SPIN RELAXATION IN SOLID ^{3}He: OBSERVATION OF EXCHANGE BOTTLENECK*

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We report here measurements of longitudinal nuclear spin relaxation in solid ^{3}He which give evidence that at sufficiently low temperatures the bottleneck in the energy transfer is between an intermediate reservoir and the lattice. The schematic of Fig. 1 shows the three-bath model which has previously been used[1-3] to interpret relaxation in solid ^{3}He. The intermediate system X has been assumed by other workers to be the exchange part of the Hamiltonian,

$$\mathcal{H}_X = \hbar J \sum_{i<j} \vec{I}_i \cdot \vec{I}_j , \tag{1}$$

where the sum is over nearest neighbors only and $\vec{I}$ is the nuclear spin vector ($I = 1/2$). We also will frequently refer to X as the exchange system, although it is worth noting that many of the results to be discussed depend only upon the existence of an intermediate system and not on its specific nature.

Let τ_{ZX}, τ_{XL}, and τ_{ZL} be the respective Zeeman-exchange, exchange-lattice, and Zeeman-lattice relaxation times and ρ be the ratio of Zeeman to exchange heat capacities. The observed relaxation time T_1 will then be given by

$$T_1^{-1} = \tau_{ZX}^{-1} + \tau_{ZL}^{-1} \tag{2a}$$

for

$$\rho\tau_{XL} \ll \tau_{ZX} \tag{2b}$$

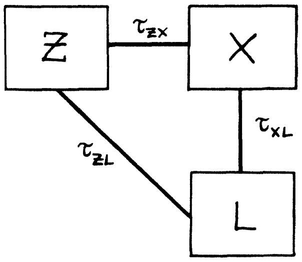

FIG. 1. Three-bath model for relaxation in solid ^{3}He. Zeeman energy Z may relax to the lattice L, assumed to have an infinite heat capacity, either by the direct route labeled τ_{ZL} or by first relaxing to an intermediate system X. The quantities τ_{ZX}, τ_{XL}, and τ_{ZL} are appropriate relaxation times for the rate of approach of $1/T_Z$ and $1/T_X$ to each other and to the inverse lattice temperature $1/T$.

and by

$$T_1 = (\rho + 1)\tau_{XL} \tag{3a}$$

for

$$\tau_{ZL} \gg \rho\tau_{XL} \gg \tau_{ZX}. \tag{3b}$$

If either condition (2b) or (3b) is satisfied, single-exponential time dependence is observed so that T_1 is well defined. In the intermediate region, $\rho\tau_{XL} \approx \tau_{ZX}$, two exponentials may be observed in general.

Reich[1] and Garwin and Landesman[2] report that (2) is apparently fulfilled for all their experiments with pulsed nmr. We, however, using a Rollin circuit[4] to observe the recovery of nmr signals following saturation, do see the "exchange bottleneck" region (3) below 1°K. At higher temperatures where (2) is expected to hold, our results are in good agreement with references 1 and 2.

Figure 2 shows the relaxation time as a function of inverse temperature at molar volumes of 19.0 cm^3 (β phase) and 20.0 cm^3 (α phase). The points refer to data taken with samples containing both $0.05\,\% \pm 0.01\,\%$ and $0.1\,\%$ concentration of ^{4}He impurities. Substantially the same results are obtained for these two concentrations, but as will be discussed later, samples containing $0.5\,\%$ ^{4}He give very different results. We may divide the data into three regions. In region I, condition (2b) holds and the observed relaxation time is given by (2a). The temperature and frequency dependence of T_1 in this region has been successfully analyzed by Hartmann.[3] In the temperature-independent portion of region I, we have

$$T_1 \sim \exp(\omega^2/2\omega_L^{\,2}), \tag{4}$$

where ω is the angular resonance frequency and ω_L is of the order of J. We find $\omega_L/2\pi = 0.78$ Mc/sec at 19.0 cm^3/mole, and Reich quotes $\omega_L/2\pi = 0.68$ Mc/sec at 18.82 cm^3/mole.

