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New Surface Relaxation Mechanism for Liquid ^3He in ^4He

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Measurements of the nuclear magnetic spin-lattice relaxation time T_1 of liquid ^3He containing 0.5% ^4He and of ^3He - ^4He mixtures as a function of magnetic field up to 22 T and at temperatures down to 40 mK show a new surface relaxation mechanism for liquid ^3He , which is proportional to the square of the magnetic field and can be described by classical relaxation theory. The intrinsic relaxation time of liquid ^3He , obtained from T_1 measurements by eliminating the surface relaxation contribution, is in good agreement with existing Fermi liquid theory. [S0031-9007(97)03546-1]

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Because of its simple atomic structure, liquid ^3He is a thoroughly studied model system for correlated fermions and for nuclear magnetic relaxation. Since the intrinsic relaxation time T_{in} is determined by the interactions between the nuclear spins, a study of T_{in} is like neutron scattering a probe of the dynamics of the Fermi liquid. However, the spin-lattice relaxation time T_1 measured in an experiment is determined both by T_{in} and by the surface relaxation T_s due to interactions of the nuclear spins with magnetic moments at the unavoidable surfaces of the experimental cell. Therefore T_{in} as a function of temperature cannot be determined unambiguously from T_1 [1–3], as both T_s and T_{in} are *a priori* unknown and temperature dependent. In this Letter we present measurements of T_1 of liquid ^3He with 0.5% ^4He and ^3He - ^4He mixtures as a function of the magnetic field up to 22 T. These data allow unambiguous determination of T_{in} and a detailed study of the surface relaxation time T_s , which is clearly found to be described by the classical Bloembergen, Purcell, and Pound theory [4], an observation not reported earlier.

We have chosen to study ^3He containing ^4He , since in this system the surface is covered with a thin layer of ^4He (even if the ^4He concentration is as low as 0.5%), which leads to much longer relaxation times than in pure ^3He [3], where a solid ^3He layer is formed at the surface, leading to a linear temperature and magnetic field dependence of T_1 [5]. Moreover, surface relaxation of ^3He in ^4He plays an essential role in most of the recent experiments on ^3He , like, e.g., in ^3He - ^4He mixture films [6] and highly spin-polarized ^3He in ^3He - ^4He systems [7]. Therefore relaxation of *liquid* ^3He is best studied in ^3He systems containing ^4He .

In the experiment T_1 is determined from the exponential decay of the nuclear magnetization, which is measured with a torsional magnetometer after a rapid change of the magnetic field. The advantage of this new, nonresonant technique, compared to nuclear magnetic resonance, is that it can be used at arbitrary magnetic field. The intrinsic and surface contributions to the measured T_1 can be separated as a result of their different magnetic field

dependence. The values and temperature dependence of T_{in} of liquid ^3He determined from our experiments are in good agreement with Fermi liquid theory [8]. On the other hand, we have discovered that the surface relaxation of ^3He in the presence of ^4He is proportional to the square of the magnetic field, markedly different from the linear magnetic field dependence observed in pure ^3He [5]. This behavior has not previously been observed in a Fermi liquid and can surprisingly be described by the theory of Bloembergen, Purcell, and Pound [4] for nuclear magnetic relaxation in classical fluids. We relate the observed relaxation to the dynamics of the ^3He atoms in the ^4He film at the surfaces, where the ^3He spin relaxes due to interaction with the inhomogeneous magnetic field of the surface. The exact nature of this interaction, however, is still subject to discussion.

The torsional magnetometer, described earlier in [9], is made of Hysol [10] and consists of a cylindrical wheel with a 100 μm manganine torque wire through its axis and an eccentric hole. The wire is horizontally stretched between two supports, holding the wheel in a house. Rotation of the wheel can be measured with capacitors on the wheel and the house. The magnetometer is located in an experimental cell (2 cm diameter, 2 cm height) filled with 3 cm^3 liquid ^3He containing 0.5% ^4He or ^3He - ^4He mixture. We have coated the surface with polystyrene, by dipping all parts of the magnetometer and the cell in a polystyrene/toluene solution, in order to reduce surface relaxation by smoothening the cell surface and reducing its effective surface area. The cell is placed in the mixing chamber of a dilution refrigerator. The wall of the cell is a 200 μm thick Kapton foil which acts as heat exchanger between the cell and the surrounding mixing chamber. By placing the magnetometer off the center of the magnetic field in a field gradient, the magnetization of the ^3He in the hole induces a torque, measured as a rotation of the wheel. T_1 is determined from the exponential decay of the nuclear magnetization after a quick step (much shorter than T_1) of the magnetic field.

