Spin relaxation in liquid ^{3}He

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We use an rf SQUID to measure the nuclear spin-lattice relaxation time T_1 in liquid ^{3}He for temperatures from 0.04 to 0.7 K at zero pressure and for different degrees of filling of the experimental cell. We identify the contributions to T_1 caused by the relaxation at the walls of the cell and by the relaxation in the bulk of the liquid. For temperatures below about 0.02 K the bulk relaxation time is larger than in other experiments. For temperatures above about 0.17 K the relaxation rate in an incompletely filled cell increases due to the appearance of vapor above the liquid; this transports magnetization to the "ceiling" of the cell.

INTRODUCTION

The spin-lattice relaxation of nuclei in liquid ^{3}He in a three-dimensional geometry has been studied in many papers.[1-6] The relaxation occurs at the walls of the experimental cell and inside it. The intrinsic, or bulk, relaxation is achieved through the dipole-dipole interaction between spins, modulated by the diffusive motion of the atoms. The experimental determination of the bulk relaxation time T_b makes it possible to check the theoretical predictions of Ref. 7. Experiments have been carried out[2-4] at temperatures $T > 0.4$ K where the contribution from the walls is small and T_b is close to the observed T_1. At lower temperatures the rate of the intrinsic relaxation decreases, approximately following a T^2 law,[5,7] while the relaxation at the walls increases as T^{-1} so that direct measurements of T_b are difficult at low pressures. When the pressure increases T_b decreases; experiments in which we have $T_b \approx T_1$ for pressures of 28 atm and with $T < 0.2$ K were carried out in Ref. 6. The time $T_b(T)$ was determined at low pressures and with $T < 0.6$ K in experiments by Beal and Hatton[1] and by Godfrin et al.[5] These authors used in their analysis of the experimental data a formula from the Bloembergen-Purcell-Pound theory:[9] $T_b = CD/\rho$, where D is the spin diffusion coefficient and ρ the liquid density. The wall relaxation time, T_w, is assumed to be proportional to $1/D$ and the constant C is determined from the slope in a $\rho/D^2 - 1/DT_1$ plot. In the experiments of Ref. 1 the main contribution to the relaxation comes from the walls and the values of T_1 did not exceed 180 s at any temperature. The spin-lattice relaxation is in Ref. 5 essentially determined by the bulk mechanism, $T_1(T = 0.1$ K$) \approx 1000$ s. We have carried out measurements with different amounts of ^{3}He in the cell; the observed values of T_1 were close to those given in Ref. 5 with $T_1(T = 0.1$ K$)$ equal to 800–900 s for cells with a degree of filling k equal to 1 and 0.75. The contribution from the walls depends on the ratio between the volume of the liquid and the area of the surface on which relaxation occurs and it can be isolated from the measured $T_1(T,k)$ functions and one can determine the bulk relaxation time. Experiments with different fillings of the cell below 25 mK are given in Ref. 10.

EXPERIMENTAL PROCEDURE

We show in Fig. 1 the cell for the ^{3}He used in our experiments. The cold-conductor of the cell was screwed to a copper disc of a ^{3}He-^{4}He solution refrigerator. The magnetic field H_0 was produced by a short-circuited superconducting solenoid with an improved field uniformity.[11] The temperature was controlled by two resistance thermometers: one was screwed onto the cold pipe of the cell and the other to the copper element of the upper part of the cell; the first one operated the temperature control. The thermometers were calibrated by the vapor pressure of ^{3}He and by the magnetic susceptibility of cerium-magnesium nitrate with an accuracy of about 3%. For the cooling of the liquid ^{3}He we placed in the lower part of the cell a copper needle plated with an indium-lead alloy and a sintered copper powder with a 0.14 m^2 surface area as determined by the absorption of ^{4}He. To make the walls of the cell smooth the inner part of the cell was covered twice by Stycast-1266 epoxy resin; in the lower part of the measuring volume which originally had a cylindrical shape with a 7.8 mm diameter and a 4.0 mm height a resin "deposit" was formed. The geometric area of the walls of the measuring volume was about 1.4 cm^2 and out of that the area S of the "ceiling" of the cell was 0.45 cm^2. The total
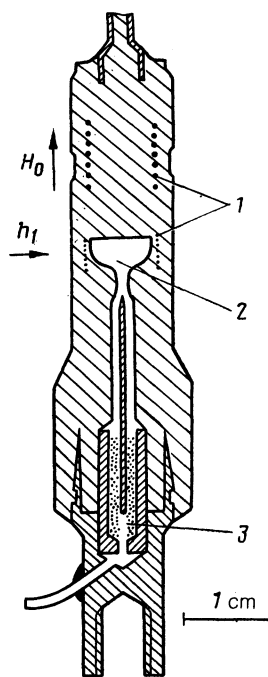

