Steady-State Production of High Nuclear Polarization in ^{3}He-^{4}He Mixtures

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We demonstrate a new technique for producing high, steady-state nuclear polarization in liquid ^{3}He-^{4}He mixtures. Polarization of 56% has been obtained in a 4% ^{3}He-in-^{4}He mixture at 200 mK, roughly 3 times that achieved to date by other means. Polarization loss due to spin relaxation is compensated for by driving a constant circulation of ^{3}He between the liquid sample and a room-temperature optical pumping volume. The weak binding properties of cesium are used both to inhibit surface relaxation and to prevent refluxing ^{4}He vapor from confining ^{3}He atoms to the low-temperature part of the cell.

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The production and study of spin-polarized ^{3}He fluids have attracted widespread attention for many years [1–3]. One reason for this interest is that atomic-scale exchange processes within the fluid are influenced by the nuclear spin polarization M. It is thus possible to modify the low-temperature equilibrium and transport properties of the fluid by changing M.

Dense ^{3}He fluids such as pure liquid ^{3}He or liquid ^{3}He-^{4}He mixtures with ^{3}He concentrations x_3 greater than 1% are degenerate Fermi fluids below temperatures of order 100 mK. This fact rather severely limits the maximum *equilibrium* nuclear polarization which can be obtained in these systems using available laboratory magnetic fields: $M \approx 5\%$ is obtained for $B = 10$ T [3]. One can, however, use magnetic fields to produce large equilibrium polarizations in very dilute ($x_3 < 10^{-3}$) liquid ^{3}He-^{4}He mixtures or in solid ^{3}He or ^{3}He-^{4}He mixtures at temperatures of order 1 mK. Very high *nonequilibrium* nuclear polarizations (up to 85%) can be obtained in room temperature helium gases using laser optical pumping techniques [4].

There are currently two methods for producing high *transient* nuclear polarization in liquid ^{3}He and ^{3}He-^{4}He mixtures. One method relies on the fast condensation of an optically pumped gas [5] while the other involves the rapid melting of a solid polarized in a high magnetic field [6]. Both methods suffer from the problem that it is often difficult to measure and control sample parameters such as pressure and temperature on time scales shorter than the polarization decay time T_1.

One would thus like to develop a means of producing high *steady-state* (albeit nonequilibrium) nuclear polarization in dense ^{3}He fluids. This requires the continuous production of nuclear magnetization to compensate for inevitable relaxation processes. The only such technique developed prior to the work reported here is the use of fractional distillation to separate the two nuclear spin states of ^{3}He in a phase separated ^{3}He-^{4}He liquid mixture [7]. To date, polarizations of nearly 20% have been achieved by this means [8].

In this Letter we report the development of a new technique which we have used to produce large steady-state ^{3}He nuclear polarizations in liquid ^{3}He-^{4}He mixtures. The helium is sealed within a cell that extends from a room temperature optical pumping volume (OPV) to a low-temperature volume where a fraction of the sample is liquid. Laser optical pumping [9] is used to polarize the ^{3}He spins in the OPV. A novel feature of the technique is that ^{3}He is continuously circulated between the OPV and the liquid sample at a rate sufficient to override spin relaxation processes. In contrast to the spin-distillation method, we are forced to operate at the saturated vapor pressure of the liquid. The ^{3}He concentration x_3 in the liquid, however, is not restricted to saturation conditions. In principle the techniques we describe could even be applied to pure liquid ^{3}He.

The experimental cell is shown in Fig. 1. It consists of an elongated loop fashioned from Pyrex glass tubes joined to the Pyrex OPV. A total of 35 μmol of ^{3}He and 1.5 mmol of ^{4}He are sealed within the cell. The U shaped lower end of the cell is thermally linked to the mixing chamber of a dilution refrigerator. Under operating conditions, liquid fills the lower 5 cm of the cell while the remaining volume is filled with vapor. The temperatures T_{evap} and T_{cond} of the liquid-vapor interfaces are independently controlled using heaters such that $T_{\mathrm{evap}} > T_{\mathrm{cond}} \approx 1$ K. ^{3}He is continuously evaporated at the interface which is at temperature T_{evap} and condensed at the other. The low thermal conductances of the sample and the cell walls allow liquid in the base of the cell to be cooled to a temperature T_0 as low as 200 mK.