The T_1 of liquid ^3He containing 0.5% ^4He has been measured as a function of the magnetic field at temperatures

ranging from 40 to 750 mK at saturated vapor pressure, and the results for four different temperatures are presented in Fig. 1. T_1 shows a gradual change from a low field region, where it has a complex temperature and magnetic field dependence and increases with magnetic field to a high field region, where it approaches a magnetic field independent saturation value T_{in} . T_1 is always smaller than T_{in} due to surface relaxation, which is clearly suppressed by the magnetic field.

To investigate the surface relaxation process in more detail, we have measured T_1 of a saturated ^3He - ^4He mixture at 100 mK (7% ^3He) in three different experimental cells: cell 1 with a volume of 3 cm³, cell 2 with a volume of 2 cm³, and cell 3 also with a volume of 3 cm³ but not coated with polystyrene. All cells have approximately the same surface area of 40 cm². The results in Fig. 2 show that T_1 is always proportional to the square of the magnetic field and does not saturate even at the highest fields, indicating that T_{in} of the mixture at 100 mK is much longer than 10⁴ s. For the two coated cells (cell 1 and 2) T_1 is within experimental error proportional to the volume, while coating of the cell leads to an increase of T_1 by a factor of almost 5, showing that the measured T_1 is dominated by surface relaxation. T_s of the ^3He - ^4He mixtures shows the B^2 dependence both in coated and in uncoated cells, and hence this behavior does not depend on the details of the surface material.

In the classical theory of Bloembergen, Purcell, and Pound [4] for nuclear spin relaxation in fluids, the nuclear spin is subject to a random fluctuating perturbing magnetic field with amplitude $\langle\delta B^2\rangle^{1/2}$ and correlation time τ_c , resulting in a relaxation rate

$$\frac{1}{T_s} = \gamma^2 \langle\delta B^2\rangle \frac{\tau_c}{1 + (\omega\tau_c)^2}, \quad (1)$$

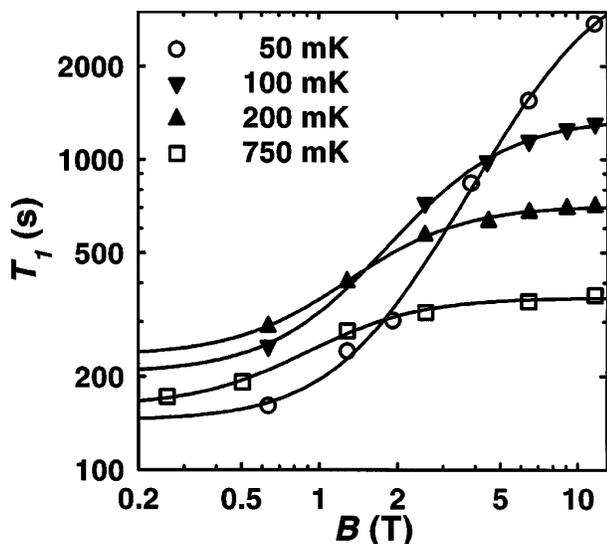


FIG. 1. Nuclear magnetic relaxation time T_1 of liquid ^3He - ^4He as a function of magnetic field. The curves are fits to the data with Eq. (3).

with ω the Larmor frequency and γ the gyromagnetic ratio ($\gamma = \omega/B \approx 32$ MHz/T for ^3He). Using this theory for the contribution of T_s to T_1 in our data, the right hand side of Eq. (1) must be multiplied by N_s/N_{tot} , the ratio of the number of spins involved in the surface relaxation process and the total number of spins which will have to relax. The atoms reach the surface by diffusion, where they have a certain probability to relax, and diffuse back into the liquid. Hence a diffusion time τ_D has to be added to T_s , as the two processes are in series, leading to our expression for the extrinsic relaxation time:

$$T_{ex} = \tau_D + \frac{N_{tot}}{N_s} \frac{1}{\gamma^2 \langle\delta B^2\rangle \tau_c} + \frac{N_{tot}}{N_s} \frac{\tau_c}{\langle\delta B^2\rangle} B^2. \quad (2)$$

The total relaxation time T_1 is determined by both the extrinsic and intrinsic relaxation processes in parallel; thus

$$\frac{1}{T_1} = \frac{1}{T_{ex}} + \frac{1}{T_{in}}, \quad (3)$$

with T_{ex} as defined in Eq. (2).

