Magnetization-Dependent Viscosity in Brute-Force-Polarized Liquid ^{3}He

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A new method to measure the magnetization dependence of the viscosity in polarized liquid ^{3}He is presented. The magnetization is obtained by "brute-force polarization" at 45 mK in magnetic fields up to 11 T; it is subsequently destroyed by saturation of the NMR signal. Our result, a relative increase of the viscosity of $(3\pm1.5)\times10^{-3}$ at 3.9% polarization and a pressure of 30 bars, disagrees with a prediction based on the "nearly metamagnetic" model.

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Since the proposals in 1979 to produce polarized liquid ^{3}He by either the rapid melting of polarized solid[1] or the condensation of polarized gas,[2] there has been considerable theoretical and experimental activity.[3,4] For example, several phenomenological theories predict a magnetic equation of state (MEOS): the paramagnon model,[5] the nearly solid model,[6] the nearly metamagnetic model,[7] and the functional density approach.[8] Experimentally, one way to investigate the polarized liquid is to study the thermodynamic properties. Several experiments to relate the MEOS of the liquid to that of the solid have been performed by rapid melting of polarized solid ^{3}He, which is supposed to show a paramagnetic behavior.[9-12] One experiment has been interpreted as indicating a metamagnetic transition in the liquid,[13] but has also been explained in terms of a phase transition in the solid.[14] Bulk-liquid experiments after a complete rapid melting of the solid—for example, sound velocity[15] and the heating due to magnetic relaxation of the polarized liquid[16,17]—may provide information concerning the liquid MEOS without reference to the solid. Thermodynamic measurements under equilibrium conditions are difficult, because presently available fields generate only tiny effects.

Another approach to probe the interactions in the polarized liquid is the study of the transport properties. The viscosity, η, is most easily accessible. Two experiments, using the method of rapid melting of solid ^{3}He and a vibrating-wire viscometer, have been published by Kopietz, Dutta, and Archie[18] and Frossati and coworkers.[4,19] The experiment in Ref. 18 showed a strong decrease of η with increasing magnetization at a temperature of 60 mK (20% decrease of η at 10% polarization), whereas Refs. 4 and 19 indicated a systematic increase with increasing polarization in the temperature range from 30 to 40 mK. A calculation by Hess and Quader[20] (HQ), based on a polarization-dependent effective interaction, which is similar to the interaction obtained within the framework of the nearly metamagnetic model,[7] is in reasonable accordance with the result of Ref. 18. This calculation predicts η to pass through a clear minimum at about 35% polarization before increasing to very large values.

In this Letter, we present a measurement of the polarization dependence of η through the destruction of magnetization by saturation of the NMR signal. This is a novel combination of NMR and viscosity techniques. The special properties of the Fermi liquid permit the measurements to proceed with high resolution. It enables us to use polarized liquid in thermal equilibrium and to circumvent the thermometry problems related to rapid-melting experiments and equilibrium methods. The saturation and subsequent relaxation of the magnetization are irreversible magnetic processes and cause temperature increases, which are easily monitored with the viscometer. The saturation perturbs the Fermi-Dirac distribution function by creating quasiparticles above the down-spin Fermi sea and holes in the up-spin Fermi sea. Since the quasiparticle lifetime is very short (about 10^{-12} s K^2/T^2), these quasiparticles and holes thermalize rapidly converting part of the absorbed energy into heat. The almost instantaneous temperature increase due to saturation and the slow temperature increase due to spin relaxation can be calculated and used to extract the polarization dependence of η without an independent measurement of the temperature.

We refer to Fig. 1, which shows the up- and down-spin Fermi sea at $T=0$, to clarify the irreversible heating due to saturation and relaxation (if we assume that the saturation time $\tau \ll \tau_1$, the relaxation time). The equilibrium polarization, m_0, is determined by the magnetic Fermi temperature T^{**} and the external field B_0: $m_0=\mu B_0/k_B T^{**}$ as long as $\mu B_0 \ll k_B T^{**}$. Here, μ is the ^{3}He nuclear magnetic moment. T^{**} has been measured as a function of pressure.[21] During a partial saturation from polarization m_0 to polarization m, quasiparticles close to the surface of the up-spin Fermi sea are kicked above the surface of the down-spin sea, gaining an extra energy, $2\mu B_0$. The number of quasiparticles which are excited is $\frac{1}{2}N(m_0-m)$ and the energy of the system is increased by an amount $E_{NMR}=N(m_0-m)\mu B_0$. N is the number of particles. These quasiparticles fall almost immediately, releasing an energy $E_{sat}=\frac{1}{2}N(m_0-m)(2\mu B_0-\delta\epsilon)$, where $\delta\epsilon=(m_0-m)\mu B_0/m_0$ ($\delta\epsilon$ is the width of the band of excited quasiparticles in Fig. 1). This energy is converted into heat, and the temperature rises over the

