Measurement of the Magnetization of Liquid ^{3}He up to 200 T

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We have measured the relaxation of energy and polarization m in transiently polarized liquid ^{3}He. From these measurements we deduce the value of the magnetic field necessary to stabilize m. This field is shown to be higher than the extrapolation of the low-field regime, and amounts to 200 T at $m=0.6$, $T=70$ mK, $P=26$ bars.

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Liquid ^{3}He has long been recognized as a model system of a strongly correlated Fermi liquid. Beyond the famous phenomenological Landau theory, several microscopic models aim to account for the physical properties of this system. Among them, the "nearly localized" model[1] and paramagnon theory,[2] respectively, ascribe the strong enhancements (compared to a free Fermi gas) of the specific heat and the magnetic susceptibility to the proximity of either a Mott transition or a ferromagnetic instability. Measuring the magnetic susceptibility χ of liquid ^{3}He up to very high polarizations has been suggested as a crucial step in determining which approach is most suitable,[3] but only small polarizations m ($m < 12\%$) can be reached in a static magnetic field (<30 T). Transient experiments are possible anyway, since the relaxation of the liquid polarization is a slow process ($T_1 \sim 10$–1000 s).[3,4] Large polarizations can be obtained in the liquid by melting solid ^{3}He, previously polarized at low temperature ($T\sim5$ mK) in a large magnetic field (~10 T). T_1 is then much longer than any other microscopic relaxation time of the system; this allows one to perform thermodynamic measurements as a function of the slowly varying magnetization. A number of experiments have been performed, using this method proposed by Castaing and Nozières[3] (CN), resulting in contradictory conclusions about the magnetic behavior of the liquid. We stress that none of them allows direct investigation of χ. Melting-pressure measurements[5,6] suggest a peak in χ around $m\sim20\%$ (Ref. 7) (as expected from the "nearly localized model"), but in this case one deals with a mixture of liquid and solid, where a specific hypothesis must be made to relate the average polarization to that of the liquid.[8] Viscosity measurements[9(a),10] would favor a decrease in χ with m, consistent with the paramagnon model.[11] But there one measures a transport coefficient, which is related to χ only through a specific hypothesis. The sound-velocity measurement[12] did not give conclusive information about χ.[13]

We report a direct investigation of the relationship between m and the magnetic field B_m, with which m would be at equilibrium. This investigation is performed at sufficiently low temperature for the liquid to be degenerate ($T<0.1$ K). It is carried out by measuring the relaxation of the polarization and the relaxational heating. Indeed, for each flipped spin an energy given by the difference between the chemical potentials of up and down spins is released as heat. This amounts[3] to $2\mu_n(B_m-B_a)$, where B_a is the applied field and μ_n the magnetic moment of ^{3}He. Then, for n atoms heat is produced irreversibly at a rate given by

$$\dot{Q}_m = -n\mu_n(B_m-B_a)\dot{m}\,. \tag{1}$$

In an adiabatic situation, a part $\dot{Q}_T$ of $\dot{Q}_m$ will warm up the cell of heat capacity C_m at fixed m ($\dot{Q}_T=C_m\dot{T}$), with the remaining being absorbed by the m dependence of the entropy of the polarized liquid. Measuring $\dot{Q}_T$ and $\dot{m}$ we obtain a field B_T defined by $B_T-B_a=-\dot{Q}_T/n\mu_n\dot{m}$ which, using the identity $(\partial B_m/\partial T)_m=-(\partial S/\partial m)_T$, is related to B_m by

$$B_T=B_m-T\left(\frac{\partial B_m}{\partial T}\right)_m\,. \tag{2}$$

The correction $T(\partial B_m/\partial T)$ is of order T^2; thus it is small at low T. With this method, we have been able to measure B_T as a function of m for $m<0.6$, and temperatures in the range $0.07<T<0.09$ K. We find, rather unexpectedly, that, even close to the melting pressure ($P\sim26$ bars), the susceptibility $\partial m/\partial B_T$ decreases monotonously when m and B_T increase.