FIG. 1. The ^{3}He cell: 1: the facing collecting coils of the SQUID; 2: measuring volume; 3: sintered copper powder.
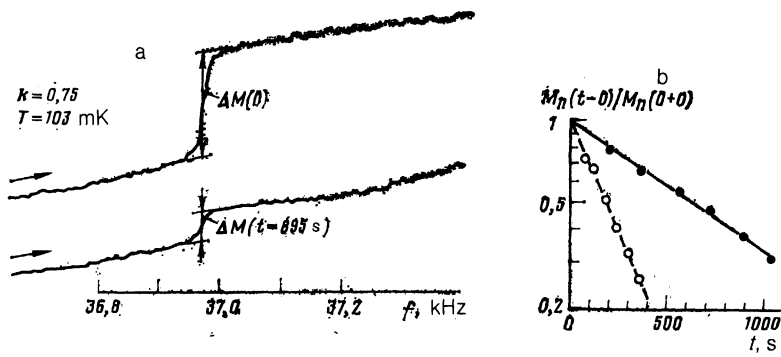
FIG. 2. a) Example of recording two successive passages through the resonance line along the vertical axis, the SQUID output. b) Graph showing the relaxation of the deviation M_n of the magnetic moment from its equilibrium value, which is used to determine T_1: O: $k = 0.75$, $T = 569$ mK, $T_1 = 266$ s; ●: $k = 0.75$, $T = 103$ mK, $T_1 = 910$ s.

volume of the cell was 0.37 cm^3 and the measuring volume V was 0.12 cm^3.

We carried out experiments with a degree of filling k of the measuring volume equal to 1, 0.75, and 0.50, which was determined by the volume of the condensed gas and calculated from the magnitude of the magnetic moment of the liquid. The contribution to the magnetic flux through the receiver spools of the SQUID from the ^{3}He in the filled channel of the cell, estimated from geometric considerations, was less than 1 to 2% of the signal from the measuring volume. The filling line of the cell passed through a trap with activated carbon at 77 K. Immediately before the experiments and the start of the cooling the experimental ^{3}He was admitted two or three times and then removed. The fraction of ^{4}He in the gas was 1.5×10^{-4}. Normally the cell was filled with liquid at the start of an experiment at $T \approx 1$ K. The pressure P in the cell for $k = 1$ was less than 10 to 15 Torr. The gas was collected at the end of the experiments by a small cryopump with activated coal.

The method for measuring the magnetic moment using a SQUID is similar to the one described in Ref. 12. Nuclear magnetic resonance was observed by the change in the longitudinal magnetization of the liquid in an adiabatically fast passage. The oscillating field h_1 with an amplitude of 4 mOe was switched on for about 40 s and its frequency was changed with a speed of 25 Hz/s. To determine T_1 we used a sequence of two such passages, reversing the magnetization (Fig. 2). From the magnitude of the change $\Delta M(0)$ in the magnetic moment in the initial passage and $\Delta M(t)$ in the passage after a time t we calculated the deviation M_n of the

magnetic moment from its equilibrium value M_0 immediately after the initial passage and immediately before the second one:

$$M_n(0+0) = 2M_0 - M_n(0-0),$$

$$M_n(t-0) = M_0 \pm \Delta M(t)/2,$$

$$M_n(t+0) = M_0 \mp \Delta M(t)/2,$$

$$M_n(0-0) = M_n(t+0)_{pr} \exp(-t_{pr}/T_1),$$

$$M_0 = \langle M_n(0-0) + \Delta M(0)/2 \rangle.$$

In the expressions for $M_n(t-0)$ and $M_n(t+0)$ one chooses the upper sign, if the direction of the change in the magnetization in the passage after the time t is the opposite of the direction of change in the initial passage (for small t). In the case shown in Fig. 2 we choose the lower sign. $M_n(t+0)_{pr}$ is the deviation of the magnetic moment from the equilibrium value at the end of the sequence preceding the one considered. The time t_{pr} between sequences was chosen in the experiment to be equal to $(2.5-3) T_1$. The correction $M_n(0-0)$ describes the weak deviation of the magnetization from the equilibrium value before the initial passage; T_1 is determined from two to three iterations. For the chosen values of T and k we carried out four to six sequences of two passages and averaged over them in the expressions for M_0.

We show in Fig. 3 the values of T_1 determined in this way. The measurements were carried out on different days with heating up to room temperature after each experiment; two to five $T_1(T,k)$ points were determined in a single experiment. Most of the experiments were carried out at a resonance frequency of 37 kHz ($H_0 = 11.4$ Oe), some at a fre-
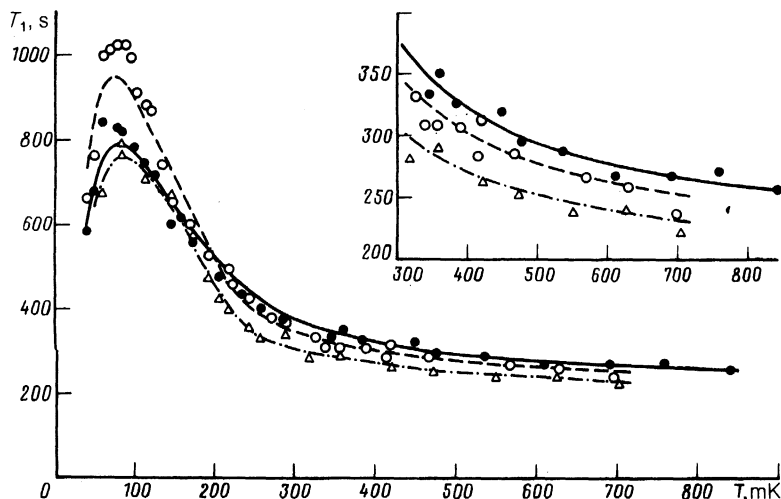

FIG. 3. Experimental T_1 points and curves calculated using Eqs. (1) to (4) for different values of k: ● and solid curve: $k = 1$; O and dashed curve: $k = 0.75$; △ and dash-dot curve: $k = 0.50$.

quency of 18 kHz. At $T \approx 70$ mK the time T_1 was determined at lower frequencies down to 2.5 kHz. Within the accuracy of the measurements T_1 was independent of the frequency. We note that a weak dependence of the rate of the surface relaxation on the resonance frequency for $f \lesssim 40$ kHz was found in Refs. 13 in experiments with a restricted geometry.

VAPOR ABOVE LIQUID ^{3}He

The results of the measurements of T_1 (T,k) shown in Fig. 3 make it possible to distinguish two temperature ranges. For $T < 170$ mK the rate of the longitudinal magnetic relaxation in the case of a full measuring volume ($k = 1$) is larger than for $k = 0.75$ and close to the $k = 0.50$ case. At those temperatures the vapor above the liquid has a negligibly small density and the ceiling of the cell does not contribute to the relaxation on the walls for $k = 0.50$ and 0.75. For $T > 300$ mK the value of T_1 (T,k) increases as a function of k. In that case the density of the saturated ^{3}He vapor is large and the nonequilibrium magnetization of the liquid is transported to the ceiling of the cell by the diffusion of atoms. The upper part of the measuring volume is covered by a film of the liquid with a thickness of several hundred atomic layers and all walls of the measuring volume take part in the relaxation. The transport of the magnetization through the vapor is diffusive in nature if the mean free path of an atom in the vapor is much shorter than the distance from the surface of the liquid to the ceiling. These lengths are comparable for a saturated vapor pressure of $\approx 2 \times 10^{-4}$ Torr; here we have used the estimate of the scattering cross-section for ^{3}He atoms from Ref. 14: an atomic diameter of ≈ 2 Å. Such a pressure corresponds to a temperature of 0.25 K.

