Frequency Dependence of T_1 for ^{3}He in Solid ^{4}He

M. G. Richards, J. H. Smith, P. S. Tofts, and W. J. Mullin*
School of Mathematical and Physical Sciences, University of Sussex, Falmer, Brighton, Sussex, England
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Measurements are reported of $T_1(\omega)$ for ^{3}He impurity in solid ^{4}He at molar volume 21 cm^3 and temperature 0.6 K. Data for fractional impurity concentrations x_3 of 1.0×10^{-3}, 0.5×10^{-3}, and 0.25×10^{-3} show striking anomalies at about 1.3 and 2.6 MHz, superimposed on a background obeying $x_3T_1=1.3\times10^{-2}\exp(\omega/7.4\times10^6)$ sec. A theory is proposed which fits this background and yields a value for the ^{3}He-^{4}He tunneling frequency of about 1 MHz. The anomalies are not fully explained.

^{3}He impurity in solid ^{4}He provides an interesting system in which the prediction[1] of Andreev and Lifshitz that any impurity that can tunnel in a crystalline lattice will become delocalized at low temperatures can be tested. In addition, the magnitude of the frequency, J_{34}, with which the impurity atoms tunnel with host atoms is of interest. Theorists[2] suggest it should not be greatly different from J_{33}, the tunneling frequency in pure solid ^{3}He, while the only published experimental data suggest it is two[3] or three[4,5] orders of magnitude smaller.

Measurements of T_1, the spin-lattice relaxation time of the ^{3}He impurity as a function of frequency, provide information from which the motion of the impurity can be deduced. The method used for obtaining data is to work at one measuring field B_0 (and frequency $\omega_0=\gamma B_0$) and make excursions to other fields for set periods and return to the original field to monitor the growth or decay of spin polarization. The method avoids the need to retune the NMR spectrometer at each of say fifty frequencies used during a single run.

The spin polarization is destroyed at B_0 and the field shifted quickly to $B=\omega/\gamma$ for time τ. It is then returned quickly to B_0 and the polarization monitored by measuring the height, h_τ, of a spin echo following a $\pi/2$-π pulse sequence. T_1 at the frequency ω is given by

$$\tau/T_1(\omega)=\ln[h_\infty{}^\omega/(h_\infty{}^\omega-h_\tau{}^\omega)],$$

where $h_\infty{}^\omega$ is the signal height recorded after waiting indefinitely at ω. It can either be measured directly or calculated from $h_\infty{}^{\omega_0}$, the equilibrium height recorded at ω_0, using $\omega_0 h_\infty{}^\omega=\omega h_\infty{}^{\omega_0}$ since polarization is proportional to field.

The data obtained at molar volume 21.0 cm^3 and at temperature 0.6 K, where the crystal is of hcp form, are plotted in Fig. 1. The run-to-run reproducibility is about 10% and fractional ^{3}He concentrations χ_3, obtained from assuming Curie's law at 1 K, follow to within about 10% concentrations in the gas used to form the samples.

The data suggest a background curve with two anomalies at about 1.3 and 2.6 MHz. In order to investigate further the nature of the anomalies, the background has been subtracted from the experimental data using the relation

$$(1/T_1)_{\text{anomaly}}=(1/T_1)_{\text{observed}}-(1/T_1)_{\text{background}}.$$