Polarized liquid obtained using the CN method is warm ($T>0.2$ K), and for measuring B_m in the fully degenerate region, we must cool this liquid below 0.1 K in a time much shorter than T_1. This is achieved by using a heat tank of 0.085 mol of saturated-vapor-pressure unpolarized liquid, in extremely good thermal contact with the polarized sample of 0.01 mol at 25 bars. We use the liquid of the heat tank as a part of the heat switch to the mixing chamber of our dilution refrigerator.[14] The sample and the tank are embedded in 700-Å sintered silver powder in order to provide a large contact area between them. These heat exchangers were sintered in holes in the silver body of our cell (Fig. 1). The longest relaxation time for temperature gradients across our cell is shorter than 1 s at 70 mK and 11 T. In a sinter the relaxation of m occurs mainly at the walls.[15] Since the

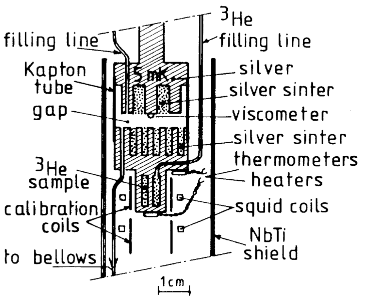

FIG. 1. The experimental cell.

diffusion of magnetization towards the relaxing centers is driven by a gradient of the effective field, the relaxation occurs under a smaller effective field than in the bulk. Thus less energy is dissipated in the relaxation process itself, but this is counterbalanced by the heat produced by (irreversible) spin diffusion: It can be shown[16] that in (1), $B_m\dot{m}$ and $\dot{m}$ then represent their spatial averages. For the actual size of the pores (0.1 μm), the spin diffusion in the liquid is very quick (10 μs) on the time scale of T_1 (70 s), so that m is homogeneous in the liquid. Moreover, only 0.5% of the atoms are adsorbed on the walls; thus the bulk values of m and B_m are close to their averages, and the use of Eq. (1) for evaluating B_m is justified.

For measuring $\dot{Q}_T$, we used a Speer resistor, directly glued on the cell. Its resistance R is measured using a sensitive four-wire bridge resulting in a noise ~ 1 μK/Hz$^{1/2}$. Absolute conversion of $\dot{R}$ to $\dot{Q}$ is obtained from a separate experiment where R is measured as a function of the heat Q applied to the isolated cell using a resistive heater. Because the liquid is unpolarized in the latter case, this neglects the difference between C_m and $C_{m=0}$. This is not a large error since the polarized liquid contributes about 10% to C_m.

Our samples are solidified using the blocked-capillary technique. The molar volume is such that, at low temperature, all of the sample is solid and just above the melting curve. The solid samples were cooled during 12–48 h, down to 6 mK. We thus achieved polarizations above 80%, in the solid, for $B_a = 11$ T. The polarization of the sample is measured by an rf SQUID. Extreme care was taken in the design of this magnetometer[16] since it is operated at 11 T, while measuring small magnetic fluxes. With our setup the uncertainty on $m(t)$ comes from the drift of the applied field during the elapsed time t, and amounts to about $\delta m \sim 0.002$ for $t = 1$ h. The absolute sensitivity of the setup was calibrated using the Curie law of the polarization in the solid phase.

