Spin-Polarizing Concentrated ^{3}He-^{4}He Mixtures by the Rapid-Melting Method

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L. P. ROOBOL(*), S. C. STEEL(*), R. JOCHEMSEN(*), G. FROSSATI(*)
K. S. BEDELL(**) and A. E. MEYEROVICH(***)

(*) *Kamerlingh Onnes Laboratorium - Nieuwsteeg 18, 2311 SB Leiden*
The Netherlands
(**) *Los Alamos National Laboratory - T11, MS-B262, Los Alamos, NM 87545, USA*
(***) *Department of Physics, University of Rhode Island - Kingston, RI 02881, USA*

Abstract. – We have achieved high-spin polarizations in concentrated mixtures of ^{3}He in ^{4}He using the rapid-melting method originally suggested by Castaing and Nozières for pure ^{3}He. Polarizations well above the equilibrium value (greater than 10%) were obtained at $T \approx 350$ mK. The observed relaxation time $T_1 = 4000$ s was of the same order as the theoretical predictions of the bulk relaxation time, and was long enough to suggest that it should be possible to study the properties of these strongly polarized mixtures.

Producing a large equilibrium spin polarization ($\Delta \approx 1$) in a degenerate Fermi system, such as mixtures of ^{3}He dissolved in ^{4}He, normally requires a field sufficient to produce a magnetic energy μH of the same order as the Fermi energy $k_B T_F$. For weakly polarized ^{3}He-^{4}He mixtures under pressure $T_F = 2.1 x^{2/3}$ K, where $x = n_3/(n_3 + n_4)$ is the concentration of ^{3}He. Producing large polarizations by brute force thus requires either unreasonably large magnetic fields ($H \sim 440$ T for $\Delta = 1$ with $x = 0.06$) or very low concentrations of ^{3}He ($x \approx 10^{-4}$). However, it is possible to produce metastable samples with nonequilibrium polarizations by making use of the Castaing-Nozières effect [1]. For pure ^{3}He this method is now well established and has produced polarizations as high as $\Delta \sim 0.6$ [2, 3]. The technique produces large spin polarizations by the formation of solid ^{3}He which, because it shows nearly Curie-paramagnet behavior down to a few mK, can be strongly polarized in available magnetic fields and temperatures ($T = 2$ mK and $H = 10$ T gives $\Delta > 0.9$). The polarized solid is then rapidly melted in a time $t \ll T_1$ the nuclear relaxation time, producing a liquid which has a polarization corresponding to much higher magnetic fields than can be produced in the laboratory. In pure ^{3}He, T_1 is about 10^2 s at 300 mK [2], and increases to $2 \cdot 10^8$ s at 10 mK [4].

Concentrated mixtures of highly polarized ($30\% \leqslant \Delta \leqslant 50\%$) ^{3}He in ^{4}He are predicted to have some very interesting properties, such as a dramatic enhancement (up to a factor of 4) in the maximum solubility at full polarization [5]:

$$x_\uparrow^{2/3} = \left(\frac{x}{2}\right)^{2/3} + (N_3 a_0^3)^{2/3} \frac{m}{m_\uparrow^*}\left(1 - \frac{m_\uparrow^*}{m^*} 2^{-2/3}\right), \tag{1}$$

where $x_\uparrow$ is the limiting concentration of a completely polarized mixture in contact with completely polarized pure ^{3}He with density N_3, a_3^0 is the atomic volume in the mixture, m is the effective mass of ^{3}He quasi-particles in the mixture, and $m_\uparrow^*$ and m^* are the effective masses for pure polarized and nonpolarized ^{3}He, respectively. Additionally, the (p-wave pairing) superfluid transition temperature of the ^{3}He component could be increased by a large factor if the mixture could be polarized [6, 7]. These relatively high-concentration ^{3}He$_\uparrow$-^{4}He mixtures are a new type of Fermi liquid, with properties very different from both pure ^{3}He$_\uparrow$ and dilute ^{3}He$_\uparrow$-^{4}He mixtures [5, 8, 9].

