Nonequilibrium spin polarization of liquid ^{3}He and of ^{3}He–^{4}He solutions

A. É. Meierovich

Institute of Physical Problems, Academy of Sciences of the USSR

(Submitted 4 June 1987)

Pis'ma Zh. Eksp. Teor. Fiz. **46**, No. 2, 77–79 (25 July 1987)

Some unconventional ways for producing liquid phases of ^{3}He with a pronounced nonequilibrium spin polarization are discussed. The depolarization time of such systems is estimated.

Spin-polarized quantum ^{3}He systems have been the subject of substantial theoretical work (see, for example, the reviews in Ref. 1), but our experimental knowledge of them is so far rather limited. The difficulties are in preparing the various phases of ^{3}He$\uparrow$. An external magnetic field can strongly polarize only solid ^{3}He and dilute ^{3}He–^{4}He solutions at ultralow temperatures. Long-lived systems with a nonequilibrium polarization are prepared by quickly melting solid ^{3}He$\uparrow$ (Ref. 2), by optical pumping in gaseous ^{3}He (Ref. 3), and by dynamic polarization of liquid ^{3}He (Ref. 4). In this letter we wish to discuss some alternative methods for polarizing ^{3}He, based on

 0021-3640/87/140092-04$01.00

the polarization of a ^3He–^4He solution by a strong magnetic field, followed by the rapid precipitation of ^4He from the mixture. We also discuss the lifetime of ^3He–^4He solutions with a nonequilibrium polarization.

1. A magnetic field $H \lesssim 10$ T strongly polarizes a ^3He–^4He solution with a ^3He concentration $x \lesssim 0.1\%$ at $T \lesssim 10$ mK. The ^3He concentration can then be raised significantly, at an essentially constant degree of polarization, by rapidly removing a large part of the ^4He through a superfluid gap. The concentration and degree of polarization of the ^3He in the solution can be monitored, and the depolarization time measured, by measuring the osmotic pressure. The difference between the osmotic pressures of two cells which are linked through a superfluid gap and which contain degenerate ($T \ll T_0$) ^3He–^4He solutions with degrees of polarization $\alpha_{1,2}$ and with $n_{1,2}$ ^3He atoms per unit volume can be written as follows when the concentrations are not too high:

$$\Pi = \Pi_0(n_1, \alpha_1) - \Pi_0(n_2, \alpha_2), \qquad \Pi_0(n, \alpha) = (3\pi^2)^{2/3}(\hbar^2/10M)n^{5/3}$$
$$\times [(1+\alpha)^{5/3} + (1-\alpha)^{5/3}] + (\pi a\hbar^2/M)n^2(1-\alpha^2),$$

where M and a are the effective mass and s-scattering length of the ^3He quasiparticles [at $T \gg T_0$ and in the limit $na^3 \to 0$ we have $\Pi_0(n,a) \approx nT$, and this pressure is essentially independent of the polarization]. When the ^4He is removed from the solution through a superfluid gap, the solution temperature rises. The ratio of the final and initial temperatures under the condition $\alpha = \text{const}$ is essentially independent of α: $T_f/T_i \approx (x_f/x_i)^{2/3}$ ($x_{f,i}$ are the final and initial concentrations). If the ^4He is removed into a volume with concentrated ^3He, a corresponding cooling will occur in this volume.

2. The method described above for producing a nonequilibrium polarization can be altered by initially using a magnetic field to polarize a solution with a ^3He concentration x close to the concentration for stratification into pure ^3He and a solution with a limiting concentration $x_c(\alpha)$ (in fields $H \lesssim 10$ T the initial polarization is $\alpha \ll 1$). If the ^4He is then rapidly removed through a superfluid gap, a stratification begins when the concentration $x_c(\alpha)$ is reached. The polarizations of the two phases that arise are related to the original polarization α by (under the condition $\alpha_{1,2} \ll 1$)

$$\alpha_{1,2} = \alpha\chi_{1,2}N/(\chi_1 N_1 + \chi_2 N_2), \qquad \alpha_1/\alpha_2 = \chi_1/\chi_2,$$

where the subscripts 1 and 2 refer to the pure ^3He and the solution, $\chi_{1,2}$ are the susceptibilities per ^3He particle, and $N_{1,2} = n_{1,2}v_{1,2}$ are the numbers of ^3He atoms in each of the phases ($N = N_1 + N_2 = \text{const}$). Since $\chi_1 \ll \chi_2$, when the ^4He is removed, the polarization of the remaining solution will increase, and in the limit $N_2 \ll N$, $N_1 \to N$ the polarization can reach a value $\alpha_2 = \alpha\chi_2/\chi_1 \gg \alpha$. The limiting polarization of the pure phase, $\alpha_1(N_2 \to 0) = \alpha$, is $\chi_2/\chi_1 \gg 1$ times its equilibrium value in the same field. The polarization also leads to a change in the limiting solubility of ^3He in ^4He ($\alpha_{1,2} \ll 1$):

$$\delta x_c(\alpha) = (\alpha^2 N^2/2)(\chi_2 - \chi_1)/(\chi_1 N_1 + \chi_2 N_2)^2(\partial\mu_2/\partial x)_P \quad (\alpha_{1,2} \ll 1).$$

The derivative of the chemical potential of ^{3}He in solution with respect to its concentration can be determined either from the theory of weak solutions, $[\partial\mu/\partial x \sim (2/3) T_0/x]$, or from experimental data on the behavior $x_c(P,T)$ (P is the pressure). The shift of the stratification curve mentioned above is significantly greater than the shift of the curve in the case of an equilibrium polarization in the same field.