initial temperature T_0 by $\delta T_{\rm sat}=T^{**}(m_0^2-m^2)/2\gamma T_0$, if we assume a linear specific heat $Cv/R=\gamma T$ and consider only temperature changes that are small compared to T_0. After the saturation the quasiparticles relax by the dipolar interaction and they release an energy $E_{\rm rel}=\frac{1}{2}N(m_0-m)\delta\epsilon$, causing a temperature rise $\delta T_{\rm rel}=T^{**}(m_0-m)^2/2\gamma T_0$. We can account for the finite temperature deviations from the ideal Fermi-liquid behavior by multiplying the susceptibility with a factor $[1-\beta(T/T^{**})^2]$ (Ref. 21) and by replacing γT with $\gamma T+\Gamma T^3\ln(T/\theta)$.[22]

Saturation of the NMR signal changes the quality factor, Q, of the vibrating-wire viscometer for two reasons: the first contribution, δQ_T, arises from the temperature increases considered above. The second, δQ_m, is caused by the magnetization dependence of η. To second order in m we can write $\eta\propto(1+\alpha m^2)/T^2$. Since $Q\propto 1/\sqrt{\eta}$, we have $Q=\Omega T(1-\frac{1}{2}\alpha m^2)$, where Ω depends on properties of the wire as well as of the liquid. Saturating from m_0 to m, we have

$$\delta Q_{T,\rm sat}=\Omega T^{**}(m_0^2-m^2)/2\gamma T_0 \tag{1a}$$

and

$$\delta Q_{m,\rm sat}=\tfrac{1}{2}\Omega\alpha T_0(m_0^2-m^2). \tag{1b}$$

During the relaxation we have $m(t)=m_0[1-\exp(-t/\tau_1)]$ and find

$$\delta Q_{T,\rm rel}=\Omega T^{**}(m_0-m)^2[1-\exp(-2t/\tau_1)]/2\gamma T_0 \tag{1c}$$

and

$$\delta Q_{m,\rm rel}=-\tfrac{1}{2}\Omega\alpha T_0(m_0-m)\{m_0+m-2m_0\exp(-t/\tau_1)+(m_0-m)\exp(-2t/\tau_1)\}. \tag{1d}$$

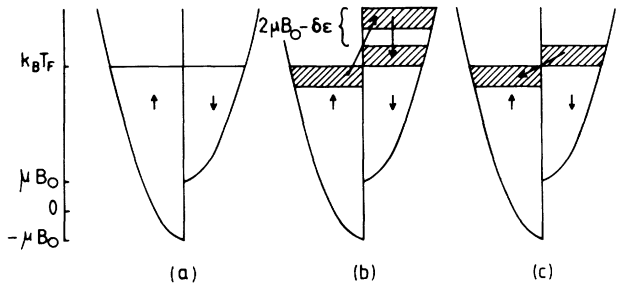

FIG. 1. The coupling between the spin bath and kinetic bath of liquid ^{3}He. The effect of the finite temperature, in fact $k_BT>\mu B$, is not shown to simplify the drawings. (a) The equilibrium situation in an external magnetic field B_0, (b) the saturation and immediate release of energy due to the quasiparticle lifetime [the simplification that all the quasiparticles in an energy band with a width $\delta\epsilon=(m_0-m)\mu B_0/m_0$ right below the surface of the up-spin Fermi sea are excited to an energy band of the same width above the surface of the down-spin Fermi sea has no consequences], and (c) the system immediately after the saturation just before it relaxes to the state depicted in (a).

In case of a full saturation, an asymmetry between $\delta Q_{\rm sat}$ $(=\delta Q_{T,\rm sat}+\delta Q_{m,\rm sat})$ and $\delta Q_{\rm rel}$ $(=\delta Q_{T,\rm rel}+\delta Q_{m,\rm rel})$ can be attributed to a polarization-dependent η. For a partial saturation we find

$$\frac{\delta Q_{\rm sat}}{\delta Q_{\rm rel}}=\frac{1+\gamma\alpha T_0^2/T^{**}}{(m_0-m)/(m_0+m)-\gamma\alpha T_0^2/T^{**}}. \tag{2}$$

Another manifestation of a polarization-dependent η is a nonexponential behavior of $\delta Q_{\rm rel}$ versus time as can be seen from (1d).