The liquid helium mixture is superfluid at the operating temperature of the cell, and thus the ^{4}He chemical potential $\mu_4(x_3, T)$ within the liquid is uniform. This imposes a relationship between the local values of x_3 and T. We have computed the distribution of ^{3}He in the cell as a function of the liquid-vapor interface temperature using the thermodynamic properties of ^{3}He-^{4}He mixtures [10]. The pressure and composition of the saturated vapor [Fig. 2(a)] are determined by the liquid temperature and composition *at the interface*, Fig. 2(b). Heating the interfaces enhances the ^{3}He concentration in the coldest volumes at the expense of other regions ("heat flush"

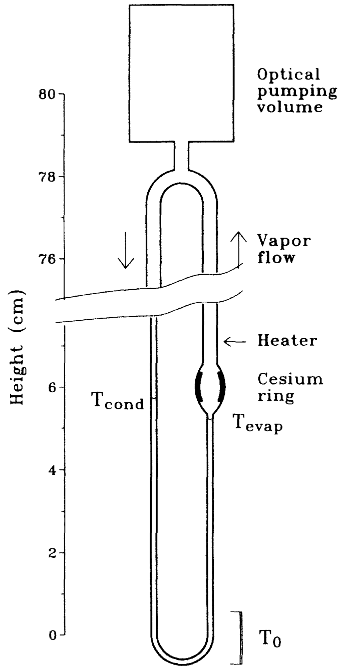

FIG. 1. Sketch of the experimental cell. The 200 cm^3 optical pumping volume is at room temperature, while the regions labeled T_{cond}, T_{evap}, and T_0 are at independently controlled cryogenic temperatures. Locations of the liquid-vapor interfaces in the two arms are shown under typical circulation conditions ($T_0 \approx 200$ mK , $T_{cond} \approx 1$ K, $T_{evap} \approx T_{cond} + 50$ mK).

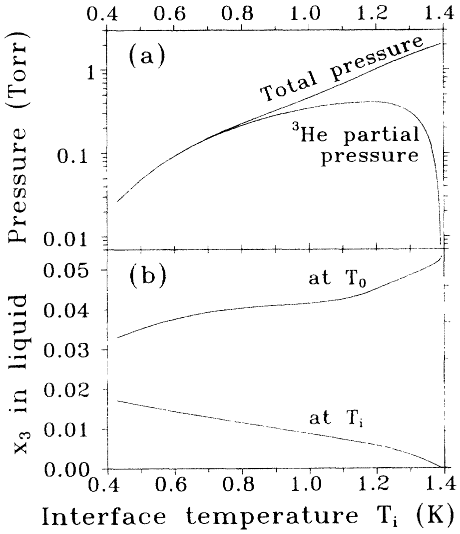

FIG. 2. Calculated (a) total and ^{3}He partial vapor pressures above the liquid-vapor interface and (b) ^{3}He concentration in the liquid near the interface and at the cell bottom. Quantities are plotted as a function of a common liquid-vapor interface temperature $T_i \approx T_{cond} \approx T_{evap}$. These results depend upon the thermodynamic properties of ^{3}He-^{4}He mixtures, as well as the volumes and filling quantities for the cell.

effect). As shown in Fig. 2(a), the vapor composition changes from almost pure ^{3}He to almost pure ^{4}He as the interface temperatures are warmed from 0.8 to 1.4 K.

Optical pumping of gaseous ^{3}He and ^{3}He-^{4}He mixtures is most efficient at pressures of 1 Torr or less and ^{3}He concentrations of 10% or higher [9]. These conditions are met over a range of interface temperatures near 1 K as shown in Fig. 2(a). Provided that the ^{3}He concentration in the OPV is the same as it is just above the liquid-vapor interface, high nuclear polarizations can be achieved within the OPV. Note [Fig. 2(a)] that a pressure difference of order 0.1 Torr is generated by a small temperature difference between T_{evap} and T_{cond}. The cell was designed such that this would be sufficient to circulate ^{3}He fast enough to override nuclear relaxation.

An essential feature of the experiment is the use of cesium metal in the cell. One function of the Cs is to inhibit nuclear spin relaxation on the cell walls. The cell was sealed after being filled with the helium sample and a small quantity of Cs. The Cs was then melted and brought successively into contact with all interior surfaces of the cell, leaving an invisible film or surface treatment. Finally, the bulk Cs in the cell was formed into a ring just above the warmer of the two liquid-vapor interfaces (Fig. 1); the sample, therefore, only contacts Cs-treated Pyrex and bulk Cs. Experiments have shown that both of these surfaces act as weak binding surfaces which exhibit low surface relaxation rates for pure ^{3}He [11]. The long T_1 values measured in the current experiment demonstrate that a similar effect occurs for ^{3}He-^{4}He mixtures.

