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Polarization Decreases the Specific Heat of Liquid ^{3}He

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We report on the first measurements of the polarization dependence of the specific heat of liquid ^{3}He. Transient polarizations m of up to 70% were reached by using the rapid melting technique. The specific heat at 60–100 mK and 27 bars is found to decrease approximately as m^2, the reduction reaching at least 30% for $m = 70\%$. These results contradict the nearly localized picture of ^{3}He, and are in agreement with the idea that a large part of the specific heat is due to spin fluctuations.

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Liquid ^{3}He is a system of strongly correlated fermions. Yet, at low temperature, its thermodynamic and transport properties are similar to those of a dilute fermion gas. This is accounted for by the Landau theory, which describes the low energy states of the liquid as those of a system of weakly interacting, spin 1/2, quasiparticles of effective mass m^*. The interactions modify the thermodynamic quantities with respect to the noninteracting case, the renormalization being larger as the pressure P is increased up to solidification. From 0 to 34 bars, the enhancement of the specific heat, equal to the ratio of m^* to the bare ^{3}He mass, increases from 3 to 5, and that of the magnetic susceptibility from 10 to 20.

A central issue in correlated Fermi systems is to understand the origin of the specific heat enhancement. In liquid ^{3}He, two models ascribe it to the proximity to a phase transition. In both models, the interaction U between atoms is considered as local, and, due to the Pauli principle, takes place only between unlike spins. In a mean field approximation, this favors spin alignment, and, beyond a critical interaction, leads to ferromagnetism. This approach is at the heart of the "nearly ferromagnetic" model [1], in which the large enhancement of the susceptibility is ascribed to the proximity of liquid ^{3}He to this transition, and that of the specific heat to the *resulting* large spin fluctuations. Anderson and Brinkman [2] rather argued that liquid ^{3}He is close to a localization transition induced by the interactions (Mott transition). In this "nearly localized" picture, based on a lattice model of ^{3}He, the on site interactions lead to local correlations, which hinder the motion of atoms and increase the effective mass [3]. The large enhancement of the specific heat and of the magnetic susceptibility *both* stem from ^{3}He being close to the critical interaction U_c above which the system would be fully localized. As shown by Vollhardt [4], this quantitatively accounts for the small pressure dependence of the Landau coefficient F_a^0.

While neutron scattering experiments find evidence of spin fluctuations in liquid ^{3}He [5], their contribution to the specific heat is still a matter of controversy [6]. In this Letter, we directly address this issue by measuring the polarization dependence of the specific heat: if the specific heat enhancement is mainly due to spin fluctuations, it is expected to decrease with polarization, since such fluctuations are "quenched" in a magnetized system. By contrast, in the nearly localized picture, Vollhardt has predicted that polarization should increase the effective mass, and therefore the specific heat [4]. While Vollhardt's calculation relies on specific assumptions, his prediction, which reflects the fact that polarization drives the system closer to localization, is probably a general feature of the nearly localized picture.

Large polarizations of liquid ^{3}He at 27 bars are obtained by rapid melting [7] of a highly polarized ^{3}He solid ($m \simeq 80\%$ at $T \simeq 8$ mK in an 11 T field). Measurements of the polarization dependence of the specific heat are performed during the slow decay of polarization from 70%, corresponding to an effective magnetic field in excess of 200 T [8], to equilibrium ($m \simeq 4\%$). In order to overcome the low thermal diffusivity of ^{3}He ($D_T \simeq 5 \times 10^{-2}$ mm^2/s at 100 mK and 27 bars), which would imply long thermal relaxation times with respect to the polarization relaxation time T_1, we confine ^{3}He inside a silver sinter. This heat exchanger increases the thermal diffusivity by an order of magnitude, and allows us to couple efficiently the experimental cell to a large heat reservoir (subsequently called heat tank) which limits the temperature rise due to melting [9,10]. The drawback is that we cannot directly measure the specific heat by an adiabatic method [11]. Instead, we measure the thermal time constant of the cell. First, we will demonstrate that polarizing the ^{3}He decreases this time constant. Second, we will show that, in our conditions, this decrease mainly reflects a polarization induced decrease of the ^{3}He specific heat.