The experimental setup uses the viscometer as a thermometer. This proves to be very convenient because it is difficult to find an accurate fast-response thermometer which operates in a high magnetic field and is not susceptible to eddy-current heating. Measurement of the magnetocaloric effect requires a long thermal relaxation time τ_T compared to τ_1 and a minimal amount of parasitic heating due to the saturating rf power. The cell is a cylinder with a diameter and length of 7 mm. The concentration of ^{4}He impurities in the ^{3}He sample is nominally 50 ppm. The walls of the cell are expected to be coated with a saturated ^{4}He layer. The large volume of the cell and the poor thermal conductivity of the cell body combine to meet the requirement $\tau_T\gg\tau_1$ ($\tau_T=10$ h at $P=30$ bars and $T=45$ mK). The NMR coil is a cylindrical copper foil with a diameter and length of 16 mm and a slit of 7 mm and is thermally anchored to the still of a dilution refrigerator. It is adapted to 50 Ω at room temperature and can be used up to at least 400 MHz. During the saturation the frequency is modulated with an amplitude of 3 kHz at a modulating frequency of 400 Hz, which gives a fairly linear sampling over the linewidth (3 kHz). The viscometer is an elongated semicircle with a radius of 2 mm and a height of 3 mm made of a single-filament NbTi wire whose original CuNi cladding has been removed by etching with HNO_3. The remaining NbTi wire has a diameter of 34 ± 2 μm, a density of 6.4 ± 0.2 g/cm^3, and a resonance frequency in low field of about 1.5 kHz with an intrinsic Q of 2200. In 10 T and at 4.2 K the resonance frequency increases to about 3 kHz and the Q decreases to 800. The dissipation of the viscometer is about 50 pW during the experiment and the amplitude of the vibrations of the wire is estimated to be about 0.1 μm. The viscometer has been soldered to two 0.5-mm copper wires. One of the Cu wires serves as a thermal anchor and is attached to a thick silver rod, which provides for thermal contact to the mixing chamber and is also an electrical ground. This Cu wire partially short circuits the poor thermal

conductivity of the Araldite cell. The large thermal resistance from the wire to the cell compared with the thermal resistance to the mixing chamber reduces the parasitic heating due to eddy currents.

An example of the response of the viscometer to the saturation and subsequent relaxation of the magnetization is shown in Fig. 2. During the same time, we measure the magnetization to extract τ_1 and m. The relaxation is exponential with a τ_1 between 600 and 700 s, depending on the exact temperature. If we put the field off the NMR resonance and apply the "saturating rf power," there is an indication of a slight temperature increase of the order of 1%–2% of $\delta T_{\rm NMR}$ $(=\delta T_{\rm sat}+\delta T_{\rm rel})$. The parasitic heating always appears after the saturation pulse and has the same order of magnitude as the noise on the viscometer signal.

Our temperature scale is based on the Q of the viscometer, which depends on η, the resonant frequency, the density of the liquid and of the wire, and the radius of the wire. The estimated temperature accuracy arising from the wire parameters is approximately 8%. The behavior of η in the temperature range of interest is described by $\eta=1/T^2(b-cT)$, where b and c depend on the pressure.[23,24] The parameter b has been tabulated[25] and we interpolated the parameter c. In addition, a systematic shift of the temperature scale might be caused by the rather large excitation of the vibrating wire. To derive the viscous force on the vibrating wire, the $\mathbf{v}\cdot\nabla\mathbf{v}$ term in the Navier-Stokes equation has been neglected.[26] This implies $x_0 \ll \delta^2/r$, where x_0 is the amplitude of the vibrating wire, δ the viscous penetration depth, and r the radius of the wire. In our case $x_0=0.1$ μm and $\delta^2/r=1$ μm. Consequently, extra damping might arise and produce a depression of the temperature value measured. If we increase the temperature scale by 15% and use the low-field values of the specific heat and the susceptibility, the measurements are thermodynamically consistent in the sense that $\delta T_{\rm NMR}$ fits well with the calculated value. By use of this temperature scale and (2), the magnetization dependence of η is given by $\alpha=2\pm1$. Note that an error in the temperature affects only the magnitude but not the sign of η. We have tried to fit (1) with two adjustable parameters, α and a constant temperature drift, to the experimental curve shown in Fig. 2. The fixed parameters are the susceptibility,[21] the specific heat,[22] $T_0=50$ mK, $m/m_0=0.05$, and $\tau_1=700$ s, the latter two obtained from a measurement of the relaxation of the magnetization. An independent very good fit, which accounts for the ratio $\delta Q_{\rm sat}/\delta Q_{\rm rel}$ as well as the nonexponential behavior during the relaxation, is obtained with $\alpha=3$. This confirms the result obtained with (2).