Cesium plays another role crucial to the success of the experiment; metallic Cs is not wet by liquid ^{4}He below a temperature of order 2 K [12]. The bulk Cs ring breaks the film of superfluid helium which would otherwise flow toward warmer temperatures. Were it not for this ring the ^{4}He film would eventually evaporate and reflux toward the cold liquid as vapor. This process, known as the HEVAC (helium vapor compression) effect, is an efficient means of confining ^{3}He atoms to the liquid and the vapor just above the interface [10,13]. The Cs ring thus allows ^{3}He atoms to reach the OPV and become polarized. The addition of small quantities of ^{3}He to the liquid ^{4}He is not expected to induce wetting except at very low temperatures where reentrant wetting has been predicted [14] and observed [15]. We estimate that the Cs ring used in our experiment should not be wet by ^{3}He-^{4}He mixtures with $x_3 < 3\%$.

The composition of the vapor in the OPV was measured using the absorption of laser light that could be tuned to atomic resonances of either isotope. During preliminary experiments in cells without Cs (H_2 wall coatings were used to inhibit relaxation) the ^{3}He concentration in the OPV was too low to be detected, demonstrating a strong HEVAC effect. In later experiments that included a Cs ring, the ^{3}He concentration in the OPV was much higher and in reasonable agreement with that expected just above the liquid-vapor interface. We believe this to be indirect evidence that Cs metal is not wet under the experimental conditions.

The ^{3}He nuclear polarization in the liquid was monitored using pulsed nuclear magnetic resonance (NMR). Coils external to the nonmagnetic cryostat provided a 400 μT vertical B_0 field, uniform to better than 0.1% over the entire cell. Additional coils permitted the application of field gradients of order 10^{-4} T/m in any direction. The NMR spectrometer employed separate transmitter and receiver coils. The transmitter coil produced a 13 kHz B_1 field over the entire liquid volume while the smaller receiver coil was primarily sensitive to the magnetization of the liquid in the base of the cell.

The high magnetization $\mathcal{M}$ of the polarized liquid produces demagnetizing fields which shift the NMR frequency ν away from the Larmor frequency $\nu_0 = \gamma B_0/2\pi$. We use this effect to measure $\mathcal{M}$ and thereby infer the polarization M. For an ellipsoidal sample, the frequency shift following a small tipping-angle pulse is $\nu - \nu_0 = A(3\cos^2\theta - 1)\gamma\mu_0\mathcal{M}/2\pi$ where θ is the angle between the symmetry axis of the sample and the static field and A is a shape factor which varies between $-1/2$ for an infinite sheet and $+1/4$ for an infinite cylinder. This shift has been observed for sheetlike drops of liquid formed by condensation of polarized ^{3}He gas [5]. For dilute ^{3}He-^{4}He liquid mixtures at low pressure $\gamma\mu_0\mathcal{M}/2\pi = (9.56\ \text{kHz})x_3M$.

The lower end of the present cell consists of a tube of inner diameter 0.6 mm, formed into a 2 cm wide U (Fig. 1). It is reasonably approximated by a continuous series of independent long cylinders, with θ ranging from zero in the vertical segments to $\pi/2$ at the bottom. Within this approximation one expects to observe two peaks in the NMR spectrum, located at the extremal values of ν. These peaks are split by $\Delta\nu = (7.17\ \text{kHz})x_3M$.

We have observed NMR spectra of this form, as shown in Fig. 3. Two principal peaks are visible in this small tipping-angle spectrum; they merge together when the tipping angle is increased to $\pi/2$. Shifts in the peak locations caused by applying field gradients indicate that the low-frequency peak is indeed produced by ^{3}He in the cell bottom, while the other is produced by ^{3}He in the vertical tubes. This spectrum was observed with the magnetization parallel to $\mathbf{B}_0$; we find that the spectrum is reflected about ν_0 when the magnetization is inverted, in agreement with the analysis above.

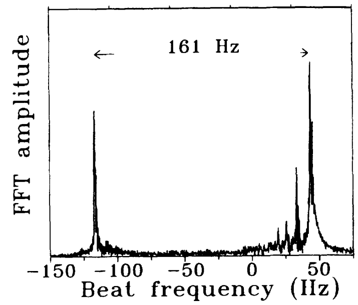

FIG. 3. Fourier transform of the free induction decay following a single $\pi/32$ NMR tipping pulse. The beat frequency is the difference between the signal frequency and a reference frequency of order 13 kHz. The dominant frequency splitting $\Delta\nu = 161$ Hz corresponds to a nuclear polarization $M = 56\%$ for the calculated ^{3}He concentration $x_3 = 0.04$. Low-polarization experiments in the same run indicated that the Larmor frequency produces a beat at approximately -50 Hz.