At lower temperatures condition (3b) is fulfilled and we observe relaxation times given by (3a) in region III. Since τ_{XL} is independent of frequency and ρ is given by

$$\rho = 8\omega^2/(3zJ^2), \tag{5}$$

where z is the number of nearest neighbors, and

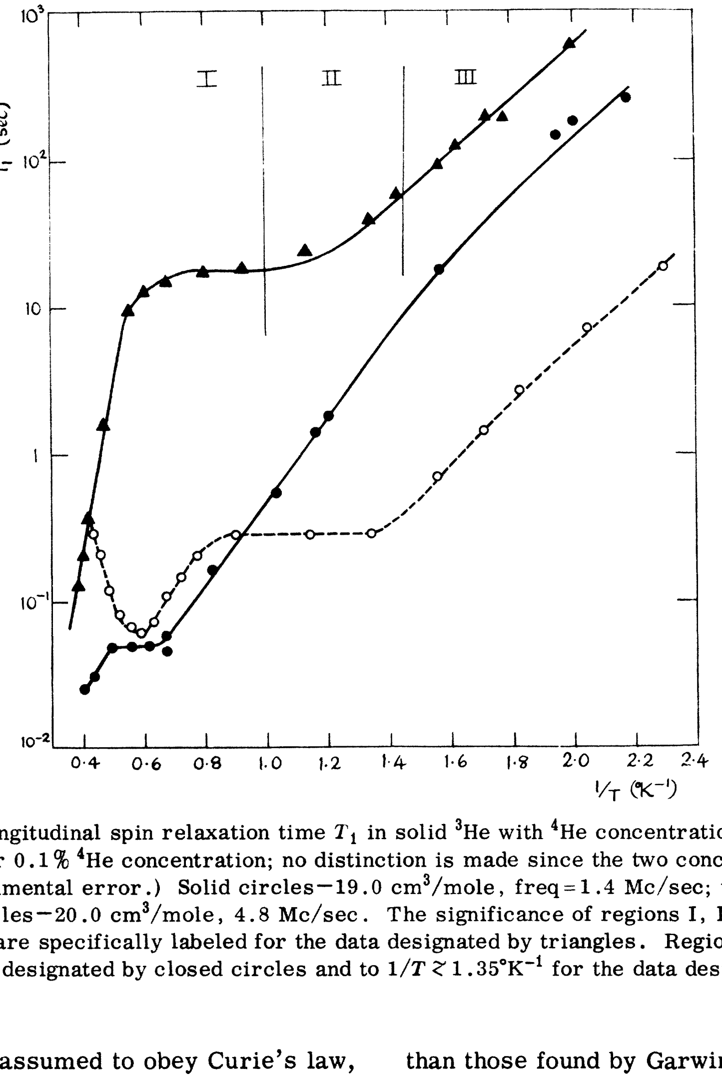

FIG. 2. Observed longitudinal spin relaxation time T_1 in solid ^{3}He with ^{4}He concentration $\leq 0.1\,\%$. (Data points are for either 0.05% or 0.1% ^{4}He concentration; no distinction is made since the two concentrations give identical results to within experimental error.) Solid circles—19.0 cm^3/mole, freq = 1.4 Mc/sec; triangles—19.0 cm^3/mole, 3.15 Mc/sec; open circles—20.0 cm^3/mole, 4.8 Mc/sec. The significance of regions I, II, and III is explained in the text. The regions are specifically labeled for the data designated by triangles. Region I corresponds to $1/T \lesssim 0.65°\mathrm{K}^{-1}$ for the data designated by closed circles and to $1/T \lesssim 1.35°\mathrm{K}^{-1}$ for the data designated by open circles.

the susceptibility is assumed to obey Curie's law, we can obtain J by measuring the frequency dependence of T_1 in this region. We observe the frequency dependence predicted by (3a), and the resulting values for J may be found in Table I. These values are roughly a factor of two greater than those found by Garwin and Landesman from an analysis of the exchange-narrowed transverse relaxation time. According to Hartmann,[3] $J = 0.42\omega_L$ for the α phase; so we see from Table I that, on this basis, J is about 30% larger than would be expected from our observed ω_L.

Table I. Experimental results for 99.95% pure[a] ^{3}He.

Molar volume (cm^3/mole)	$\omega_L/2\pi$[b] (Mc/sec)	$J/2\pi$[c] (Mc/sec)	τ_0[d] (msec)	Δ[d] (°K)
19.0	0.78	0.37	6.25	4.5
20.0	1.2	1.2	0.11	4.5

[a] Samples 99.90% pure give the same results to within experimental error.

[b] See Eq. (4).

[c] See Eqs. (3) and (5); for 19.0 cm^3/mole there are 12 nearest neighbors (hcp), while for 20.0 cm^3/mole there are 8 (bcc).

[d] See Eq. (6).

The exchange-lattice relaxation time τ_{XL} has an exponential temperature dependence

$$\tau_{XL} = \tau_0 \exp(\Delta/T) \qquad (6)$$

in region III, and the results for τ_0 and Δ are shown in Table I.