We have applied a rapid-melting technique, similar to that used for pure ^{3}He, [2, 10] to ^{3}He-^{4}He mixtures with $x \cong 5.5\%$ and 8.5%. These mixtures were pressurized to 4.0 MPa at $T = 1.5$ K inside a plastic Pomeranchuk cell (shown in fig. 1) and cooled down to 60 mK by a
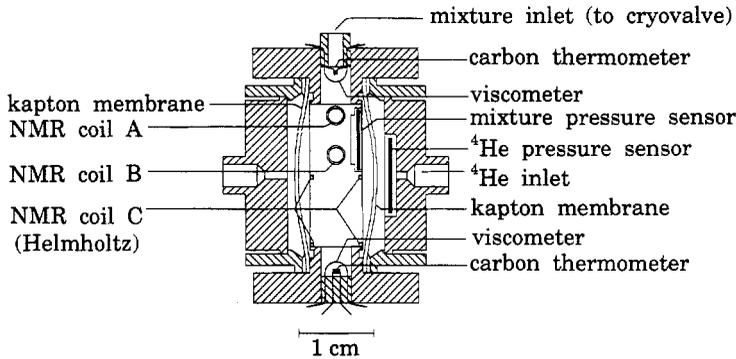

Fig. 1. – A cross-sectional view of the experimental Pomeranchuk cell, constructed of talc-filled epoxy. Except for the central slot with the NMR coils (shown here), most of the volume between the two circular Kapton membranes is occupied by a talc-filled epoxy filler to reduce the volume. The cryogenic valve (not shown) is mounted immediately above the cell.

dilution refrigerator in a magnetic field of 9.3 T. The magnetization was monitored by three NMR coils in different places of the cell (coils A, B and C in fig. 1), while the temperature was measured by two sliced Matsushita 51 Ω carbon resistance thermometers, one at the top and one at the bottom of the cell. Both thermometers were mounted inside the loop of a vibrating-wire viscometer [11] which detected the liquefaction of the mixture, while the mixture pressure and the ^{4}He pressure were measured with sapphire capacitive pressure transducers [12]. To minimize magnetic relaxation at the cell walls there was no sintered heat exchanger inside the cell; thermal contact to the mixing chamber of our dilution refrigerator was provided by a silver sinter heat exchanger inside the cryogenic valve [11] at the top of the cell. This valve also served to keep the number of atoms in the cell volume constant during the decompression.

