 \exp(\Delta/T) \) analogous to that invoked by Beamish et al.[4, 5]. The additional term in the correlation time is given by

\[
\tau_{RL} = \int \langle \langle \tau_f(t) \tau_k(0) \rangle \rangle_\text{avg} = \Re \int \langle j|k \rangle \exp[i\omega_p t - t/\tau_L] dt = R_0^{-1} \left[ \frac{u}{1 + u^2} \right]
\]

where \( u = z\omega_p \tau_L \) and \( R_0 \) is the phonon induced transition rate that includes the lattice relaxation. We estimate \( R_0 \) from Abragam’s treatment[39] of single phonon induced transitions (Eq. 83 of Ref. [39]) and find \( R_0^{-1} = 2600T^{-1} \) s for \( x_3 = 16 \) ppm. The best fit shown in Fig. 4 for the total relaxation (adding \( \tau_{RL} \) to the temperature independent coherent relaxation) is given by \( R_0^{-1} = 1610T^{-1} \) s with \( \tau_0 = 18 \) ns and \( \Delta = 1.01 \) K. This fit implies a value \( K_0/(2\pi) = 1350\text{kHz} \), slightly higher than that cited by Landesman[29]. The dotted line is the calculated variation for \( \Delta = 0.67 \) K, the mean value for binding energy for \( ^3\text{He} \) atoms on dislocations[3].

\[
\tau_{RL} = \int \langle \langle \tau_f(t) \tau_k(0) \rangle \rangle_\text{avg} = \Re \int \langle j|k \rangle \exp[i\omega_p t - t/\tau_L] dt = R_0^{-1} \left[ \frac{u}{1 + u^2} \right]
\]

6. Discussion
The value inferred for the activation energy of the relaxation process, \( \Delta = 1.01 \) K, is appreciably larger than the typical energy deduced by Fefferman et al.[3] for the binding of \(^3\text{He} \) atoms to dislocations in very pure crystals (0.3 ppm \(^3\text{He} \)) and much larger than that observed by Beamish et al. [4, 5] for the anomalous peaks in the shear modulus for crystals of similar quality. This is not necessarily surprising for the high \(^3\text{He} \) concentration of the samples used for the NMR studies. For the expected concentrations of dislocations,[40, 41] less than 1 ppm of \(^3\text{He} \) would saturate all dislocations and their effect would not be seen in the current NMR experiments.

![Figure 4](image_url)

**Figure 4.** Comparison of the temperature dependence of the nuclear spin-lattice relaxation times for dilute \(^3\text{He} \) in solid \(^4\text{He} \) for concentrations \( x_3 = 16, 24 \) and 500 ppm \(^3\text{He} \) with phenomenological model for lattice relaxation. A typical error bar is shown for the 16 ppm data. The broken vertical bars mark the phase separation into small Fermi liquid droplets. The 24 ppm sample was never cooled below the phase separation temperature.
The mechanism for the thermal activation in these samples is therefore expected to be associated with a different physical process, possibly the motion of grain boundaries in solid $^4$He and a deeper theoretical understanding of grain boundary dynamics is needed to determine if this is the case.

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