In the intermediate region II where $\rho\tau_{XL} \approx \tau_{ZX}$ it should, in principle, be possible to see signal recoveries consisting of two exponentials. However, in terms of the values for ρ, τ_{ZX}, and τ_{XL} inferred from regions I and III, it turns out that it is not possible for us to observe the shorter of the two relaxation times with our present apparatus.[5] This conclusion is based on a detailed solution[6] of the rate equations which will not be repeated here. It shows that we would have to saturate the signal in a time short compared to about 50 milliseconds in order to see the faster exponential. Hence the fact that we have single-exponential recoveries in region II is not inconsistent with the three-bath model when account is taken of our present electronic limitations.

However, if there were a phase transition in region II so that both the α and β phases were present, as was supposed by Reich, then we certainly ought to have observed nonexponential recoveries. (We have, in fact, found nonexponential recoveries at other molar volumes where the α-β phase transition is known to occur from the data of Grilly and Mills.[7]) The fact that we have not been able to observe nonexponential recoveries at a molar volume of 20 cm^3 in region II is, we believe, strong evidence against the mixed-phase hypothesis put forward by Reich to explain his nonexponential recoveries at 20.12 cm^3/mole and 21.10 cm^3/mole (curves *E* and *D* of Fig. 6, reference 1). Since he used pulsed nmr, it is perhaps possible that he was instead observing the two relaxation rates appropriate to the three-bath model.[5]

In samples containing 0.5% ^{4}He concentration we do see nonexponential recoveries for all temperatures below about 1°K. Above 1°K the results agree with those obtained in the purer samples. Between about 1°K and 0.7°K the recoveries can be described by two exponentials. A typical case is shown in Fig. 3, where in the lower curve (*A*) the high rf power used to saturate the signal was applied for a relatively short time, and in curve (*B*) the saturating period was considerably longer. That the signal takes longer to recover in (*B*) than in (*A*) is a characteristic feature of the three-bath model: The rate of recovery depends on the initial temperature T_X of the system *X*, and this can be raised well above the lattice temperature during a long saturation. To our knowledge direct observation of this effect has not previously been reported in the literature of magnetic relaxation. We emphasize that the lattice heat capacity is, of course, very much larger than that of the nuclear spins, and any variation in lattice temperature during the course of a saturation or recovery is completely negligible.

Analysis[6] of the rate equations enables us to compute ρ, τ_{ZX}, and τ_{XL} in terms of the two observed exponentials and the proportion in which they combine.[8] This can be done either for conditions appropriate to a very short period of saturation (curve *A*) or for those appropriate to a long period of saturation (curve *B*). The results are quoted in Fig. 3, and it may be seen that the two curves give roughly the same values for ρ, τ_{ZX}, and τ_{XL}, thus showing the internal consistency of the three-bath model.

In general, we find that τ_{ZX} is independent of ^{4}He concentration between 0.05% and 0.5% but that ρ is an order of magnitude less in the samples with 0.5% ^{4}He. Thus the addition of ^{4}He impurities appears to increase the heat capacity of the intermediate system by an order of magni-