Several systematic errors might affect either $\delta Q_{\rm sat}$ or $\delta Q_{\rm rel}$. (i) An increase in $\delta T_{\rm sat}$ because of relaxation during the saturation is estimated to be smaller than 5% of

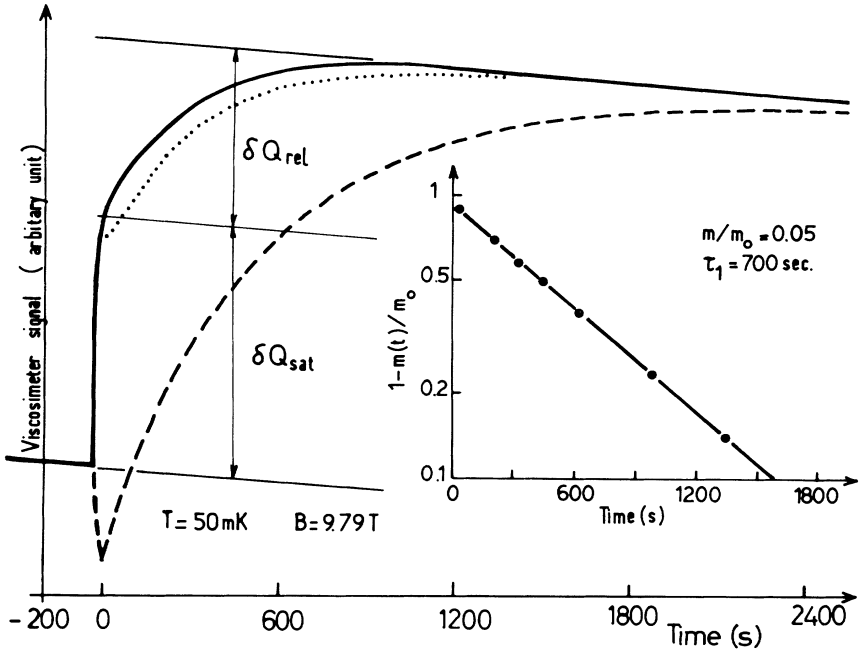

FIG. 2. Typical experimental recorder trace of the viscometer during and after a saturation (solid line). The ratio $\delta Q_{\rm sat}/\delta Q_{\rm rel}=1.36$. By our taking into account $m/m_0=0.05$ and using (4) this is interpreted as an increase of η with increasing magnetization ($\alpha=2$). For comparison, we have shown calculated curves with $\alpha=-30$ as predicted by HQ (dashed line) and $\alpha=0$ (dotted line). The experimental curve is very well fitted by (1) with $\alpha=3$. We remark that even the relaxation only cannot be well fitted with $\alpha=0$. Note that if $\alpha=-30$, δQ is negative during saturation and shortly after magnetization because of the large negative contribution of δQ_m. Inset: A logarithmic plot of the relaxation of the magnetization after saturation.

δT_{sat} at $P=30$ bars and $T=45$ mK. The consequent decrease in α is much smaller than its statistical error. (ii) The thermal relaxation time is not infinite compared to τ_1. A computer simulation shows that the systematic errors (a decrease in δT_{rel}) due to this effect at $P=30$ bars and $T=45$ mK are negligible. (iii) Inhomogeneous saturation of the NMR signal causes magnetization and temperature gradients. A temperature gradient is most likely to introduce a systematic error during the saturation, whereas a magnetization gradient causes irreversible heating due to the diffusion of magnetization in an external field. The time constant of this process is about 200 s (the spin diffusion time). Therefore, an inhomogeneous magnetization is most likely to introduce errors during the relaxation. However, from geometric considerations we conclude that the saturating field is rather homogeneous and the systematic errors are negligible.

To conclude, the relative increase in η of $(3\pm1.5)\times10^{-3}$ at 3.9% magnetization at $P=30$ bars, i.e., $\alpha=2\pm1$, is in clear disagreement with the experimental result obtained in Ref. 23 and the calculation by HQ, which is based on the nearly metamagnetic model[7] and predicts $\alpha=-30$. Our result lies within the error bars of the result obtained in Ref. 4. Note that comparison between η in zero field and in 10 T would require a temperature reproducibility of the order of 10^{-3} to resolve such an effect. The decrease measured by Kopietz, Dutta, and Archie, could be an artifact due to the strong temperature dependence of η ($\propto 1/T^2$) and the difficult conditions with respect to thermal equilibrium in this experiment. We remark that spin-flip scattering does not enter in the MEOS, but it does affect the transport properties. The assumption made by HQ to generalize the expression for the spin-flip scattering at zero to finite polarization may be questionable.[27] In the paramagnon model, we expect η to increase with increasing magnetization.

As a second result we would like to emphasize that degenerate liquid ^{3}He is a very peculiar system in the sense that the saturation of the NMR signal and subsequent relaxation of the magnetization in liquid ^{3}He can be observed by means of a measurement of the kinetic (lattice) temperature of the quasiparticles.

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