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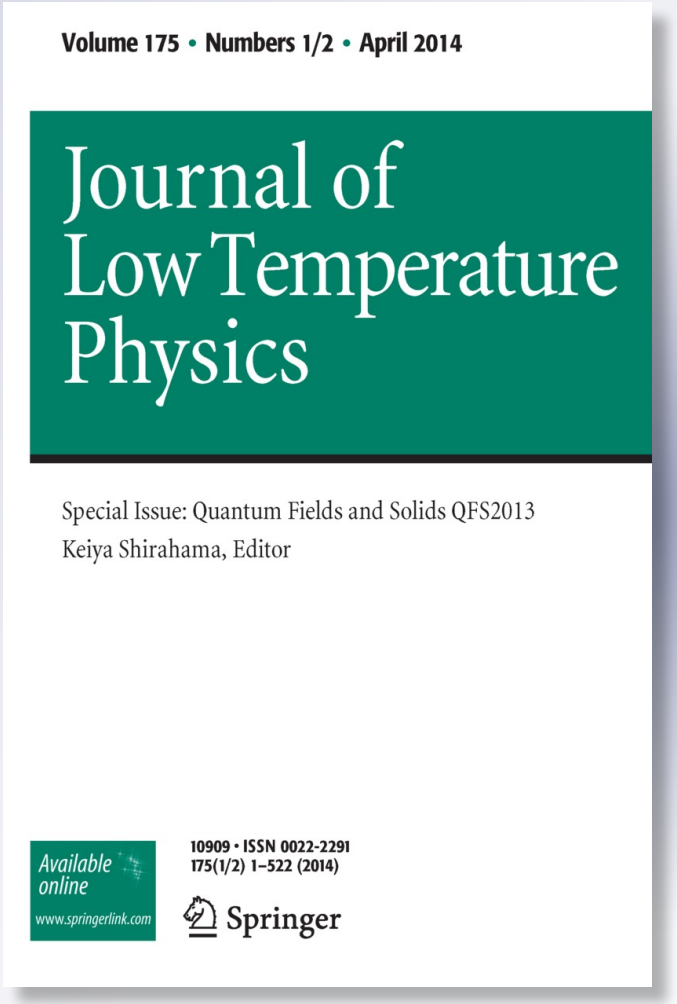

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Lattice Relaxation in Solid ^{4}He: Effect on Dynamics of ^{3}He Impurities

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Abstract We present a new simplified derivation of the effect of lattice relaxation that accompanies the quantum tunneling of ^{3}He impurities in solid ^{4}He on the nuclear spin-lattice relaxation of the ^{3}He impurities for very low impurity concentrations. As a result of the larger zero point motion of the ^{3}He impurity compared to the ^{4}He atoms, a significant lattice distortion accompanies the impurity as it moves through the lattice and the dynamics of the impurity depends on both the interaction energy between two ^{3}He atoms and on the relaxation of the lattice for the tunneling impurity. Using a phenomenological model for the lattice relaxation we compare the nuclear spin-lattice relaxation rates observed at low temperatures with the dependence on temperature expected for a ^{4}He lattice relaxation comparable to that observed by Beamish et al. (Phys. Rev. Lett. 96:195304, 2006).

Keywords Tunneling · NMR · Lattice relaxation

1 Introduction

The observation of a number of distinct anomalies in the physical properties of solid ^{4}He at low temperatures has attracted considerable interest as they point to a pre-

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viously unsuspected behavior for the elastic properties of the solid. The anomalies range from peaks in the sound attenuation near 0.2 K [2], anomalous frequency and dissipation responses of torsional oscillators (TOs) [3, 4], a prominent frequency dependent change in the shear modulus [1, 5], apparent transport through the bulk using porous glass to transmit atoms [6], response to bulk rotation [8], ^{3}He dependent hysteresis [9], unusual excitation spectrum of ^{4}He [10], to a large softening of the shear modulus of the solid at low temperatures referred to as a "giant plasticity" effect [7]. For a recent survey see Balibar et al. [11].

