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## New Surface Relaxation Mechanism for Liquid $^3\text{He}$ in $^4\text{He}$

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Measurements of the nuclear magnetic spin-lattice relaxation time  $T_1$  of liquid  $^3\text{He}$  containing 0.5%  $^4\text{He}$  and of  $^3\text{He}$ - $^4\text{He}$  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  $^3\text{He}$ , which is proportional to the square of the magnetic field and can be described by classical relaxation theory. The intrinsic relaxation time of liquid  $^3\text{He}$ , 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  $^3\text{He}$  is a thoroughly studied model system for correlated fermions and for nuclear magnetic relaxation. Since the intrinsic relaxation time  $T_{\text{in}}$  is determined by the interactions between the nuclear spins, a study of  $T_{\text{in}}$  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  $T_{\text{in}}$  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  $T_{\text{in}}$  as a function of temperature cannot be determined unambiguously from  $T_1$  [1–3], as both  $T_s$  and  $T_{\text{in}}$  are *a priori* unknown and temperature dependent. In this Letter we present measurements of  $T_1$  of liquid  $^3\text{He}$  with 0.5%  $^4\text{He}$  and  $^3\text{He}$ - $^4\text{He}$  mixtures as a function of the magnetic field up to 22 T. These data allow unambiguous determination of  $T_{\text{in}}$  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  $^3\text{He}$  containing  $^4\text{He}$ , since in this system the surface is covered with a thin layer of  $^4\text{He}$  (even if the  $^4\text{He}$  concentration is as low as 0.5%), which leads to much longer relaxation times than in pure  $^3\text{He}$  [3], where a solid  $^3\text{He}$  layer is formed at the surface, leading to a linear temperature and magnetic field dependence of  $T_1$  [5]. Moreover, surface relaxation of  $^3\text{He}$  in  $^4\text{He}$  plays an essential role in most of the recent experiments on  $^3\text{He}$ , like, e.g., in  $^3\text{He}$ - $^4\text{He}$  mixture films [6] and highly spin-polarized  $^3\text{He}$  in  $^3\text{He}$ - $^4\text{He}$  systems [7]. Therefore relaxation of *liquid*  $^3\text{He}$  is best studied in  $^3\text{He}$  systems containing  $^4\text{He}$ .

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  $T_{\text{in}}$  of liquid  $^3\text{He}$  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  $^3\text{He}$  in the presence of  $^4\text{He}$  is proportional to the square of the magnetic field, markedly different from the linear magnetic field dependence observed in pure  $^3\text{He}$  [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  $^3\text{He}$  atoms in the  $^4\text{He}$  film at the surfaces, where the  $^3\text{He}$  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  $\mu\text{m}$  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  $\text{cm}^3$  liquid  $^3\text{He}$  containing 0.5%  $^4\text{He}$  or  $^3\text{He}$ - $^4\text{He}$  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  $\mu\text{m}$  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  $^3\text{He}$  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  $^3\text{He}$  containing 0.5%  $^4\text{He}$  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  $^3\text{He}$ - $^4\text{He}$  mixture at 100 mK (7%  $^3\text{He}$ ) in three different experimental cells: cell 1 with a volume of 3 cm<sup>3</sup>, cell 2 with a volume of 2 cm<sup>3</sup>, and cell 3 also with a volume of 3 cm<sup>3</sup> but not coated with polystyrene. All cells have approximately the same surface area of 40 cm<sup>2</sup>. 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 10<sup>4</sup> 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  $^3\text{He}$ - $^4\text{He}$  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  $\tau_c$ , resulting in a relaxation rate