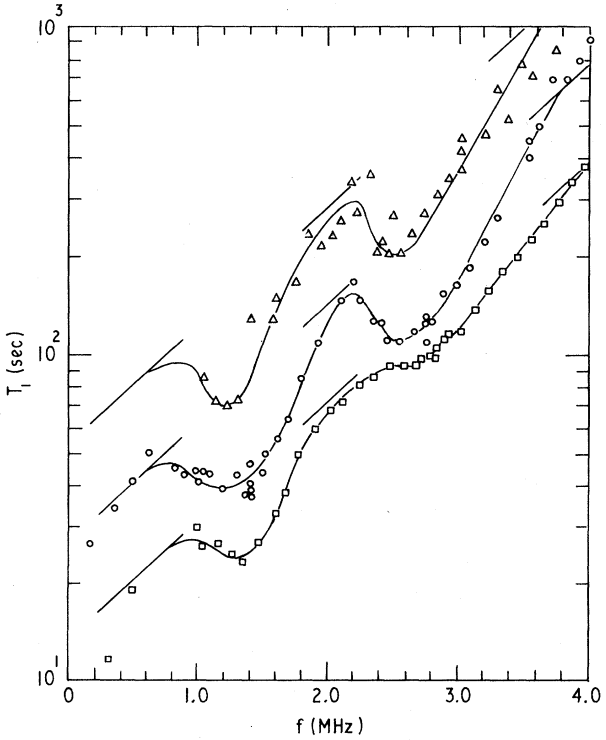

FIG. 1. The spin-lattice relaxation time T_1 for ^{3}He impurity of three concentrations in solid ^{4}He as a function of frequency. The molar volume of the samples is 21.0 cm^3 and the temperature 0.53 K. Fractional ^{3}He concentrations: squares, 10^{-3}; circles, 5×10^{-4}; triangles, 2.5×10^{-4}.

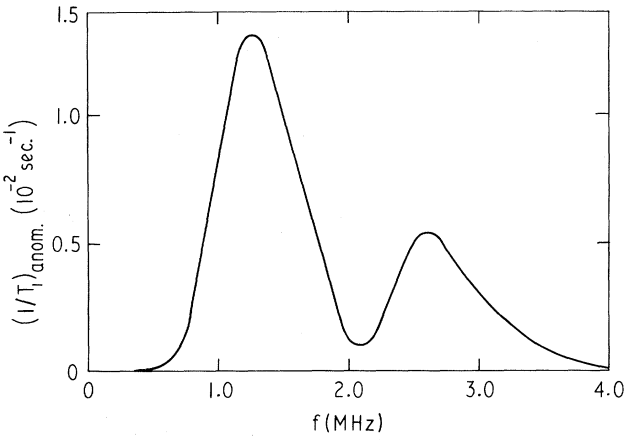

FIG. 2. The anomalous contribution to the spin-lattice relaxation rate as a function of frequency. The fractional ^{3}He content is 5×10^{-4}, the molar volume 21.0 cm^3, and the temperature 0.53 K.

For $(T_1)_{observed}$, the solid lines of Fig. 1 are used and for $(T_1)_{background}$, the expression

$$(T_1)_{background} = (1.3\times10^{-2}/x_3)\exp(f/1.17), \quad (1)$$

where T_1 is in seconds and f is in megahertz. Lines following Eq. (1) for each of the three impurity concentrations, 1.0×10^{-3}, 0.5×10^{-3}, and 0.25×10^{-3}, are shown partially in Fig. 1. $1/(T_1)_{anomaly}$ is plotted for $x_3 = 0.5\times10^{-3}$ in Fig. 2, and the positions and areas of the peaks for each concentration given in Table 1. For a motionally narrowed, isotropic spin system,[6]

$$\int_0^\infty (1/T_1)\,d\omega = \pi M_2, \quad (2)$$

where M_2 is the second moment of the NMR absorption line. If we assume that the ^{3}He spins are distributed at random, then $M_2(x_3) = x_3 M_2(1)$, where[2] $M_2(1) = 22.6\times10^{10}/V_m^2$ is the second moment for pure solid ^{3}He of molar volume V_m. These relations have been experimentally verified[7] at $x_3 = 0.25\times10^{-2}$.

The experimental contribution to the area in Eq. (2) from the background and anomalies is about 50% of the total predicted area. Since the nearest-neighbor contribution to M_2 is about 83% in an hcp crystal, the observed T_1 values must partly reflect the nature of the collisions between two impurities when they are on neighboring lattice sites.

Since the characteristic frequency that is observed (1.17 MHz) is close to the tunneling frequency in pure solid ^{3}He of this density,[2] it appears that when two impurities are on adjacent sites they can tunnel freely without being strongly affected by their own strain-induced interaction.[8] This is due to the fact that in an hcp lattice one of two adjacent impurities has four sites that it can tunnel to, which are also nearest neighbors of the other impurity. Such tunneling conserves strain energy and can therefore be expected to occur at approximately the uninhibited rate J_{34}, which is the rate that a single isolated impurity would tunnel with neighboring ^{4}He atoms.

The model that we have investigated theoretically is one in which ^{3}He impurity atoms move around in the lattice in some unknown way but must spend a fraction $12x_3$ of their time adjacent to another ^{3}He atom (there are 12 near-neighbor pairs). During this time, the pair of impurities tumble around each other at rates of the order of J_{34}. Thus their local (dipolar) field is modulated at frequencies up to this amount, and $T_1(\omega)$ will be scaled by a frequency of this order. The states of the "tumbling pair" will be linear combinations of localized states and will form a band of width $\sim\hbar J_{34}$. Other configurations (second-nearest neighbor, etc.) will form similar bands separated by gaps $\gg\hbar J_{34}$ due to the distortion-induced interaction.