During cooling, the ^{3}He quasi-gas separates in small liquid ^{3}He pockets, while the ^{4}He solidifies. The liquid ^{3}He distribution is more or less homogeneous throughout the cell as we could see by the homogeneous growth of the NMR signals of the three NMR coils. The pressure of the ^{3}He-^{4}He system drops to $P \sim 2.9$ MPa. The mixture is then compressed by increasing the pressure of the liquid ^{4}He outside the flexible walls of the Pomeranchuk cell. When the mixture pressure reaches the ^{3}He melting curve the cryogenic valve is closed. Further slow compression of the mixture produces additional cooling [13] by the Pomeranchuk effect. This is continued until the cooling stops at $T \cong 35$ mK and $P \sim 3.3$ MPa. During this process, the magnetization of the ^{3}He has slowly been increasing as it solidifies
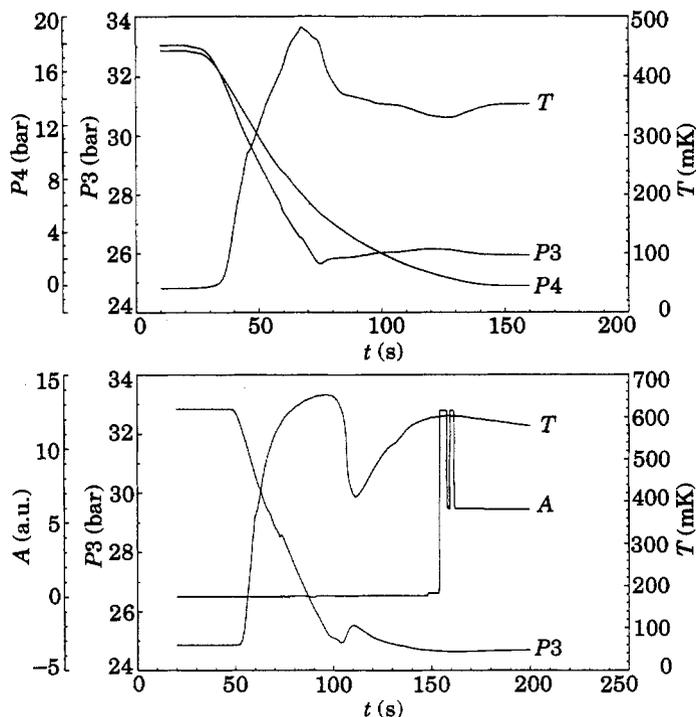
Fig. 2. – The behaviour of the sample during the decompressions. The upper plot shows the temperature T, mixture pressure $P3$, and external ^{4}He pressure $P4$ for the mixture with $x = 8.5\%$. The lower plot shows the temperature T, mixture pressure $P3$, and the viscometer amplitude A ($\propto \eta^{-1/2}$) for the mixture with $x = 5.5\%$. The viscometer amplitude is not meaningful until about $t = 160$ s when the liquid melts and the servo feedback loop driving the viscometer locks on to the resonance.

in the external 9.3 T field. This solid mixture is then rapidly decompressed by reducing the pressure of the liquid ^{4}He outside the cell to zero in about 100 s while keeping the cryogenic valve closed (see fig. 2). The viscometer confirms that the liquid does indeed melt. The larger Helmholtz NMR coil (Coil C in fig. 1) is then used to monitor the decay of the magnetization.

Figures 3 and 4 show the decay of the magnetization for decompressions made with $x = 8.5\%$ and 5.5%, respectively. We assumed that, after the decompression, the area of the continuous wave NMR signal varied as $A(t) = A_0 + A_1 \cdot \exp\left[-t/T_1\right]$ and fitted the parameters A_0, A_1 and T_1 to the data, and then divided the areas by A_0. Because the NMR area is proportional to the magnetization, $A/A_0 = \Delta/\Delta_0$, where Δ_0 is the equilibrium polarization of the mixture. Near 350 mK, Δ_0 is about 3% for the mixture concentrations we used. The relaxation time T_1 was found to be $1.2 \cdot 10^3$ s for $x = 8.5\%$ and $4.0 \cdot 10^3$ s for $x = 5.5\%$. The scatter in the data represents the uncertainty in the absolute value of Δ, which is $\pm 15\%$, while that in T_1 is $\pm 5\%$.