At lower temperatures the transport of the magnetization through the vapor is determined by the number of atoms colliding with the surface of the liquid, the reflection coefficient in that case being close to zero.[15] In calculating the function T_1 (T,k) for $T > 170$ K we assumed that the time for the transport of the magnetization through the vapor is the sum of the time determined by the diffusion of the atoms in the gas and the time set by the rate at which the atoms collide with the surface of the liquid, which becomes negligibly short for $T > 300$ mK. The rate of the bulk relaxation of the spins in gaseous ^{3}He is small[16] and does not contribute to T_1.

DETERMINING THE TIME FOR RELAXATION IN THE BULK OF THE LIQUID AND AT THE WALLS

The rate of the relaxation of the magnetization can be written in the form

$$\frac{1}{T_1} = \frac{1}{T_b} + \frac{1}{k_w T_w + T_D} + \frac{AD}{VLk}. \qquad (1)$$

The small third term on the right-hand side corresponds to spin diffusion along the filling channel of the measuring volume; A and L are the area of the cross-section and the length of the channel and we have $A/VL \approx 0.2$ cm^{-2}. The second term on the right-hand side of (1) corresponds to the contribution from the walls; T_w is the intrinsic wall relaxation time in a full cell; the coefficient k_w is proportional to k and inversely proportional to the area of the walls involved in the relaxation, and for $k = 1$ we find $k_w = 1$; the time T_D describes the diffusion of the magnetization to the walls. In our

conditions T_D is at most $(0.1-0.15)$ T_w and the nonexponential behavior of the relaxation which is connected with the diffusion is in that case small.

For $T > 300$ mK diffusion in the vapor above the liquid transports the magnetization to the ceiling of the cell and we have $k_w = k$. The coefficient for the diffusion of the spins through the vapor is $D_e = D_{V\rho V}/\rho T\chi$ where D_V and ρ_V are the spin diffusion coefficient and the density of the vapor, and $\chi(T)$ is the magnetic susceptibility of the ^{3}He nuclei, expressed in units of the constant in the Curie law.[17] The time T_D was for $k = 1$, 0.75, and 0.50 calculated from the data for the spin diffusion coefficients for the liquid[5,18] and the vapor:[19] we have $D_e/D = 1.1$ for $T = 0.5$ K; the measuring volume was assumed to be cylindrical. The experimental points T_1 (T) for $k = 1$, 0.75, and 0.50 and $T > 300$ mK were approximated by smooth curves and from them we calculated T_b (T) and T_w (T) using (1) in the narrower temperature range $350 < T < 650$ mK. We estimate the accuracy of the determination of T_b (T) in that temperature range to be about 10%.

For $T < 170$ mK there is no vapor above the liquid ^{3}He; k_w depends on the area of the walls which are in direct contact with the liquid. We looked for T_b and T_w in the form

$$T_b = B_2/T^2 + B_1/T + B_0, \qquad T < 650 \text{ mK}, \qquad (2)$$
$$T_w = 1/(W_1/T + W_0), \qquad T < 170 \text{ mK}. \qquad (3)$$

The first term in (2) describes T_b (T) at low temperatures corresponding to a degenerate Fermi liquid.[5,7] Equation (3) uses the results of measurements of the surface spin relaxation time of liquid ^{3}He,[5,8] according to which we have $T_w(T) \propto T$ for $T \lesssim 0.1$ K while it changes slowly with T at higher temperatures. The quantities B_2, W_1, W_0, $k_w(k = 0.75)$, and $k_w(k = 0.50)$ were found by a best fit of (1) with the measurements of T_1 for $T < 170$ mK; B_1 and B_0 are then determined so that at higher temperatures the function T_b (T) calculated using (2) goes over into the function found for $T > 350$ mK. The result of such calculations is $B_2 = 17.1$ s·K^2, $B_1 = 17$ s·K, $B_0 = 273$ s. The values $k_w(k = 0.75) = 1.38$ and $k_w(k = 0.50) = 1.04$ found here are close to the estimate using the geometry of the measuring volume. Two independent methods to determine the wall relaxation time for the temperature ranges $T < 0.17$ K and $0.35 < T < 0.65$ K agree, when they are extrapolated into the intermediate range, to within about 20%.