All of these anomalous results involve the use of external stresses or other macroscopic probes. In order to gain more insight into the low temperature behavior of solid ^{4}He we have carried out non-invasive studies of the microscopic dynamics of solid ^{4}He by using NMR techniques to study the motion of ^{3}He impurities at very low concentrations [12–14] compared to earlier studies at higher concentrations and higher temperatures [15, 16]. The ^{3}He impurities move through the ^{4}He lattice by ^{3}He-^{4}He particle exchange. Because the zero point motion of the ^{3}He atom is larger than that of a ^{4}He atom, there is an appreciable distortion of the lattice surrounding the impurity and this lattice deformation must move with the tunneling impurity. As a result, the effective tunneling observed by NMR depends on the elastic properties of the ^{4}He lattice. Any significant changes in the elastic properties would be expected to result in an associated change in the NMR relaxation rates. NMR studies of solid solutions of dilute ^{3}He in ^{4}He have been carried out by Sasaki et al. [17] and Kim et al. [13]. The latter did observe a peak in the nuclear spin relaxation time at $T \cong 0.17$ K that resembles the dissipation peak observed for the shear modulus by Day and Beamish [1]. In order to understand these NMR results we review the theory of NMR relaxation for ^{3}He impurities in solid ^{4}He and include a semi-phenomenological treatment of the ^{4}He lattice in the calculation of the overall relaxation. Landesman [18] has used a fictious spin method to treat the spin-lattice relaxation but gives few details of the calculation. We will use a more straightforward treatment that makes the role of the lattice interaction between defects more transparent.

2 Nuclear Spin-Lattice Relaxation Theory

The nuclear spin-lattice relaxation of dilute ^{3}He impurities in solid ^{4}He is determined by the modulation of the ^{3}He-^{3}He nuclear dipole-dipole interactions. At low temperatures the modulation results from the quantum tunneling of the ^{3}He impurities by ^{3}He-^{4}He atom-atom exchange, the binding and unbinding of ^{3}He atoms on dislocations, and by any motion of the dislocations. For the ^{3}He concentrations we discuss here, $x_3 > 10$ ppm, the number of ^{3}He atoms bound on the dislocations (for even medium quality samples) is only a very small fraction of x_3. The ^{3}He atoms at the pinning sites on the dislocation lines therefore do not contribute directly in any significant manner to the observed effects. The pinning of the dislocations of course affects the ^{4}He lattice dynamics very significantly and that will indirectly affect the NMR relaxation. The samples discussed here were prepared by very slow cooling using the blocked capillary method with a final pressure of 27.75 ± 0.05 bar at 1.2 K [14]. The

tunneling Hamiltonian is expressed in the standard form

$$H_T = -2\hbar J_{34} \sum_{ij} I_i \cdot I_j \tag{1}$$

where I_i is the nuclear spin operator at site i. $J_{34}/2\pi \cong 1$ MHz [19]. We also need the elastic interaction induced by virtual phonon exchange between two impurities at sites j and k for large distances r_{jk}. This interaction can be written as [18, 20]

$$H_{K(ij)} = -2\hbar K_0 \left[\left(1 - 3\cos^2(\theta_{jk})\right)/r_{jk}^{-3}\right] \tau_i \tau_j \tag{2}$$

where τ_i is a creation operator for the impurity at site j. θ_{jk} is the polar angle specifying the orientation of the internuclear vector r_{jk} with respect to the magnetic field, and $K_0/2\pi \cong 1200$ MHz.

The dipole-dipole interaction between two atoms at sites i and j, H_{DD}, can be expressed in terms of irreducible nuclear spin operators, T_l^m, that transform analogously to the spherical harmonics, Y_l^m, with [22]

$$H_{DD} = \frac{\mu_0}{4\pi}\left(\frac{4\pi}{5}\right)^{1/2} \hbar^2\gamma^2 \sum_{ij}\sum_{m} Y_2^{-m}(\Omega_{ij}) T_{ij}^m / r_{ij}^3 \tag{3}$$

The magnitude of H_{DD} is $\approx 10^3$ Hz for $x_3 \approx 1000$ ppm. In general the relaxation rate is given by $\frac{1}{T_1} = J_1(\omega_L) + 4J_2(2\omega_L)$ where $J_m(\omega_L)$ is the component of the spectral density at the nuclear Larmor frequency, ω_L, of the correlation function

$$\begin{aligned} G_m(t) &= \frac{4\pi}{5}\frac{\hbar^2\gamma^4}{N}\sum_{ij}\sum kl \frac{Y_2^m(\theta_{ij})}{r_{ij}^3}\frac{Y_2^{-m}(\theta_{kl})}{r_{kl}^3}\Gamma_{ijkl}^m(t) \\ \Gamma_{(ij),(kl)}^m(t) &= \frac{1}{\langle I_j^z\rangle^2}\left\langle T_{ij}^m \left(T_{kl}^m(t)\right)^\dagger\right\rangle = \frac{4}{x_3}\left\langle T_{ij}^m e^{-i(H_K+H_J)t}\left(T_{kl}^m\right)^\dagger e^{i(H_K+H_J)t}\right\rangle \end{aligned} \tag{4}$$