Figure 2 shows the behavior of the pressure, temperature and viscometer amplitude during the decompressions on an expanded time scale. The cooling occurring near $t = 100$ s indicates that the ^{4}He matrix is melting and that mixing of ^{3}He and ^{4}He occurs. This coincides with the moment at which the magnetization decay rate changes drastically, in fig. 3. The sharp drop shows a time constant of 10^2 s, similar to the T_1 of pure ^{3}He at 300 mK
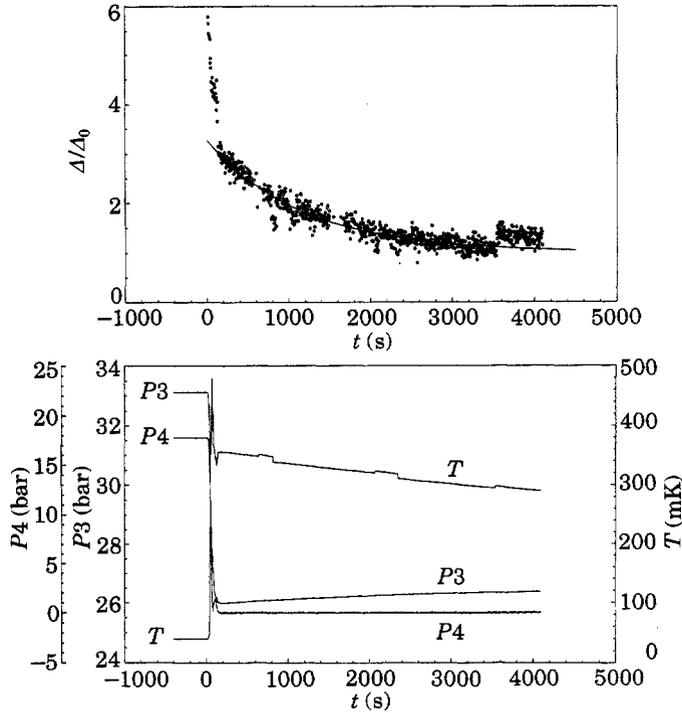
Fig. 3. – The $x = 8.5\%$ mixture relaxing back to equilibrium. The upper plot shows the normalized polarization Δ/Δ_0 (each point shown is the mean value of 4 successive data points) as well as the exponential fit. The lower plot shows the temperature T, the mixture pressure $P3$, and the external ^{4}He pressure $P4$.

and much smaller than T_1 for a bulk mixture. The fact that this drop is absent in fig. 4 suggests that the melting processes of the two decompressions are quite different.

The long-time constants (compared to pure ^{3}He) are to be expected, since there are only two processes to diminish the polarization: magnetic relaxation at the walls and bulk dipole-dipole relaxation. The theory of bulk dipole relaxation is rather well developed for dilute systems [14-18]. We can summarize the theoretical expressions for T_1 in the various temperature regimes as follows:

$$T_1^{\text{bulk}} = \frac{\hbar^2 \lambda^6}{\beta^4} \left(\frac{v}{\lambda}\right)^2 \frac{a_0^3}{xv\lambda^2} f(T) . \tag{2}$$

Here v is the characteristic velocity of the particles with magnetic moment β and atomic volume a_0^3, λ is the de Broglie thermal wavelength of the particles and x is the molar concentration of ^{3}He. At high temperatures $\lambda \sim a_0$, but in the quantum regime (both in the degenerate and in the Boltzmann case) λ is of the order of the de Broglie wavelength, $\lambda = \hbar/(m^* v)$. In the degenerate limit $T \ll T_F$ (where $T_F = k_F^2/2m^*$ is the Fermi temperature), the function $f(T) = b_0(T_F/T)^2$ [14-17], while for $T > T_F$ it is independent of temperature $f(T) = b_1$ [18, 19]. The values for the temperature independent coefficients b_0 and b_1 can be found in ref. [14-19]. In the low-temperature limit T_1 is nearly independent of the concentration x, while at high temperature T_1 will vary as x^{-1}. This theoretical estimate gives a dipole relaxation time $T_1^{\text{bulk}} \sim 10^4$ s for $T \simeq 350$ mK. In this temperature range we
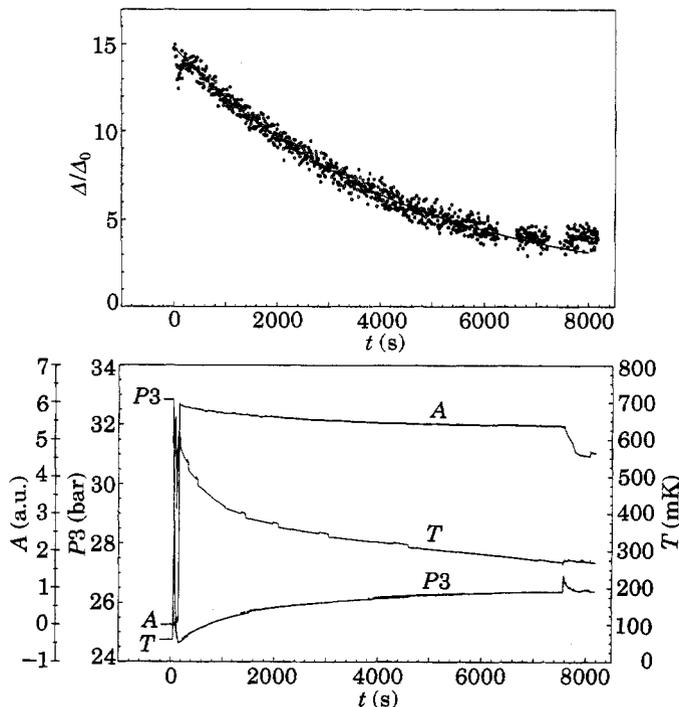
Fig. 4. – The $x = 5.5\%$ mixture relaxing back to equilibrium. The upper plot shows the normalized polarization Δ/Δ_0 (each point shown is the mean value of 4 successive data points) as well as the exponential fit. The lower plot shows the temperature T, the mixture pressure $P3$, and the viscometer amplitude A $(\propto \eta^{-1/2})$.

