

# <sup>3</sup>He melting in a magnetic field

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The almost-localized Fermi liquid theory is generalized to low temperatures and arbitrary magnetic fields. The results are used to analyze previous experimental studies, in which strongly polarized liquid <sup>3</sup>He obtained by melting in a magnetic field was investigated. © 1996 American Institute of Physics. [S1063-7761(96)01001-7]

## 1. INTRODUCTION

The spin-polarized Fermi liquid <sup>3</sup>He has attracted the attention of both theoreticians and experimentalists for a long time.<sup>1</sup> The polarization of equilibrium liquid <sup>3</sup>He in magnetic fields accessible to experiment is quite low. For example, its specific magnetic moment in a ≈6 T field at  $T=0.1$  K amounts to only 3% of the peak value.<sup>2</sup> Therefore, a method for obtaining strongly polarized liquid <sup>3</sup>He by rapidly melting the polarized solid phase in a magnetic field was proposed in Ref. 3. This method was used widely in subsequent experimental studies,<sup>2,4,5</sup> and it is presently the only method for obtaining quasiequilibrium, strongly polarized liquid <sup>3</sup>He in macroscopic volumes.<sup>6</sup> Simple models of the melting process were constructed in Refs. 7 and 8; however, they employed rather arbitrary assumptions instead of the equation of state of liquid <sup>3</sup>He. In Ref. 5 liquid <sup>3</sup>He with a very high magnetic moment was obtained after rapid melting. A procedure for calculating the effective magnetic field from the heat evolved when the magnetic moment relaxes was used in that work. We show below that such an approach is not correct.

Along with the paramagnon,<sup>9</sup> mean-field,<sup>10</sup> induced interaction,<sup>11</sup> and nearly metamagnetic <sup>3</sup>He theories,<sup>12</sup> the almost localized Fermi liquid model,<sup>13</sup> which is also sometimes called the Brinkman–Rice gas model, has been employed to describe the state of equilibrium polarized liquid <sup>3</sup>He. This model was successfully used to describe the properties of liquid <sup>3</sup>He at nonzero temperatures in the absence of a magnetic field.<sup>14</sup>

In Sec. 2 of this paper the almost-localized Fermi liquid model is generalized to low temperatures (much lower than the effective Fermi energy) and arbitrary magnetic fields. In Sec. 3 the melting process of <sup>3</sup>He is modeled in a magnetic field with consideration of the equation of state of polarized liquid <sup>3</sup>He. The pressure depression for rapid melting is calculated in Sec. 4, and then the values obtained are compared with the experimental results in Refs. 2 and 4. The results in Ref. 5 are also discussed in that section. The main conclusions are summarized in Sec. 5.

## 2. ALMOST-LOCALIZED FERMI LIQUID AT NONZERO TEMPERATURES IN AN ARBITRARY MAGNETIC FIELD

Liquid <sup>3</sup>He is a system of spin-1/2 fermions, that exhibit a strong short-range interaction. Such systems are often described by the Hubbard Hamiltonian<sup>12</sup>

$$H = \sum_{\mathbf{k}, \sigma} \varepsilon_{\mathbf{k}, \sigma} a_{\mathbf{k}, \sigma}^{\dagger} a_{\mathbf{k}, \sigma} + U \sum_{i, \sigma} n_{i\sigma} n_{i-\sigma}, \quad (1)$$

where  $a_{\mathbf{k}, \sigma}^{\dagger}$  ( $a_{\mathbf{k}, \sigma}$ ) is the creation (annihilation) operator of a fermion with spin  $\sigma$  and wave vector  $\mathbf{k}$ ,  $n_{i\sigma}$  is the operator of the number of particles with spin  $\sigma$  in the coordinate representation, i.e.,  $n_{i\sigma}$  corresponds to the set of the vectors  $\mathbf{R}_i$  in the coordinate space, which are related to the vectors  $\mathbf{k}$  by Fourier transformation, and  $U$  is the interaction constant. This Hamiltonian is usually used for a system of interacting fermions in a lattice with an initial  $s$  band. However, in the Gutzwiller approximation to the solution of the Hamiltonian (1), on which the almost-localized Fermi liquid model is based, the structure of the lattice is inconsequential. It is only important that the number of nearest neighbors is constant and that the coordinate wave functions (the Wannier functions) are of the  $s$  type. This accounts for the applicability of this model to liquid <sup>3</sup>He.<sup>13</sup>