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

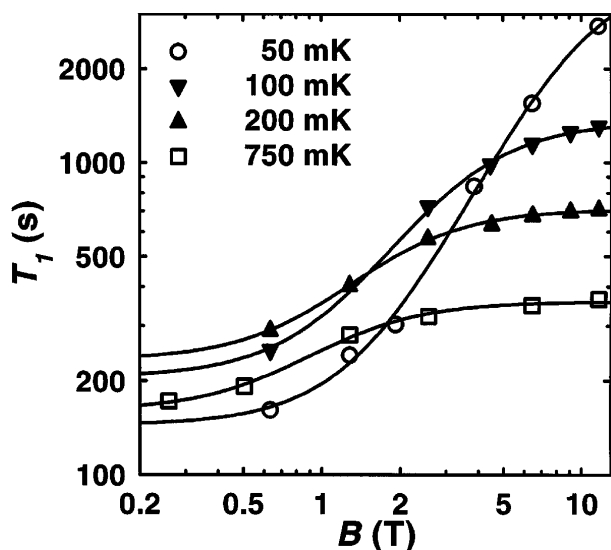


FIG. 1. Nuclear magnetic relaxation time  $T_1$  of liquid  $^3\text{He}$ - $^4\text{He}$  as a function of magnetic field. The curves are fits to the data with Eq. (3).

with  $\omega$  the Larmor frequency and  $\gamma$  the gyromagnetic ratio ( $\gamma = \omega/B \approx 32$  MHz/T for  $^3\text{He}$ ). 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  $\tau_D$  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  $^3\text{He}$  containing  $^4\text{He}$  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.

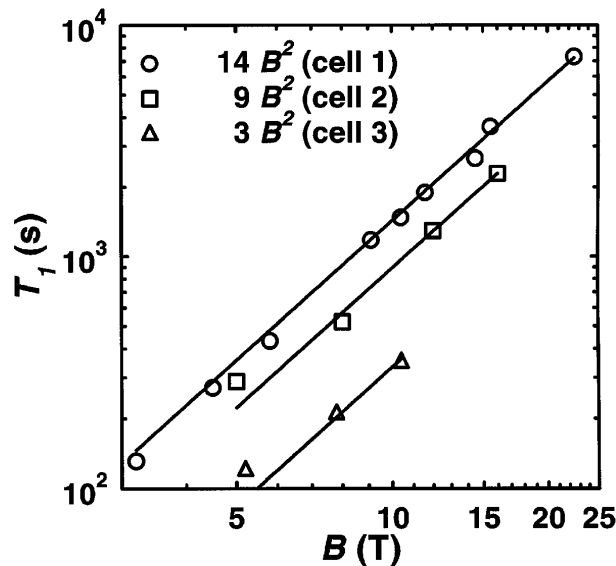


FIG. 2. Nuclear magnetic relaxation time  $T_1$  of a saturated  $^3\text{He}$ - $^4\text{He}$  mixture at 100 mK as a function of magnetic field and the quadratic fits for three different cells.

The correlation time  $\tau_c$  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  $\tau_D$  is estimated. Using the known diffusion coefficient for liquid  $^3\text{He}$  [11] and approximating the sample volume of the magnetometer by a sphere,  $\tau_D$  is found to increase from 20 s at 40 mK to 150 s at 200 mK. Using these numbers,  $\tau_c$  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  $\tau_c$  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  $T_{\text{in}}$ , negligibly short  $\tau_D$  and  $\omega \tau_c > 1$ , Eq. (3) reduces to the  $B^2$  term of Eq. (2). An estimate of  $\tau_D$  for the mixture is  $\sim 1$  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  $\tau_c$  is about 30 ns or longer. All other data of  $T_1$  of  $^3\text{He}$ - $^4\text{He}$  mixtures with both lower and higher  $^3\text{He}$  densities, not shown here, were always well described by Eq. (3).

Comparing  $T_s$  of the concentrated  $^3\text{He}$  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  $^3\text{He}$  density ratio of the concentrated  $^3\text{He}$  and the saturated  $^3\text{He}$ - $^4\text{He}$  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  $^3\text{He}$  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].  $T_{\text{ex}}$  is therefore not determined by  $\tau_D$ , contrary to the previous assumption, which is often used to extract  $T_{\text{in}}$  from  $T_1$  measurements as a function of temperature [1–3]. Since  $T_{\text{ex}}$  is not found to be equal to  $\tau_D$ , this naturally explains why  $T_1$  in  $^3\text{He}$ - $^4\text{He}$  mixture films does not scale with diffusion coefficient  $D$  [6].