A detailed calculation is possible for the nearest-neighbor contribution. In a preliminary study, we have considered the case where the field is applied along the c axis of the crystal, though for our samples a powder average would

TABLE I. Parameters describing the frequencies and strengths of the anomalies in the frequency dependence of T_1.

x_3	Low-frequency peak position (MHz)	High-frequency peak position (MHz)	Area[a] under $(1/T_1)_{b.g.}$ (10^4 sec^{-2})	Area under lf anomaly (10^4 sec^{-2})	Area under hf anomaly (10^4 sec^{-2})	$\pi M_2(x_3)$ (10^4 sec^{-2})
10^{-3}	1.38± 0.05	2.78± 0.10	56.4	6.0± 0.5	1.6± 0.2	157
5×10^{-4}	1.28± 0.05	2.61± 0.05	28.2	6.2± 0.3	3.0± 0.2	78.5
2.5×10^{-4}	1.26± 0.10	2.53± 0.10	14.1	2.6± 0.4	1.3± 0.2	39.2

[a] Using Eq. (1).

probably be more appropriate. Also, in separating the motion of the two particles into relative motion and center-of-mass motion, the tunneling frequency becomes a function of the direction of tunneling, and in the present calculation a spherical average has been taken. With these approximations the theoretical expression for $T_1(\omega)$ oscillates about the function

$$1/T_1 = (6.7\times 10^8 x_3/J_{34}) \exp(-0.62\omega/J_{34}). \quad (3)$$

It is interesting to note that while the single- and double-frequency terms contributing to T_1 are both approximately Gaussian in ω, their sum is approximately exponential in the range $\omega = 0$ to $\omega \sim 8J_{34}$.

If Eq. (3) is fitted to the experimental data described by Eq. (1), we obtain values for J_{34} at molar volume 21.0 cm^3 as follows: $J_{34}/2\pi$ from intercept, 1.39 MHz; $J_{34}/2\pi$ from slope, 0.73 MHz.

This agreement must be regarded as satisfactory in view of the approximations made. $J_{33}/2\pi$, the equivalent tunneling frequency for pure solid ^{3}He at this density (a value has to be extrapolated from higher densities because the hcp phase only exists below molar volume 19.6 cm^3) is 0.7 ± 0.3 MHz.[2]

The origin of the anomalies in Fig. 1 is not understood. The theoretical curve does have structure though not of the simple and striking form shown in Fig. 2. Whether a more precise theory of the "tumbling pairs" would exhibit the observed structure is not at present clear. It should meanwhile be pointed out that any pair of non-spin-dependent energy levels of two adjacent ^{3}He impurities, with splitting ΔE, will yield two peaks in the $1/T_1(\omega)$ curve in consequence of the relation

$$1/T_1 = J(\omega) + 4J(2\omega),$$

where $J(\omega)$ is the spectral density of the dipole field fluctuations at ω. The two peaks will come at $\hbar\omega = \Delta E$ and $2\hbar\omega = \Delta E$ and the ratio of the areas under these two peaks will be 1:2. The anomalies of Fig. 2 satisfy both these relationships.

In conclusion, two features of the frequency variation of T_1 for these dilute solutions of ^{3}He in ^{4}He may be noted. Firstly, there is a smoothly varying background which is scaled by a frequency of the order of 1 MHz. This suggests a tumbling frequency of this order for a pair of neighboring atoms. Previous published estimates for the ^{3}He-^{4}He tunneling frequency, based on spin-diffusion coefficient D and transverse spin-relaxation times T_2, are two or three orders of magnitude smaller, and this is probably due to the existence[7] of a low-frequency branch to Fig. 1 in the kilohertz range, representing the slow tunneling of ^{3}He-^{4}He neighbors inhibited[8] by other ^{3}He impurity atoms a few lattice spacings distant. Secondly, there is a well-defined structure in the frequency dependence of T_1 in the megahertz region. It is argued that this is due to some simple (but unknown) splitting in the energy spectrum of a neighboring pair of ^{3}He impurities.

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*Present address: Department of Physics and Astronomy, University of Massachusetts, Amherst, Mass. 01002.

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