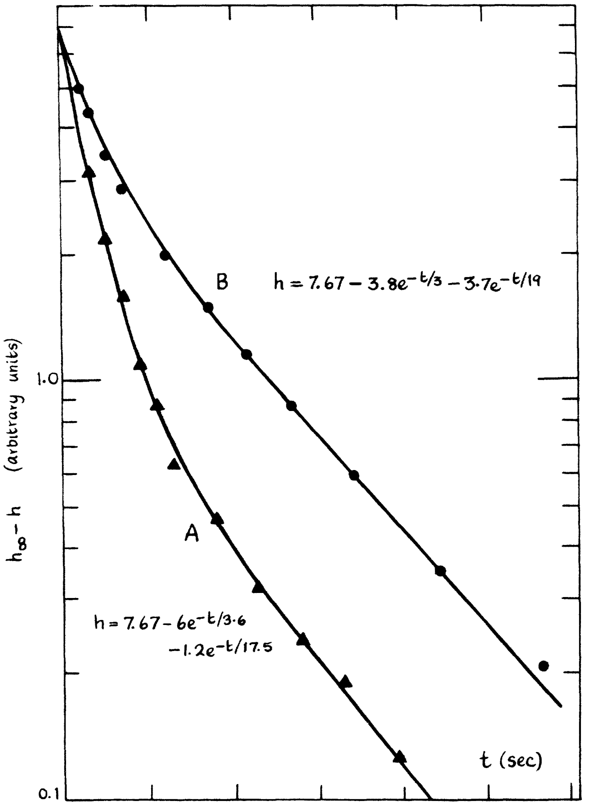

FIG. 3. Recovery of signal height h—subtracted from equilibrium height h_∞—following two different periods of saturation. Data are for molar volume of 19.0 cm^3, frequency 2.71 Mc/sec, $T = 0.71$°K, and ^{4}He concentration of 0.5%. (*A*), rf saturating power applied for 2 seconds; (*B*), rf saturating power applied for 36 seconds. The solid lines represent the best double-exponential fits to the data. From the observed decay constants (which should be identical in *A* and *B*) and the proportion in which they combine, we compute (*A*) $\tau_{ZX} = 4.18$ sec, $\tau_{XL} = 15.2$ sec, $\rho = 0.14$; (*B*) $\tau_{ZX} = 3.45$ sec, $\tau_{XL} = 16.1$ sec, $\rho = 0.11$. In (*A*) the temperature T_X of the intermediate system is equal to the lattice temperature T at $t = 0$, while in (*B*) $T_X \approx 1.5T$ at $t = 0$, where $t = 0$ is the time at which the rf saturation is turned off.

tude without altering the Zeeman-exchange relaxation rate. The quantity τ_{XL} is roughly independent of ^{4}He concentration at 1°K, but it is less strongly dependent on temperature—i.e., has a lower activation energy (6)—for the more impure samples.

We observe at least three relaxation times in the more impure samples below 0.7°K; so the three-bath model does not provide a complete description at the lowest temperatures. Because of this, we cannot perform a meaningful analysis of the nonexponential recoveries at the lowest temperatures. However, it may be noted that the difference between recoveries following short and long saturation periods is even more marked at these low temperatures than shown in Fig. 3. The mechanism for this additional bottleneck is at present unknown to us. It occurs at higher temperatures than those for which the ^{3}He-^{4}He phase separation has been observed.[9]

An extensive program of measurement is currently in progress.

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[1] H. A. Reich, Phys. Rev. 129, 630 (1963).

[2] R. L. Garwin and A. Landesman, Phys. Rev. 133, A1503 (1964).

[3] S. R. Hartmann, Phys. Rev. 133, A17 (1964).

[4] J. Hatton and B. V. Rollin, Proc. Roy. Soc. (London) A199, 222 (1949).

[5] With some electronic modifications we have now been able to observe nonexponential recoveries in region II for a molar volume of 20.6 cm^3. The recoveries are similar to those shown in Fig. 3, having the characteristic features of the three-bath model and not of a mixed phase. Values of ρ obtained from an analysis of the double-exponential recovery are in rough agreement with the observed frequency dependence of T_1 in the region where Eq. (4) is applicable.

[6] To be published.

[7] E. R. Grilly and R. L. Mills, Ann. Phys. (N.Y.) 8, 1 (1959).

[8] We assume in this analysis that the direct Zeeman-lattice relaxation rate τ_{ZL}^{-1} is negligibly slow at temperatures ($\lesssim$1°K) for which nonexponential recoveries are observed.

[9] D. O. Edwards, A. S. McWilliams, and J. G. Daunt, Phys. Letters 1, 218 (1962); Phys. Rev. Letters 9, 195 (1962).

DIFFERENTIAL MOTION OF EXCITED He$^+$ IONS IN A HOLLOW CATHODE PLASMA*

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In a recent study[1,2] of the HeII (3*s*,*p*,*d*) - (4*s*,*p*, *d*,*f*) line complex excited in a liquid helium-cooled hollow cathode plasma at pressures between 0.12 and 0.06 Torr and current densities of about 5 mA/cm^2, an important shift of the position of the lines was discovered. It appears that the most likely origin of the shift of the lines is a Doppler effect caused by a drift of the ions in the direction of the small axial electric field in the plasma. A comparison of measurements of the relative line positions with the calculated positions and with those measured by Series[3] indicates a possible differential shift apparently arising from different drift velocities for ions in the 4*s*, 4*p*, 4*d*, and 4*f* states. Because of the importance of this type of shift for spectroscopic measurements, and because of its potential use as a tool for studying the hollow cathode plasma and atomic properties, an investigation of this effect has been begun. This note is a report of the preliminary experiments.

The discharge tube consisted of three cylindrical copper electrodes held coaxially one above the other by insulating glass spacers. The assembly was closed at the bottom by a glass light trap, and attached at the top to a 40-cm long conical tube with an end window for viewing the plasma along the axis of the electrodes. The middle electrode, which had an 8-cm bore, was the cathode within which the plasma was contained. The two identical, symmetrically placed extremal electrodes were alternately made anodes so that the plasma could be viewed with the electric field either toward or away from the observer. The discharge was operated under conditions similar to those of the first study.