The last parameter determined with Eq. (3) from  $T_1$  measurements of  $^3\text{He}$  containing 0.5%  $^4\text{He}$  as a function of the magnetic field is  $T_{\text{in}}$ . The results are plotted in Fig. 3 as a function of temperature together with the calculated  $T_{\text{in}}$  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  $T_{\text{in}}$  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  $T_{\text{in}}$  within experimental error of about 10%, which implies that the theory underestimates  $T_{\text{in}}$  with about 50% at the high temperature plateau.

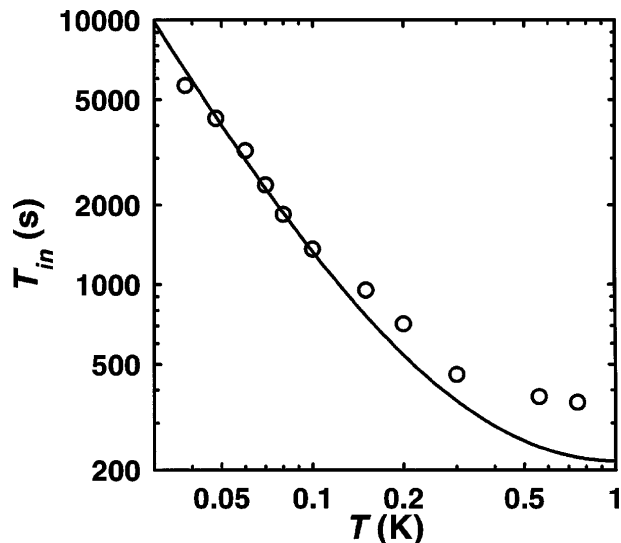


FIG. 3. Intrinsic relaxation time  $T_{\text{in}}$  of liquid  $^3\text{He}$  as a function of temperature determined with Eq. (3). The solid line represents the theory of Ref. [8].

We have shown that surface relaxation of  $^3\text{He}$  in the presence of  $^4\text{He}$  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  $^3\text{He}$  samples with solid  $^3\text{He}$  at the surface [5]. The difference between these two systems is that the  $^4\text{He}$  replaces the  $^3\text{He}$  at the surface, forming a superfluid  $^4\text{He}$  film on top of a solid  $^4\text{He}$  layer at the surface of the cell [14,15]. Because of the finite solubility of  $^3\text{He}$  in this film, the system near the surface should be the same for a saturated mixture as for liquid  $^3\text{He}$  containing a fraction of  $^4\text{He}$  to cover the surfaces, which explains why the observed surface relaxation is independent of the  $^3\text{He}$  density in the cell. This finite solubility also explains the observation in  $^3\text{He}$ - $^4\text{He}$  mixture films that after a certain coverage of the surface with  $^4\text{He}$ ,  $T_1$  becomes nearly independent of the  $^4\text{He}$  coverage [15].

A possible mechanism for the observed surface relaxation process is that a  $^3\text{He}$  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  $^4\text{He}$  film. Taking for  $D$  the bulk value for a 5%  $^3\text{He}$ - $^4\text{He}$  mixture [12], the observed  $\tau_c$  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  $^4\text{He}$  film [14]. This correlation length could be related to the surface roughness. The observed increase of  $\tau_c$  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  $^3\text{He}$  layers on top of the solid  $^3\text{He}$  layer [16]. Another mechanism might be that a  $^3\text{He}$  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  $^3\text{He}$  atoms at the surface, and  $\tau_c$  to the sticking time. This sticking behavior has also been observed in  $^3\text{He}$  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  $^3\text{He}$  containing  $^4\text{He}$ , unlike when there is a solid layer of  $^3\text{He}$  at the surface, is proportional to  $1 + (\gamma B \tau_c)^2$ . We relate the observed relaxation to the dynamics of the  $^3\text{He}$  atoms in the  $^4\text{He}$  film at the surface. The correlation time  $\tau_c$ , 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  $^3\text{He}$  we have experimentally determined with high precision is in good agreement with the theoretical predictions by Vollhardt and Wölfle [8].

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