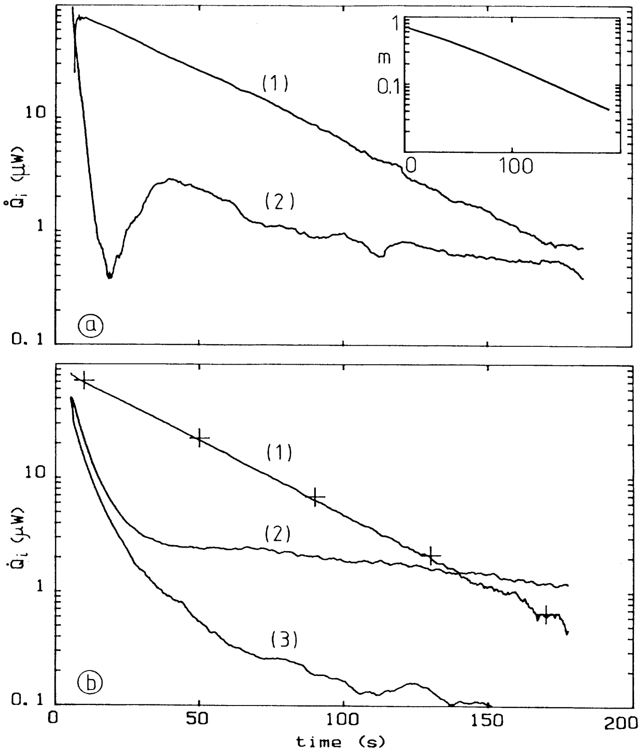

FIG. 2. (a) Curve 1: Relaxation of the liquid excess polarization (inset) and the power $\dot{Q}_T$, following a rapid melting of spin-polarized solid ^{3}He at 11 T (starting at $t = 0$). Also shown is the warming rate following a rapid melting at zero field (curve 2); this is due to heat coming from the hot capillary ($\dot{Q}_{\rm cap}$). (b) Curve 1: measured power, when $\dot{Q}_a = [(3\text{ mJ})/(34\text{ s})]\exp[-t/(34\text{ s})]$ is applied to the cell after a heat pulse of 10 mJ at 11 T. + indicates the applied power. The cooling rate following the application of a 9-mJ heat pulse to the cell containing cold liquid is shown in curve 2 (curve 3) for an applied field of 11 T (0 T). It is mainly due to the silver spin relaxation ($\dot{Q}_{\rm Ag}$) (to the heat $\dot{q}$ flowing to the cold capillary). In (a), curve 1, and (b), curve 1, the contribution of silver spins to the magnetization and the warming rate have been subtracted, as well as $\dot{Q}_{\rm cap}$ for (a), curve 1. $\dot{Q} = 1$ μW corresponds to $\dot{T} = 5$ μK/s.

Figure 2(a), curve 1, shows the decay of m and the heating $\dot{Q}_T$, following a melting. The most relevant parameters of these experiments are the following: initial temperature, 6 mK; just after melting, $P = 26$ bars and $T = 70$ mK; and after the relaxation, $T = 90$ mK. The total magnetic energy released amounts to about 3 mJ. The polarization relaxes with a time constant close to 70 s, and the warming rate $\dot{Q}_T$ about 2 times faster, as expected from Eq. (1). Note that while the temperature gradients due to the melting are large (about 9 mJ are generated in 3 s), they relax rapidly in our cell, the corresponding $\dot{T}$ being negligible after 15 s [see Fig. 2(a), curve 2, and Fig. 2(b), curves 2 and 3]. There are two extraneous sources of heat which must be subtracted from the warming rate in order to get $\dot{Q}_T$. First, the liquid ^{3}He which flows out of the cell to the narrow filling line is hot (0.3 K) and injects heat into the cell at a rate $\dot{Q}_{\text{cap}}$. We measure $\dot{Q}_{\text{cap}}$ in a separate experiment where the solid is melted at zero field, where $\dot{Q}_T=0$ [see Fig. 2(a), curve 2]. Second, the nuclear moments of the silver, which are initially substantially polarized, relax in about 120 s and absorb $\dot{Q}_{\text{Ag}}$. This was studied by applying heat pulses of 9 mJ in 3 s to the cell, initially at 6 mK, 26 bars, and $B_a=11$ T [Fig. 2(b), curve 2]. In this case we measure $\dot{Q}_{\text{Ag}}+\dot{q}$, where $\dot{q}$ is the heat absorbed by the cold capillary. $\dot{q}$ was measured with pulses at zero field [Fig. 2(b), curve 3]. The silver spins also generate a flux on the pickup coil that we measure during the heat pulses at 11 T, and subtract from the signal in the decompressions (it corresponds to 6% of the ^{3}He signal). As a test of our ability to measure $\dot{Q}_T$ we applied a pulse of 10 mJ to the cell at 6 mK, 26 bars, and 11 T, followed by a heating $\dot{Q}_a=[(3\text{ mJ})/(34\text{ s})]\exp[-t/(34\text{ s})]$ which mimics $\dot{Q}_T$. By subtracting $\dot{Q}_{\text{Ag}}$, we are then able to calculate an incoming power which is equal to $\dot{Q}_a$ within 3%, as shown in Fig. 2(b), curve 1.

From Fig. 2(a) and Eq. (1), we obtain the relation m vs B_T shown in Fig. 3. The error bars represent changes of 20% in $\dot{Q}_{\text{cap}}$ and $\dot{Q}_{\text{Ag}}$, which is an upper bound of their irreproducibility. The straight line represents the expected curve at low m, including the effect of the "entropic" correction $T\,\partial B_m/\partial T$, as calculated from Ref. 17. Within the error bars, the curve is a straight line at low m, with the right slope and extrapolation to the origin. The corrections $\dot{Q}_{\text{Ag}}$ and $\dot{Q}_{\text{cap}}$ are important in order to get this behavior, but do not influence the results for $m>0.2$ (i.e., smaller times). Figure 3 shows that the susceptibility decreases monotonously when m and B_T increase. For the highest m ($=0.6$), B_T is then 30% higher than the extrapolation of the initial behavior. This is well outside our errors bars[18] and is certainly our most salient result.