The ground-state energy of an almost-localized Fermi liquid, normalized to a single lattice site or, in our case, to a single vector  $\mathbf{R}_i$ , has a simple form:<sup>13</sup>

$$E = \sum_{\sigma} q_{\sigma} \bar{\varepsilon}_{\sigma} + Ud, \quad (2)$$

where  $\bar{\varepsilon}_{\sigma}$  is the mean kinetic energy of the electrons with spin  $\sigma$ ,  $d$  is the fraction of the vectors  $\mathbf{R}_i$  occupied by two electrons with different spins. Liquid <sup>3</sup>He is assumed to have a half-filled band, i.e., the number of particles is assumed to equal the number of vectors  $\mathbf{R}_i$ .<sup>12</sup> Then the function  $q_{\sigma}$  has the form<sup>13,15</sup>

$$q = q_{\sigma} = q_{-\sigma} = \frac{4d}{1-m^2} [1 - 2d + \sqrt{(1-2d)^2 - m^2}], \quad (3)$$

where  $m = \langle n_{i\sigma} - n_{i-\sigma} \rangle$  is the magnetic moment, which is also normalized to a single vector  $\mathbf{R}_i$ .

In Ref. 15 a thermodynamic method was used to pass to nonzero temperatures and arbitrary magnetic fields; however, it unfortunately does not permit accurate determination of the entropy of an almost-localized Fermi liquid. Therefore, to calculate the thermodynamic parameters, we use the one-electron spectrum of an almost-localized Fermi liquid:<sup>16,17</sup>

$$E_{\mathbf{k}, \sigma} = q_{\sigma} \varepsilon_{\mathbf{k}, \sigma}. \quad (4)$$

The spectrum (4) makes it possible to accurately calculate the thermodynamic potential and the free energy of an almost-localized Fermi liquid in an arbitrary magnetic field. We pass to the dimensionless variables<sup>15</sup>

$$\xi = \frac{2\varepsilon}{\Delta}, \quad t = \frac{2k_B T}{\Delta}, \quad h = \frac{2\mu_0 H}{\Delta}, \quad f = \frac{2F}{\Delta},$$

$$\omega = \frac{2\Omega}{\Delta}, \quad \nu = \frac{2\mu}{\Delta}, \quad u = \frac{2U}{\Delta}, \quad (5)$$

where  $\Delta$  is the width of the original electron band,  $k_B$  is the Boltzmann constant,  $\mu_0$  is the Bohr magneton,  $T$  is the temperature,  $H$  is the magnetic field,  $\mu = \mu_\sigma = -\mu_{-\sigma}$  is the chemical potential of uncorrelated fermions (in the original band), and  $F$  and  $\Omega$  are, respectively, the free energy and the thermodynamic potential of an almost-localized Fermi liquid. As is usually done,<sup>13,15</sup> we use the Hubbard model band

$$\rho(\xi) = \begin{cases} \frac{2}{\pi} \sqrt{1-\xi^2}, & |\xi| \leq 1, \\ 0, & |\xi| \geq 1. \end{cases} \quad (6)$$

The effective band (4) for  $^3\text{He}$  has a width of about 1 K, which is much greater than the typical temperatures in melting experiments ( $<0.2$  K); therefore, we shall henceforth investigate only low temperatures  $t \ll q$ . As shown in Ref. 15, there are two types of solutions, depending on the value of the magnetic moment.