For relatively high concentrations, $x_3 > 1000$ ppm, $\Gamma_{(ij),(ij)}(t)$ can be calculated using Anderson's method [23] for dilute systems, and we find (Appendix A)

$$\tau_c^{-1} = 19\frac{J_{34}^2}{K_0} x_3^{-1/3} \tag{5}$$

For the experiments under consideration [13, 14] we have $\omega_L\tau_c > 1$ and from Eq. (4)

$$T_1 = \frac{\omega_L^2}{38M_2}\frac{K_0}{J_{34}^2} x_3^{-2/3} = 0.86 x_3^{-1/3} \tag{6}$$

where M_2 is the second moment for pure ^{3}He. T_1 is temperature independent and more than an order of magnitude below the observed relaxation time $T_1 = 5200$ s for $x_3 = 16$ ppm. The values of x_3 are determined directly and accurately from the amplitudes of the NMR signals themselves [14]. This discrepancy between the data [13, 14] and Landesman's model [18] is due to the fact that at very low

concentrations, collisions between impurities are rare when the mean separation $r_m = a_0 x_3^{-1/3}$ is much greater than the scattering length for interacting impurities given by $R_K = a_0(3K_0/(2J_{34})^{1/4}) \cong 8a_0$ (a_0 is the lattice spacing.) For low concentrations, $x_3 < 100$ ppm, the characteristic time for the modulation of the dipolar interactions is given by the time to tunnel between neighboring atoms. This time is $\tau_{Coh} = r_m/v_G$ where $v_G = a_0 z J_{34}$ is the group velocity for the impurity motion. z is the co-ordination number. We find $\tau_{Coh} = (zJ_{34})^{-1} x_3^{1/3}$ which gives a good fit to the data described by $T_1^{Coh} = 2.410^{-3} x_3^{-4/3}$ for $J_{34}/(2\pi) = 0.6$ MHz in the temperature independent regime but does not account for the peak in the relaxation time observed at $T = 175$ mK.

The above calculation only describes the coupling between the nuclear Zeeman interactions and the tunneling degrees of freedom. It neglects the relaxation of the strain around the tunneling impurity as the impurity moves. This latter relaxation acts in series with the nuclear spin to tunneling coupling, much like a phonon bottleneck [21]. To evaluate this term we consider the correlation function

$$\langle \tau_j(t)\tau_k(0)\rangle = \left\langle \exp[i \sum_p \left[(H_{K(jp)} - H_{K(kp)})t\right] e^{-t/\tau_L} \right\rangle \tag{7}$$

where we have introduced a phenomenological term $\tau_L = \tau_0 e^{\Delta/T}$ for the lattice relaxation analogous to the treatment of Day and Beamish [1]. Integrating Eq. (7) we find for the additional relaxation time $\tau_{TL} = R^{-1}[u/(1+u^2)]$ where $u = \omega_p \tau_L$ and $\omega_p/(2\pi) = 3\sqrt{8/3} x_3^{4/3} K_0/(2\pi) = 2.35$ kHz is the spatial average of the gradient of $K(r_{ij})$ for $x_3 = 16$ ppm. R is the transition rate for a phonon induced transition in K_{ij} and is estimated from the golden rule

$$R = \frac{4\pi}{\hbar} \sum_E |\langle E_1|H_K|E_2\rangle|^2 \rho(E) N_1 \delta(E_1 - E_2 - \hbar\omega_p).$$

$\rho(E)$ is the density of states and N_1 is the number of thermally excited phonons at temperature T. Adopting the treatment of Abragam [24] we have

$$R = 36\pi z \left(\frac{\omega_D}{x_3}\right)\left(\frac{\omega_p}{\omega_D}\right)^2 \left(\frac{K_0}{\omega_D}\right)^2 \left(\frac{k_B T}{\hbar\omega_D}\right) \tag{8}$$

ω_D is the Debye frequency. We finally have for the overall relaxation

$$T_{1Total} = T_1^{Coh} + \tau_{TL} \quad \text{with } \tau_L = 9100 T^{-1}\left[u/\left(1+u^2\right)\right] \text{ s.} \tag{9}$$

The Lorentzian term leads to a peak in the temperature dependence when $\omega_p \tau_o E^{\Delta/T} = 1$. Because of the dependence of ω_p on x_3 the intensity and position of the peak varies considerably with concentration. This dependence is illustrated in Fig. 1 for a lattice relaxation with $\tau_0 = 9$ nsec. and $\Delta = 1.77$ K. Note that the linear temperature dependence is overwhelmed by the Lorentzian term in Eq. (9).