The peaks we observe are extremely narrow. The corresponding free-induction decays are several seconds long. Spectra such as that shown in Fig. 3 exhibit a detailed structure not predicted by the infinite-cylinder approximation. A proper analysis of the spin dynamics must consider the precession of each spin in the dipolar field produced by the remainder of the sample. This analysis predicts the existence of a set of modes which depend upon sample shape and field gradients. Similar "magnetostatic modes" have previously been observed in highly polarized solid ^{3}He [16] and in ferrites [17]. We have carried out a numerical analysis of the magnetostatic modes of our experimental cell which successfully reproduces several features of the experiment; details will be published elsewhere. A key result of this analysis is that the splitting between the dominant modes is accurately given by the formula for $\Delta\nu$ in the infinite-cylinder approximation. We thus take $\Delta\nu$ to be a quantitative measure of the sample magnetization $\mathcal{M}$.

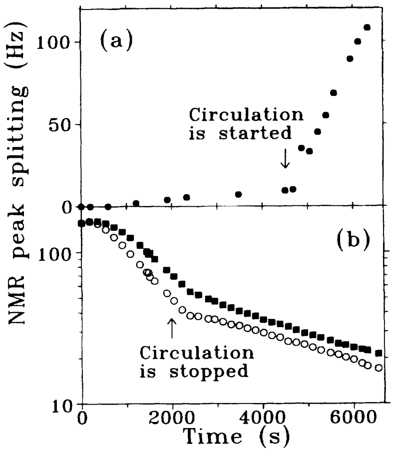

FIG. 4. Evolution of the NMR peak splitting $\Delta\nu$ (which measures the magnetization of the sample) under various optical pumping and circulation conditions. In (a), gas in the OPV is constantly pumped. ^{3}He circulation is started at $t = 4500$ s and results in a sharp increase in the rate of polarization buildup. In (b), optical pumping is stopped at $t = 0$ and the polarization in the OPV is maintained at zero thereafter. An abrupt decrease in the rate of decay of $\Delta\nu$ is observed when the circulation is stopped. The magnetization on the condensing side (open circles) as measured by $\Delta\nu$ decays earlier in time than that on the evaporating side (filled squares) due to the sense in which ^{3}He atoms circulate.

To characterize the ^{3}He circulation, we have studied the time evolution of the magnetization by recording the response of the system to a series of small tipping-angle pulses. Figure 4 shows the results of two such experiments. In Fig. 4(a) circulation is initiated by heating T_{evap} to a temperature 50 mK above T_{cond}. This causes the magnetization of the liquid to increase with a time constant of order 1500 s (the limiting value is not attained in the figure). Figure 4(b) illustrates the decay of the liquid magnetization caused by circulation while the polarization in the OPV is forced to remain at zero.

In conclusion, we have demonstrated that a liquid ^{3}He-^{4}He mixture can be highly polarized by combining thermally driven ^{3}He circulation with room-temperature optical pumping. The use of Cs is crucial to the experiment; it serves to inhibit ^{4}He vapor refluxing which would otherwise keep the ^{3}He from reaching the OPV and to reduce surface spin relaxation [$T_1 > 5000$ s in Fig 4(b)]. The largest frequency splitting we have measured to date is more than 160 Hz, implying a ^{3}He nuclear polarization $M = 56\%$ at a temperature of 200 mK with $x_3 = 0.04$. Although x_3 is not directly measured in this experiment, the values plotted in Fig. 2(b) are relatively insensitive to experimental parameters. The greatest uncertainty arises from imperfect knowledge of the temperature distribution in the cell, from which x_3 is inferred.

In addition to the magnetostatic modes mentioned above, we have observed a number of interesting effects which we attribute to the nonlinear spin dynamics of the highly polarized fluid. The most striking of these is an instability leading to the spontaneous appearance of a precessing transverse magnetization [18]. Another is the absence of ordinary spin echoes following two resonant pulses separated by less than the free induction decay time. Understanding these new effects will likely require further experiments, as well as an analysis of the spin dynamics which incorporates both nonlocality and nonlinearity.

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