For $170 < T < 300$ mK the additional time needed to transport the magnetization through the vapor is determined by the number of atoms colliding with the surface area S of the liquid:

$$T_V = (4\rho V k T\chi/P_V S)(\pi R T/8\mu)^{1/2}. \qquad (4)$$

Here μ is the molar mass of ^{3}He, R the molar gas constant, and $P_V(T)$ the vapor pressure. Using for $P_V(T)$ the 1976 temperature scale, extrapolated below 0.2 K, we find $T_V(T = 170 \text{ mK}, k = 0.5) = 3 \times 10^4$ s; this value is about an order of magnitude larger than the relaxation time of the spins on the ceiling of the cell.

To calculate the functions T_1 (T) with $k = 1$, 0.75, and 0.50 (Fig. 3) from (1) we used Eq. (2) for the bulk relaxation time with the coefficients found here; the wall relaxation time in the temperature range $170 < T < 350$ mK was ob-

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tained by "matching" Eq. (3) with T_w found for $T > 350$ mK.

EXPERIMENTS INVOLVING FILLING OF THE CELL AT LOW TEMPERATURES AND INVOLVING ADDING ⁴He TO THE ORIGINAL GAS

The ⁴He contained in the original gas ($1.5 \times 10^{-2}\%$) can lead to coating of the walls of the measuring volume and the surface of the sintered powder in the lower part of the cell with a thickness of about 0.6 monatomic layer; for this estimate we used a value of the monolayer density of 0.11 atoms per Å² given in Ref. 16. The ⁴He on the surfaces causes the wall spin relaxation of liquid ³He to slow down.[5] To check the effect of ⁴He impurities on T_w we performed two series of experiments.