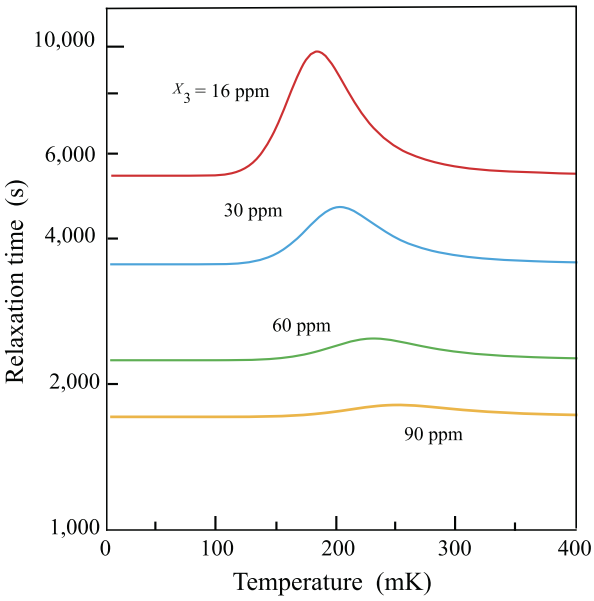

Fig. 1 Calculated temperature dependence of the nuclear spin-lattice relaxation time for four different concentrations. The peak results from the relaxation of the lattice surrounding the moving ^{3}He impurities. The temperature independent contribution is due to the modulation of the nuclear dipole-dipole interactions by ^{3}He-^{4}He particle exchange (Color figure online)

3 Discussion

Figure 2 shows the comparison of the calculated temperature dependence of the relaxation time with that observed experimentally for $x_3 = 16$ ppm [13]. The fit shown is for phenomenological values of an activation energy $\Delta = 1.75$ K and a tunneling time $\tau_0 = 9$ nsec. which are to be compared with values $\Delta = 0.77$ K and $\tau_0 = 8.9$ nsec. deduced by Day and Beamish [1] from studies of the shear modulus at much lower values of x_3. Values of Δ smaller than 1.75 K lead to much broader peaks than that of Fig. 2. The sharp drop at $T = 90$ mK marks the onset of the phase separation of the solid mixture into separate ^{3}He and ^{4}He components [12]. Despite the crude approximations used to calculate the rate R, the fit is qualitatively good. The capture of ^{3}He atoms by dislocations would lead to an increase in the nuclear spin lattice relaxation time but would not be expected to lead to a peak. Also for the density of dislocations in solid ^{4}He observed by Paalanen et al. [25] and Iwasa [26, 27], less than 1 ppm of ^{3}He would saturate all the dislocations and the overall effect would be small. Furthermore, If the effect of dislocations was much larger than that expected from the density of dislocations, the NMR echo would show two components, a small peak on top of the normal broader echo (in the time domain), corresponding to the signal from quasi-static ^{3}He atoms on the dislocations. Within experimental sensitivity no such component has been observed down to the lowest temperatures studied (10 mK).

4 Conclusions

Calculations that include a lattice relaxation following the motion of ^{3}He impurities and their lattice strain are shown to be in good qualitative agreement with the observed nuclear spin-lattice relaxation at very low ^{3}He concentrations. The unusual