The field dependence of T_1 obtained with Eq. (3), with $\tau_D + N_{tot}/N_s \gamma^2 \langle\delta B^2\rangle \tau_c$, $N_{tot} \tau_c / N_s \langle\delta B^2\rangle$ and T_{in} as the only temperature dependent but field independent fitting parameters, is shown as solid lines in Fig. 1, and is in excellent agreement with the experimental data. This result implies that T_{in} is indeed independent of magnetic fields in the range of the experiment and that the field dependence of T_s of liquid ^3He containing ^4He is well described by Eq. (1). It should be stressed that the data cannot be fitted with a linear field dependence, which would not only give a bad fit but would also lead to unphysical negative relaxation times at zero magnetic field.

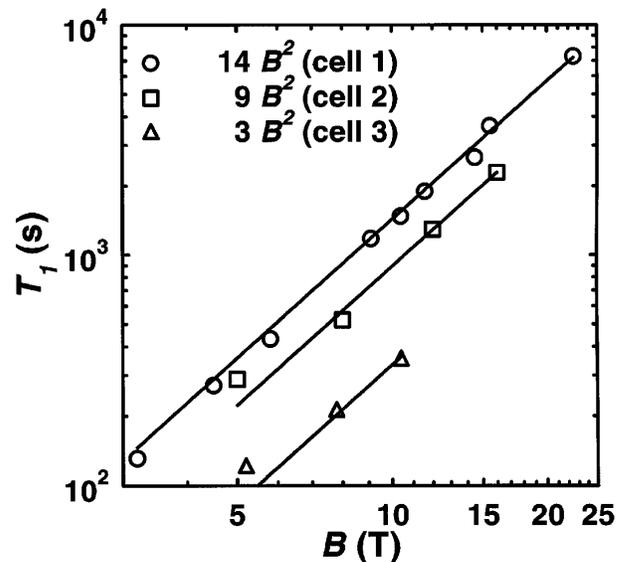


FIG. 2. Nuclear magnetic relaxation time T_1 of a saturated ^3He - ^4He mixture at 100 mK as a function of magnetic field and the quadratic fits for three different cells.

The correlation time τ_c directly results from the ratio of the two fit parameters $\tau_D + N_{\text{tot}}/N_s \gamma^2 \langle \delta B^2 \rangle \tau_c$ and $N_{\text{tot}} \tau_c / N_s \langle \delta B^2 \rangle$ if the diffusion time τ_D is estimated. Using the known diffusion coefficient for liquid ^3He [11] and approximating the sample volume of the magnetometer by a sphere, τ_D is found to increase from 20 s at 40 mK to 150 s at 200 mK. Using these numbers, τ_c is then found to increase from 20 ns at 40 mK to 50 ns at 200 mK. These numbers are reasonable, since a magnetic field of 1 T and a τ_c of 30 ns correspond to $\omega \tau_c = 1$, while we indeed see influence of the magnetic field on T_1 in this range of magnetic field.

Our T_1 data of the saturated mixture (Fig. 2) are essentially also described by Eq. (3). However, in the limit of long T_{in} , negligibly short τ_D and $\omega \tau_c > 1$, Eq. (3) reduces to the B^2 term of Eq. (2). An estimate of τ_D for the mixture is ~ 1 s at 100 mK [12], which is indeed negligible. $\omega \tau_c$ can be seen to be larger than 1 for fields of the order of 3 T indicating that τ_c is about 30 ns or longer. All other data of T_1 of ^3He - ^4He mixtures with both lower and higher ^3He densities, not shown here, were always well described by Eq. (3).

Comparing T_s of the concentrated ^3He and the saturated mixture, we observe that the ratio of the coefficients of B^2 from the T_1 measurements in the same experimental conditions is 13, which is within the experimental error of 10% equal to the ^3He density ratio of the concentrated ^3He and the saturated ^3He - ^4He mixture, which is 14. Since both systems are described by Eq. (3), this result shows that the number of spins near the surface N_s is independent of the ^3He density in the cell.

The field dependence of T_1 (Figs. 1 and 2), shows that surface relaxation is not infinitely fast, as assumed by Low and Rorschach [2]. T_{ex} is therefore not determined by τ_D , contrary to the previous assumption, which is often used to extract T_{in} from T_1 measurements as a function of temperature [1–3]. Since T_{ex} is not found to be equal to τ_D , this naturally explains why T_1 in ^3He - ^4He mixture films does not scale with diffusion coefficient D [6].