We use the same experimental setup as in Ref. [9]. Its main features are shown in Fig. 1. Both the heat tank and the cell are machined from 4N silver and annealed to obtain a very large thermal conductivity, so as to avoid vertical temperature gradients. The experimental volume is a 4 mm diameter, 15 mm long, cylinder, fully sintered except along a 0.2 mm wide slit, which contains a vibrating wire viscometer. The average polarization inside the cell is measured with a SQUID magnetometer. A thermometer

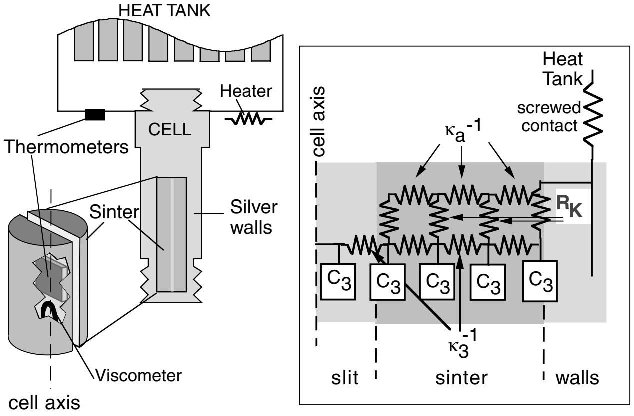

FIG. 1. The experimental cell. The inset sketches the thermal path involved in the thermal relaxation. κ_3 and κ_a are the ^{3}He and silver thermal conductivities, R_K is the sinter Kapitza resistance, and C_3 is the ^{3}He specific heat.

and a heater glued on the heat tank allow one to regulate the temperature of the silver walls. The thermal time constant of the cell is then obtained from the time dependence of the viscometer temperature. We use the viscometer because, unlike the carbon thermometer also located inside the slit [9], it has no thermal inertia of its own. The viscometer temperature is obtained from the measured viscosity $\eta(m, T)$, by computing the viscosity $\eta_0(T)$ that would be measured in the unpolarized liquid according to $\eta_0(T) = \eta(m, T)/(1 + 3m^2)$ (appropriate below 100 mK [9]), and converting it to temperature from a separate calibration of $\eta_0(T)$.

The procedure we use to measure the cell thermal response time is illustrated in Fig. 2: Right after melting, both the heat tank temperature T_{HT} and the viscometer temperature T_V increase. After 10 s, T_{HT} stabilizes around 45 mK, while T_V rapidly decreases towards this value. At this point, we regulate the heat tank temperature at 60 mK and measure the relaxation of T_V during 7.5 s, at a polarization of 70%. The regulation is then turned off and the system cools down until we apply a new temperature step. Between the steps, the viscometer is warmer than the heat tank, due to the heat released by the magnetic relaxation [8].

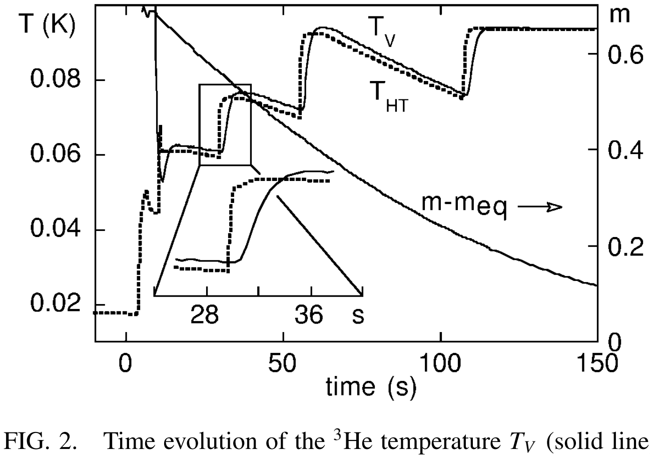

FIG. 2. Time evolution of the ^{3}He temperature T_V (solid line) inside the cell, in response to steps of the heat tank temperature T_{HT} (dotted line), applied during the relaxation of the ^{3}He polarization m towards $m_{\mathrm{eq}} \approx 4\%$, after melting the solid at zero time. The inset blows up the second step.