3. Yet another way to increase the ^{3}He concentration in a solution at a constant polarization is to raise the pressure to a level above the pressure at which pure ^{4}He would crystallize but below the pressure at which ^{3}He$\uparrow$ would crystallize. In this case, only the ^{4}He will crystallize, and if the liquid-crystal interface moves at a velocity lower than the diffusion rate of the ^{3}He in the liquid solution, but faster than the depolarization rate, then all of the ^{3}He$\uparrow$ will remain in the liquid phase, and its concentration will increase at an essentially constant polarization. In this case we are left with the open question of whether a pronounced polarization will lead to a penetration of ^{3}He atoms into the solid ^{4}He above a certain threshold, even at low temperatures.[5]

4. The possibility of carrying out the experiments described above is limited by the depolarization time of the spin system. The primary mechanism for the depolarization of weak solutions at low temperatures is magnetic relaxation at the cell walls. In the absence of convection the corresponding time is determined by spin diffusion, $\tau \sim L^2/Dw$, or—if the mean free path is long—by the transit time (L is a characteristic dimension of the cell, and w is the probability for spin flip upon a collision with the wall). Numerically, the spin diffusion coefficient is $D \sim 10^2 x^{2/3}/a^2T^2$ cm^2/s, and the mean free path is $l \sim 10^{-1} x^{1/3}/a^2T^2$ cm (a is expressed in angstroms, and I in millikelvins). If the polarization is pronounced, the mean free path is even larger (by a significant amount).

We can estimate a lower limit on τ. The shortest times τ correspond to the case $T < 1$ mK, in which we have $l \gtrsim L$. We are interested in a high nonequilibrium degree of polarization, $T/T_0 \ll \alpha \lesssim 1$, in which case factors T/T_0 do not arise upon inelastic scattering at the wall. The spin of a ^{3}He quasiparticle may flip as the result of a magnetic dipole interaction with an electron paramagnetic center at the wall. If the concentration of these centers is at the atomic level, the depolarization time in the ballistic regime, $T \lesssim 1$ mK, is on the order of a second, if we ignore the presence of a narrow barrier (a layer of essentially pure ^{4}He) at the wall. Other possibilities are indirect processes involving the presence of a few mobile ^{3}He atoms in the thin layer of helium which has solidified at the wall. Such atoms are magnetically (and strongly) bound to paramagnetic centers of the wall (or to ^{19}F nuclei), and they undergo an effective exchange interaction with ^{3}He quasiparticles in the volume. The number of such atoms falls off with the temperature, in proportion to $\exp(-\Delta/T)$. If $\Delta \gtrsim 0.1$ K, the corresponding processes are inconsequential at $T \lesssim 1$ mK. At $\Delta \lesssim 10$ mK, the indirect processes become the governing factors; τ falls off sharply and is an exponential function of the temperature.

Large values of τ should therefore be expected either in the absence of a significant convection if the temperature is moderately low ($T \gtrsim 10$ mK; short mean free paths) or (better) if there is a reliable magnetic shielding of the walls. A condensation of several layers of molecular hydrogen on the walls would apparently be the most convenient approach here.

I wish to thank Yu. Anufriev, F. Laloë,[1)] and M. Chapellier for constructive discussions.

[1)] In the course of a discussion of the results of this study, F. Laloë stated that some similar arguments had been expressed independently by W. Gully (unpublished results; I do not know the details).

[1]A. E. Meyerovich, "Spin-polarized ^{3}He–^{4}He solutions," in Progress in Low Temperature Physics, Vol. 11 (ed. D. F. Brewer), North-Holland, Amsterdam, 1987, p. 1; A. E. Meyerovich, "Spin-polarized phases of ^{3}He," in Anomalous Phases of ^{3}He (ed. W. P. Halperin and L. P. Pitaevski), North-Holland, Amsterdam (to be published).

[2]B. Castaing and P. Nozieres, J. Phys. (Paris) **40**, 257 (1979); G. Shumacher D. Thoulouze, B. Castaing, Y. Chabre, P. Segransan, and J. Joffrin, J. Phys. (Paris) **40**, L143 (1979); M. Chapellier, G. Frossati, and F. B. Rasmussen, Phys. Rev. Lett. **42**, 904 (1979); H. Godfrin, G. Frossati, A. S. Greenberg, B. Hebral, and D. Thoulouze, Phys. Rev. Lett. **44**, 1695 (1980); G. Bonfait, L. Puech, A. S. Greenberg, G. Aska, B. Castaing, and D. Thoulouze, Phys. Rev. Lett. **53**, 1092 (1984); A. Dutta and C. N. Archie, Phys. Rev. Lett. **55**, 2949 (1985).

[3]M. Leduc, P. J. Nacher, S. B. Crampton, and F. Laloë, "Nuclear polarization of ^{3}He gas at low-temperature by optical pumping," in Quantum Fluids and Solids (ed. E. D. Adams and G. G. Ihas), American Institute of Physics, New York, 1983, p. 179.

[4]A. Schuhl, S. Maegawa, N. M. Meisel, and M. Chapellier, Phys. Rev. Lett. **54**, 1952 (1985); S. Saito, M. Okuyama, and T. Satoh, Phys. Rev. Lett. **55**, 1757 (1985).

[5]A. E. Meyerovich, J. Low Temp. Phys. **53**, 487 (1983).

Translated by Dave Parsons