The last parameter determined with Eq. (3) from T_1 measurements of ^3He containing 0.5% ^4He as a function of the magnetic field is T_{in} . The results are plotted in Fig. 3 as a function of temperature together with the calculated T_{in} by Vollhardt and Wölfle [8]. Excellent agreement between experiment and theory is observed in the Fermi liquid regime (below 200 mK), both in temperature dependence and in absolute value, showing the validity of our previous analysis. In the high temperature plateau, the experimentally determined T_{in} is 1.5 times longer than the theoretical value, in agreement with previous T_1 measurements of Romer [13]. Moreover, above 100 mK the contribution of the wall relaxation to T_1 at the highest fields was so small, that T_1 could reach T_{in} within experimental error of about 10%, which implies that the theory underestimates T_{in} with about 50% at the high temperature plateau.

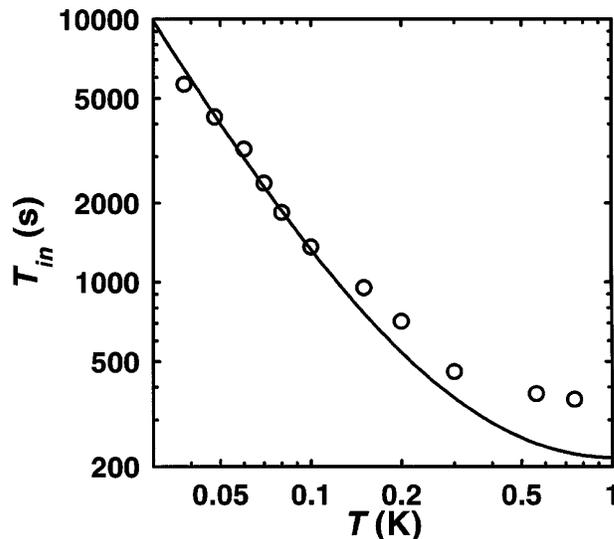


FIG. 3. Intrinsic relaxation time T_{in} of liquid ^3He as a function of temperature determined with Eq. (3). The solid line represents the theory of Ref. [8].

We have shown that surface relaxation of ^3He in the presence of ^4He depends on B^2 as described by Eq. (1). This observation is in sharp contrast to the generally seen linear B dependence of T_s in ultrapure ^3He samples with solid ^3He at the surface [5]. The difference between these two systems is that the ^4He replaces the ^3He at the surface, forming a superfluid ^4He film on top of a solid ^4He layer at the surface of the cell [14,15]. Because of the finite solubility of ^3He in this film, the system near the surface should be the same for a saturated mixture as for liquid ^3He containing a fraction of ^4He to cover the surfaces, which explains why the observed surface relaxation is independent of the ^3He density in the cell. This finite solubility also explains the observation in ^3He - ^4He mixture films that after a certain coverage of the surface with ^4He , T_1 becomes nearly independent of the ^4He coverage [15].

A possible mechanism for the observed surface relaxation process is that a ^3He spin relaxes due to diffusional motion in the inhomogeneous magnetic field of the surface. In this case, $\tau_c = l^2/\pi^2 D$, with l the correlation length of the inhomogeneous field and D the diffusion coefficient in the ^4He film. Taking for D the bulk value for a 5% ^3He - ^4He mixture [12], the observed τ_c of 30 ns corresponds to a correlation length of the order of $0.5 \mu\text{m}$, which is about 10 times larger than the thickness of the ^4He film [14]. This correlation length could be related to the surface roughness. The observed increase of τ_c with temperature is consistent with a decrease of D with temperature [12]. This surface relaxation mechanism might also be responsible for the observed relaxation in the liquid ^3He layers on top of the solid ^3He layer [16]. Another mechanism might be that a ^3He atom sticks to the surface

and relaxes due to the perturbing field of a paramagnetic impurity at the surface of the cell. N_s would then correspond to the number of ^3He atoms at the surface, and τ_c to the sticking time. This sticking behavior has also been observed in ^3He gas in hydrogen coated cells in the low field limit [17], while at higher fields the B^2 dependence was observed but not attributed to the surface relaxation process [18].

In conclusion, we have shown that surface relaxation of liquid ^3He containing ^4He , unlike when there is a solid layer of ^3He at the surface, is proportional to $1 + (\gamma B \tau_c)^2$. We relate the observed relaxation to the dynamics of the ^3He atoms in the ^4He film at the surface. The correlation time τ_c , determined from our experiments, is of the order of 30 ns. The surface and intrinsic contributions to T_1 can be separated due to their different magnetic field dependence, and the intrinsic relaxation time of ^3He we have experimentally determined with high precision is in good agreement with the theoretical predictions by Vollhardt and Wölfle [8].

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