1. We perform operations similar to those in Ref. 15. Then from the spectrum (4), retaining only the term quadratic in  $t/q$ , for the moderate magnetic moments  $1-m > t/q$  we obtain

$$\omega = -q \left[ \frac{4\sqrt{(1-\nu^2)^3}}{3\pi} + m(0)\nu + \frac{2}{3} \pi \left( \frac{t}{q} \right)^2 \sqrt{1-\nu^2} \right] + ud, \quad (7)$$

where

$$m(0) = \frac{2}{\pi} \left[ \nu \sqrt{1-\nu^2} + \arcsin \nu \right]$$

is the magnetic moment at zero temperature. The total energy, the entropy, and, thus, the free energy of an almost-localized Fermi liquid can be calculated directly from the one-electron spectrum (4):

$$f = -q \left[ \frac{4\sqrt{(1-\nu^2)^3}}{3\pi} + \frac{2}{3} \pi \left( \frac{t}{q} \right)^2 \frac{1}{\sqrt{1-\nu^2}} \right] + ud. \quad (8)$$

2. A "semiconductor" solution,<sup>15</sup> which corresponds to a ferromagnetic insulator, at  $T=0$  K, is possible when  $\nu > 1$ . Retaining the first nonvanishing term in  $t/q$ , we obtain the thermodynamic potential and free energy in the form

$$\omega = -q \left[ \nu + 2 \sqrt{\frac{2}{\pi} \left( \frac{t}{q} \right)^3} \exp\left( q \frac{1-\nu}{t} \right) \right] + ud, \quad (9)$$

$$f = -2q \sqrt{\frac{2}{\pi} \left( \frac{t}{q} \right)^3} \exp\left( q \frac{1-\nu}{t} \right) \nu + ud. \quad (10)$$

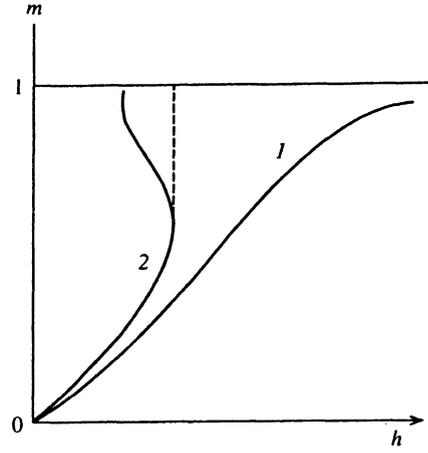


FIG. 1. General form of the solutions obtained from Eqs. (12) and (13). 1) small values of  $u$ ; 2) large values of  $u$ .

There is still a narrow range  $1-t/q < m < 1$ , in which there is no simple analytic solution. This is due to a purely technical difficulty: the derivative  $d\rho/d\xi$  in the Hubbard band diverges at  $\xi=1$ , and it is consequently impossible to use an expansion in powers of  $t/q$  when  $\nu$  is near that point.

It is convenient to express the free energy and the thermodynamic potential of an almost-localized Fermi liquid in terms of the corresponding potentials  $\omega_0$  and  $f_0$  of uncorrelated fermions in the form

$$\omega = q\omega_0 + ud, \quad f_0 = qf_0 + ud.$$

We note that the magnetic moment of an almost-localized Fermi liquid equals the magnetic moment of the uncorrelated fermions; therefore, it can be found from the following expression:

$$m = - \frac{\partial \omega_0}{\partial \nu}. \quad (11)$$

Then the state of an almost-localized Fermi liquid is determined by minimizing the free energy with respect to the parameter  $d$  (Ref. 15):

$$\frac{\partial f}{\partial d} = \frac{\partial q}{\partial d} f_0 + u = 0. \quad (12)$$