Figure 3 shows the relaxation of the heat tank and viscometer temperatures for each step, the time origin being taken at the beginning of the step. Here, the viscometer temperature is corrected for the effect of the magnetic relaxation (which gives a constant shift over the short time scale of the steps). For both temperatures, we plot their normalized deviation from their final value T_f $[T_f - T(t)]/[T_f - T(0)]$. For the first temperature step, which is applied during the initial fast decrease of the viscometer temperature, the corresponding background contribution was subtracted from the raw signal (by fitting to an exponential prior to the step). In order to separate the effect of polarization from that of temperature, we compare each temperature step of this series to an identical one in the unpolarized liquid: starting with cold unpolarized liquid, we apply a heat pulse to reproduce the initial heating in the melting experiment, and repeat the procedure of Fig. 2. The comparison of the two experiments in Fig. 3 shows *unambiguously* that the polarization decreases the time scale of relaxation [12].

The relaxations are nonexponential due to the complex thermal scheme depicted in Fig. 1. A measure of their time scale is the time integral of the (normalized) temperature difference $T_V(t) - T_{\mathrm{HT}}(t)$, pictured as the hatched area in Fig. 3(a). This integral is related to the response function of the viscometer temperature $S(t)$, defined such that, for a given small temperature change $T_{\mathrm{HT}}(t)$, $T_V(t)$ obeys the

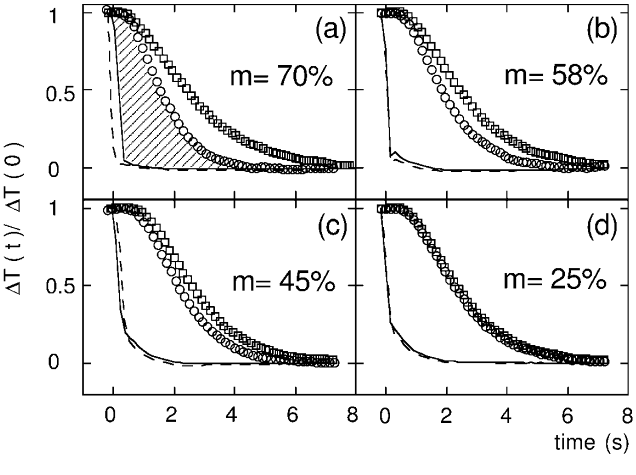

FIG. 3. In polarized ^{3}He, the normalized relaxation of the inner temperature T_V (circles) for the steps of T_{HT} (continuous lines) shown in Fig. 2 is faster than that observed for similar steps in unpolarized ^{3}He (squares and dashed lines). The hatched area defines the delay time τ used in the analysis.

general relation [13]

$$T_V(t) - T_{\mathrm{HT}}(t) = -\int_{t'=-\infty}^{t} S(t - t')\dot{T}_{\mathrm{HT}}(t')\,dt'. \quad (1)$$

For a step of $T_{\mathrm{HT}}(t)$, Eq. (1) implies that the hatched area above is given by $\tau = \int_0^\infty S(t)\,dt$, even in our case of a nonideally sharp step. The same equation also implies that, in a hypothetical experiment where T_{HT} would be ramped linearly with time ($\dot{T}_{\mathrm{HT}} = \text{const}$), corresponding to a stationary distributed heating across the cell ($\propto -C_3\dot{T}_{\mathrm{HT}}$), $T_V - T_{\mathrm{HT}} = -\tau\dot{T}_{\mathrm{HT}}$. This shows that the measured integral τ is the product of the ^{3}He specific heat C_3 (the silver specific heat being negligible) times an appropriate static thermal resistance between the viscometer and the heat tank. This resistance involves the thermal conductivities κ_a and κ_3 of the silver sinter and ^{3}He, and the Kapitza resistance R_K between them. The problem is then to know if changes of R_K or κ_3, rather than of C_3, could explain the observed polarization induced decrease of τ.