This new result quantitatively agrees with the simple RPA picture of the strong Stoner enhancement of the susceptibility, due to short-range repulsion between antiparallel spins in direct space. When m increases, the system goes rapidly away from the ferromagnetic instability and χ levels off (Fig. 3 and its inset). Such a quantitative agreement was also observed for the slope[9,10] of the viscosity versus m^2, compared to the prediction of the paramagnon model.[11]

It is in strong contrast to the metamagnetic behavior predicted by Vollhardt in the framework of the almost localized theory,[1] since this theory predicts a first-order transition to a fully polarized state for $m\sim0.20$.[19] It is necessary here to mention what would happen in our experiment if such a transition would occur. The ^{3}He in each pore of the sinter should split into two phases, with different polarizations. The relaxation would happen at the same effective field $B_m=B_{\text{crit}}$ in the two phases, where they have the same chemical potential.[3] In such a case B_m and B_T would also show a Maxwell "plateau,"[16] which we do not observe. The polarization we achieve is far above the spinodal point ($m\sim0.40$),[19] so that metastability cannot be invoked to reconcile our result with this calculation. In the phenomenological approach of Bedell and Sanchez-Castro,[20] the transition is limited to an increase of χ at some polarization m_c. In such an occurrence we should worry about the entropic correction $T\,\partial B_m/\partial T$. In the latter calculation, no anomaly of S is expected at the pseudotransition, but we expect (consistent with Ref. 1) that an anomaly in S would be a maximum at m_c.[21] This implies that $-T\,\partial B_m/\partial T$ is positive for $m<m_c$ and negative for $m>m_c$, so that its effect would be to create in $B_T(m)$ a stronger plateau than in $B_m(m)$.

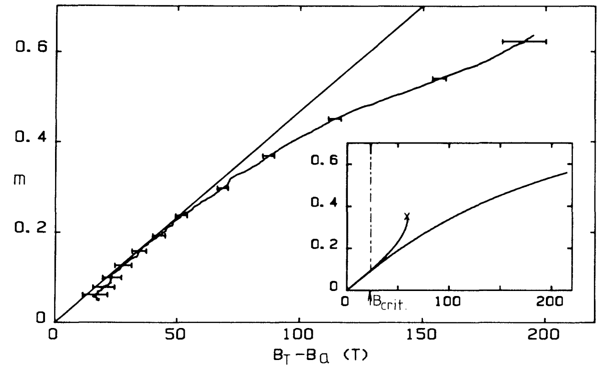

FIG. 3. $m(t)-m(\infty)$ vs $(B_T-B_a)(t)$ for the melting experiment of Fig. 2(a), curve 1. The error bars are displayed each 15 s. The straight line is the expected behavior at low m. Inset: m vs B_m as predicted by the RPA (lower trace) and for the Gützwiller ansatz (Ref. 1) (upper trace), for a twentyfold-enhanced initial susceptibility (corresponding to 25 bars). Also shown is the spinode (×) and the field B_{crit} at which the transition occurs in the latter model.

By analyzing the melting-pressure experiments,[5,6] the authors of Ref. 7 have proposed a peak in χ, the peak found from Ref. 5 being narrow and not very high. With our accuracy on differences between fields being better than the error bars in Fig. 3, since they represent a bias error in B_T, this peak is hardly compatible with our result. The authors also proposed a crude way of determining the (unknown) polarization in the decompressions of Ref. 6, and suggested a strong first-order transition at 0.07 K. Such a transition is certainly not compatible with our results.

We conclude that our calorimetric method allows one to measure the effective field in polarized ^{3}He at 70 mK $<T<$ 90 mK and for polarization up to 60%. None of the previous experiments could probe ^{3}He at such a large polarization. The present investigation is made possible by the extremely short thermalization time in our setup, and by the principle of direct measurement of B_m. We report fields of 200 T, the highest value ever achieved in ^{3}He. Our results show that the field is higher than the extrapolation of the paramagnetic regime, by an amount compatible with the Stoner model. This rules out a significant metamagneticlike behavior in liquid ^{3}He, even at low temperature.

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