would expect the T_1^{bulk} to vary as x^{-1}, thus giving a 60% reduction in T_1^{bulk} when going from concentration $x = 5.5\%$ to $x = 8.5\%$.

The value of this estimate is in rough agreement with the experimental data, however, the observed values of T_1 are somewhat shorter and show a more pronounced concentration dependence: $T_1(5.5\%)/T_1(8.5\%) \sim 3.3$. This discrepancy is too big to be explained by the uncertainties in the experimental values of x (which are known to within $\pm 10\%$) or by the fact that eq. (2) is derived in the limit of low concentrations. The most likely explanation is that the experimentally observed relaxation rate includes a substantial contribution from relaxation occurring at the walls of the cell.

If we take the spin diffusion data from Murdock, Mountfield and Corrucini[20] and extrapolate to 350 mK, we get a spin diffusion constant $D = 3.5 \cdot 10^{-4}$ cm^2/s, which gives a spin diffusion time $\tau \simeq 70$ s for our experimental cell. A theoretical estimate for dilute systems [5] gives a similar value: $\tau \sim 10^2$ s. Thus, if there is some spin relaxation at the walls, it would not be surprising if it were to dominate the observed relaxation rate T_1^{-1}. In these

TABLE I. – *A summary of some relevant parameters for the two concentrations measured ($H = 9.3$ T).*

x	5.5%	8.5%
T_F (mK)	$3 \cdot 10^2$	$4 \cdot 10^2$
$\mu H/kT_F$	2.4%	1.8%
T_1 (s)	$4.0 \cdot 10^3$	$1.2 \cdot 10^3$

experiments there were almost no metallic surfaces in contact with the sample, and the walls of the cell were certainly coated with a few layers of pure ^{4}He. As a result, the observed relaxation times were reasonably close to the above estimates of bulk dipole-dipole relaxation, suggesting that, at worst, the surface relaxation rate was comparable to that in the bulk.

Conclusion. – We have shown, for the first time, that concentrated ^{3}He-^{4}He mixtures can be strongly polarized, by applying the Castaing-Nozières method and we have measured relaxation times which are one order of magnitude longer than for pure ^{3}He at the same temperature, making it possible to do extensive measurements on these novel polarized Fermi systems.

If extrinsic relaxation at the walls can be kept small, a T^{-2}-dependence of T_1^{bulk} might lead to relaxation times in the 10^7 s range at a few mK. The heating due to the relaxation of the magnetization would then be small enough that further cooling by means of a nonmagnetic heat exchanger could be possible eventually entering the region where the ^{3}He component would undergo *p*-wave superfluidity.

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