In order to investigate this point, we use the (continuous) thermal model [10] of the cell sketched in Fig. 1. The input parameters R_K and κ_a/T of the model were measured in separate experiments; κ_3 was taken from Ref. [14]. The model correctly accounts for the temperature dependence of τ in nonpolarized ^{3}He between 20 and 400 mK, as shown by Fig. 4. In the temperature range of interest (60–150 mK), it implies that the thermal resistance is dominated by the sinter effective thermal conductivity, in which the ^{3}He contribution amounts only to 10%. Hence, changes in τ mainly come from those of C_3. More specifically, a factor of 2 increase in κ_3 or $1/R_K$ would decrease τ at most by 9% and 6%, respectively. Such a large change is a maximum for κ_3: by using the viscometer both as a heater and a thermometer, we recently estimated a 20% increase for a 40% polarization [15]. As for the change in R_K, it should be much smaller as the sound velocity in ^{3}He at 27 bars increases by less than 10^{-3} for a 30% polarization [16]. Thus, the observed 40% reduction of τ at $m = 70\%$ implies a minimal reduction of the specific heat by 30%. From now on, we will therefore assume that τ depends on polarization only through the specific heat C_3.

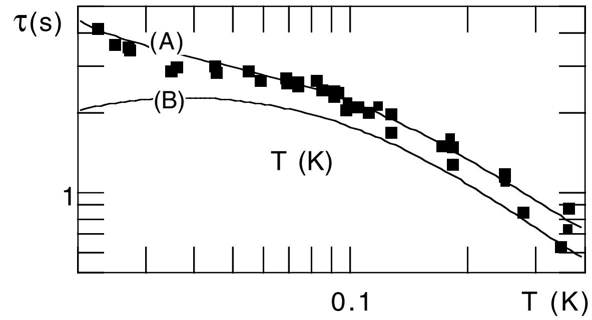

FIG. 4. Comparison of the delay time measured in unpolarized ^{3}He at 27 bars to the prediction of the thermal model sketched in Fig. 1 [line (*A*)]. The dominant contribution is due to the sinter effective thermal conductivity [line (*B*), corresponding to the parallel conductions of ^{3}He and silver, with $\kappa_3 < \kappa_a/10$ at 80 mK]. As a consequence, the polarization dependence of τ mainly comes from that of C_3.

Let us now discuss the polarization dependence of C_3, inferred from experiments similar to that of Fig. 2, and shown in Fig. 5. The observed decrease contradicts the nearly localized model. Assuming C_3 [17] to be proportional to m^*, we computed from Ref. [4] its variation for arbitrary m, using $U/U_c = 0.9$ as determined by Vollhardt for 27 bars [curve (*A*) of Fig. 5]. In this model, $m_0 \approx 40\%$ and $m_1 \approx 15\%$ are, respectively, the polarizations at which the system with homogeneous polarization m becomes absolutely and relatively unstable with respect to the fully polarized state. Accordingly, beyond m_1, the system should separate into two phases ($m = 1$ and $m = m_1$) [18]. We do not see any evidence for such a transition. This agrees with our previous result that the susceptibility decreases with polarization [8] between $m \approx 20\%$ and $m \approx 60\%$. However, in the latter case, it could have been argued that the discrepancy could be corrected by, e.g., improving [19] on the Gutzwiller approximation used by Vollhardt, without renouncing the basic idea that the specific mass enhancement results only from the proximity to localization.