In separate experiments the filling coefficient k was increased from 0.75 to 1 during the experiment without heating the cell. For $T > 250$ mK the values of T_1 obtained were the same as the results of the main set of experiments, while they were smaller for $T < 200$ mK. This is possibly connected with a different thickness of the ⁴He layer on the ceiling of the cell. For $k = 1$ we performed measurements of T_1 for $50 < T < 210$ mK with a fraction of about 30% of ⁴He in the original gas. The experimental data are shown in Fig. 4. The ⁴He concentration in the bulk of the liquid above a demixed solution of a helium isotope mixture is 0.6% at $T = 0.21$ K (Ref. 20) and it decreases rapidly when the temperature is lowered. Thus, while the measuring volume can be assumed to be filled with liquid ³He, its walls are covered by a film of a dilute phase of the ³He-⁴He mixture. We note that in these experiments the temperature was rapidly—over a time less than 2 to 5 minutes—equalized in the cell when it was cooled down, which is clearly connected with the film on the walls. In the case of pure ³He the time needed to establish thermal equilibrium was about 1.5 hr at $T = 75$ mK. For $T \lesssim 120$ mK the measured values of T_1 were appreciably larger than those obtained in the main series of experiments. The values of the wall relaxation calculated from $T_b(T)$, which had been determined before, using (1) are shown for these cases in Fig. 5.
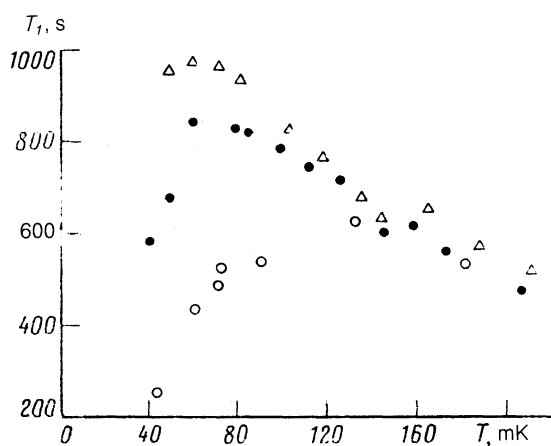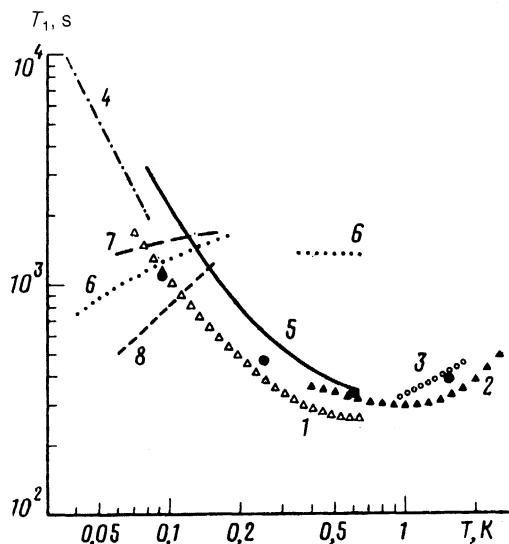
FIG. 5. Bulk relaxation time, experiments: 1: extrapolation from Ref. 1 to $P = 0$; 2: Ref. 2; 3: Ref. 3, ●: T_1 measurements with saturated vapor pressures;[5] 4: theory,[7] extrapolation to $P = 0$; 5: result obtained in the present paper, calculations using (2) with the given coefficients. Wall relaxation times: 6: main set of experiments, $T < 170$ mK and $T > 350$ mK; 7: filling of the cell from $k = 0.75$ to $k = 1$ during the experiment; 8: ⁴He added to the original gas.

DISCUSSION OF THE RESULTS

The function $T_b(T)$ found here is shown in Fig. 5. In Eq. (2) we used the minimum number of terms, and it cannot be extrapolated above 0.65 K. In particular, (2) does not describe the minimum of $T_b(T)$ at about 0.8 K (Fig. 5).[1-3] We estimate the accuracy of the result for T_b to be about 20% for $T < 0.2$ K. The possibility of spin relaxation of ³He due to the presence of impurities suspended in the liquid was discussed in Ref. 1. The reproducibility of the T_1 measurements performed in different experiments shows the small effect of impurities; the function $T_b(T)$ obtained here can be considered to be the lower limit for the bulk relaxation time. The values of $T_b(T)$ found for $T < 0.2$ K are higher than the data of Refs. 1–5 by a factor of 1.5–1.8 while $T_b(T)$ is the same as the results of Refs. 2, 3, and 5 at higher temperatures.

The authors of Refs. 1–5 compare the measured values of T_1 with those predicted by the Bloembergen-Purcell-Pound theory which describes spin relaxation in liquids:[9]

$$1/T_b = 0{,}4\pi\gamma^4\hbar^2 n/Dd,$$

where γ is the gyromagnetic ratio, n the number of spins per unit volume, and d the distance of closest approach of the atoms which usually is assumed to depend only weakly on the temperature and the liquid density. Calculating the magnitude of d from the function $T_b(T)$ obtained in the present paper and the value of $D(T)$ from Refs. 5 and 18 leads to 3.2 Å for $T = 0.1$ K and 2.4 Å for $T = 0.6$ K. From the data of Ref. 5 we have $d = 2.1 \pm 0.1$ Å for $0.1 < T < 1.5$ K; a calculation using the measurements of T_1 from Ref. 3 gives $d = 2$ Å for $1 < T < 1.8$ K. Therefore, the value of d possibly decreases with increasing temperature.

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FIG. 4. T_1 measurements, $k = 1$: ●: main series of experiments, filling of the ³He cell at $T \approx 1$ K; O: filling of the measuring volume from $k = 0.75$ to $k = 1$ during an experiment without heating the cell; △: experiments with a ³He-⁴He mixture.

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