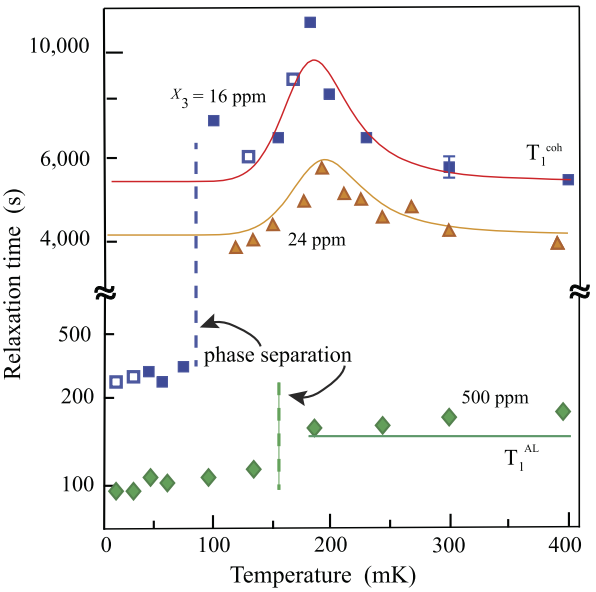

Fig. 2 Comparison of the calculated temperature dependence of the nuclear spin-lattice relaxation times for $x_3 = 16$, 24 and 500 ppm with observed relaxation times shown by the blue squares (*solid squares* for cooling, *open squares* warming), *orange triangles* and *green diamonds*, respectively. For 500 ppm the comparison is with Landesman's model[13], T_1^{AL} valid for high concentrations. The 24 ppm sample was never cooled below the phase separation [14]. A typical error bar is shown for 16 ppm at 300 mK (Color figure online)

peak in the spin-lattice relaxation time mimics that observed for the dissipation peak in the shear modulus [1] but the excitation energies are appreciably higher. Further studies at lower ^{3}He concentrations are needed to obtain a clearer separation of the NMR anomaly from the phase separation transition.

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Appendix A: Calculation of the Correlation Time for High ^{3}He Concentrations

Following the method of Kubo (Cowan [22], Chap. 7), one calculates the time derivative of the autocorrelation function, $\ddot{\Gamma}_{(ij),(ij)}(t)$, and assuming an exponential decay one determines a correlation time $\tau_c^{-1} = G_0(0)^{-1} \int dt \ddot{\Gamma}_{(ij),(ij)}(t)$ The time derivative

$$\ddot{\Gamma} = \frac{4}{x_3}\langle [T_{ij}^m, H_J] e^{-iH_K t} [T_{ij}^{-m}, H_J] e^{iH_K t} \rangle = 6J_{34}^2 \exp\left[\sum_{p(ij)} i(K_{jp} - K_{ip})t\right]$$

with an impurity tunneling from site p to site i with spectator at site j. The energy difference $K_{jp} - K_{ip}$ can be replaced by the average of the spatial gradient of the interaction energy $K_{ij} = K_0(3\cos^2\theta_{ij} - 1)/r_{ij}^3$ to yield

$$\ddot{\Gamma}(t) = 12J_{34}^2 \cos\left(\sum_p \omega_p t\right) \quad \text{with } \omega_p = \frac{3K_0}{(R/a_0)^4}.$$

The sum of the oscillatory terms $\cos(\omega_p t)$ rapidly averages to zero and only small deviations from unity of the cosine term will contribute to the overall average. Using the method of Anderson[23] we write $\ddot{\Gamma}(t) = 12J_{34}^2 \prod_p [1 + (\cos\omega_p t - 1)]$ which can be expanded and the term $\cos\omega_p t - 1$ replaced by its average to yield $\ddot{\Gamma}(t) = 12J_{34}^2 \exp[N\langle\cos\omega_p t - 1\rangle]$. The average

$$N\langle\cos(\omega_p t) - 1\rangle = \frac{2\pi N}{V}\int_0^{\pi/2} d\theta \sin\theta \int_0^{\infty} R^{-4} dR\left(\cos\left[\frac{3K_0(1-3cos^2\theta)t}{R^4}\right] - 1\right)$$
$$= -\frac{\pi}{2} x_3 I_1 I_2 (3K_0 t)^{-3/4} = -0.71 x_3 (K_0 t)^{3/4}$$

where $I_1 = \int_0^{\pi/2} \sin\theta d\theta |1 - 3\cos^2\theta|^{3/4}$ and $I_2 = \int_0^{\infty} dx[\frac{\cos x - 1}{x^{1/4}}] = 2^{3/4}\Gamma(\frac{3}{4}) \times \cos(\frac{3\pi}{8})$. From Eq. (6) we find $\tau_c^{-1} = 19\frac{J_{34}^2}{K_0} x_3^{-1/3}$ in reasonable agreement with Landesman [18].

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