Our present result rules out this hypothesis, unless, quite unexpectedly, polarization drives the system away from localization. This conflict cannot be due to the fact that Vollhardt's calculation does not take into account the decrease of specific heat due to the polarization induced changes of the Fermi levels of up and down spins (which is the only effect for free fermions). For a parabolic dispersion relation, this effect alone corresponds to

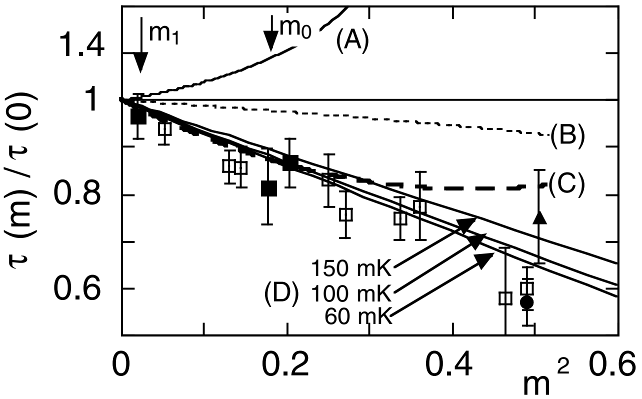

FIG. 5. Polarization dependence of the measured thermal relaxation time scale τ for different final temperatures: ● 60 mK, □ 75–85 mK, ■ 90–105 mK, ▲ 160 mK. The lines correspond to predictions for the polarization dependence of the specific heat: (*A*) nearly localized model (note the compressed scale); (*B*) free fermions; (*C*) paramagnons; (*D*) experimental dependence of the susceptibility plus Maxwell relation for three temperatures (see text).

$C_3(m)/C_3(0) \simeq 1 - m^2/9 - \cdots$. As shown by curve ($B$) of Fig. 5, this is too weak to compensate for the strong enhancement originally predicted by the nearly localized model.

A finite temperature extension [20] of the nearly localized model indeed predicts a decrease of the specific heat with increasing polarization. However, this prediction seems disputable as the very mechanism responsible for the increase of specific heat in the original model (the polarization dependence of the double occupancy of sites) is neglected in this approach [18].

We rather believe that the observed 30%–40% reduction of the specific heat shows that this strong fraction of the specific heat cannot be described in terms of a near localization of the ^{3}He atoms. Such a reduction, much larger than for free fermions [curve (B) of Fig. 5], is more in line with the idea that spin fluctuations contribute to a significant part to the entropy.

This conclusion is in qualitative agreement with diagrammatic calculations [21,22] which predict a smaller m^* for the fully polarized system than for the unpolarized one, because the density and spin fluctuations which dress the particles are both reduced by polarization. Because of our partial polarization, a direct comparison is possible only for the nearly ferromagnetic model, where only the spin fluctuations are taken into account [1]. Béal-Monod and Daniel [23] have computed the temperature and field dependence of the magnetization M within this model up to order B^3. Inverting this relation for $M = M(B,T)$, and using Maxwell's relation $(\frac{\partial S}{\partial M})_T = -(\frac{\partial B}{\partial T})_M$, we deduced the polarization dependence of specific heat up to order m^4. As shown by Fig. 5, this roughly accounts for the observed behavior, up to $m \simeq 40\%$, when we normalize this change by the measured zero field specific heat [6]. At low polarization, the agreement is not surprising as the nearly ferromagnetic model correctly accounts for the experimental temperature dependence of the susceptibility [24]. The striking result is that it extends, up to large polarizations, the discrepancy for $m > 40\%$ being possibly due to the fact that Béal-Monod and Daniel use a small m expansion. Alternatively, if we directly use the experimental low field susceptibility $\chi(T)$ [25] to predict the polarization dependence of the specific heat up to order m^2, we account for the order of magnitude of the effect up to $m \approx 70\%$, as well as its small dependence on temperature [curves (D) in Fig. 5]. Repeating the calculation for other pressures, we expect the polarization effect at $T \simeq 80$ mK to depend little on pressure. This is qualitatively consistent with two preliminary experiments at 2 and 10 bars which employed the method described herein, and show a reduction of specific heat of 28 ± 5% and 33 ± 3% for $m = 50\%$ and $m = 74\%$, respectively.

In summary, our measurements support the idea that a large part of the specific heat of liquid ^{3}He is due to spin fluctuations. This contrasts with the assumption of the nearly localized model, and implies that a proper description of the low temperature specific heat of liquid ^{3}He should go beyond purely local effects.

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