This equation relates the independent variables  $d$  and  $q$ . Finally, the chemical potential of an almost-localized Fermi liquid is the derivative of the free energy with respect to the magnetic moment along the line in the  $md$  plane defined by Eq. (12),

$$h = \frac{\partial f}{\partial m} = q \frac{\partial f_0}{\partial m} + \left( \frac{\partial q}{\partial m} + \frac{\partial q}{\partial d} \frac{\partial d}{\partial m} \right) \times f_0 + u \frac{\partial d}{\partial m} = q\nu + \frac{\partial q}{\partial m} f_0. \quad (13)$$

The form of the functions  $m(h)$  obtained by solving Eqs. (12) and (13) depends on  $u$  (see Fig. 1). When  $u$  is small (curve 1), this function increases monotonically, and the "semiconductor" solution appears in the limit  $h \rightarrow \infty, m \rightarrow 1$ . When  $u$  is large (curve 2), as in the case of  $^3\text{He}$ , the function

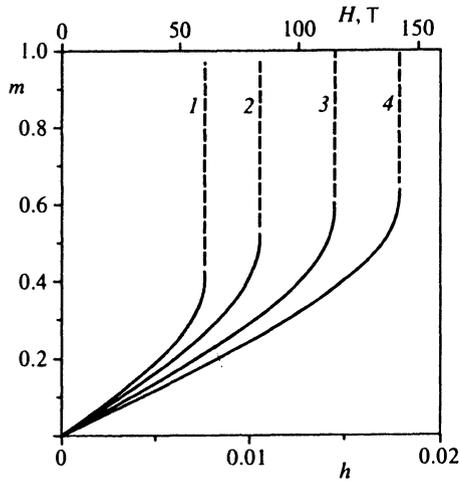


FIG. 2. Plots of  $m(h)$  for various values of the reduced temperature  $t$ : 1)  $t=0$ ; 2)  $t=0.01$ ; 3)  $t=0.02$ ; 4)  $t=0.03$ .

becomes two-valued, and a first-order transition to the state of a ferromagnetic insulator takes place.<sup>13</sup> A known thermodynamic inequality<sup>18</sup> gives the condition for the transition

$$\frac{\partial h}{\partial m} = 0. \quad (14)$$

We take the parameters of the Hamiltonian (1) for  $^3\text{He}$  according to the data in Ref. 19. As usual, we neglect the magnetostriction. Typical pressures in the  $^3\text{He}$  melting experiments were 30 bar. In that case the Fermi energy of the uncorrelated fermions is 6.23 K, and the interaction constant

$$u = \frac{32}{3\pi} \sqrt{1 - \frac{1}{m^*}} = 3.1,$$

where  $m^*$  is the effective mass of a fermion in units of the mass of a free fermion. Plots of  $m(h)$  for various temperatures are shown in Fig. 2.

### 3. MODEL OF $^3\text{He}$ MELTING IN A MAGNETIC FIELD

The liquid and solid phases of  $^3\text{He}$  have rather long magnetic moment relaxation times at low temperatures. At temperatures below 0.1 K they can reach 1000 s (Refs. 14 and 15). Therefore, the melting rate in the experiments was chosen such that, on the one hand, equilibrium values of the temperature and pressure could be established and, on the other hand, the relaxation of the magnetic moment was insignificant.<sup>2,4</sup> The spin diffusion depth in the solid phase is only about  $1 \mu\text{m}$  (Ref. 7), and it is usually assumed that there are three phases: a liquid phase, the surface layer of the solid phase, and the bulk of the solid phase. The liquid phase is in equilibrium with the surface layer of the solid phase, i.e., their chemical potentials are equal. At temperatures of the order of 0.1 K the solid phase is a paramagnet, and for a magnetic moment of the surface layer  $m_s$ , the effective magnetic field equals the chemical potential of the liquid:  $h = t \arctan m_{ss}$  (Ref. 4). The chemical potential of the bulk solid is assumed to be independent. Therefore, it may be assumed that its magnetic moment is maintained at low temperatures.

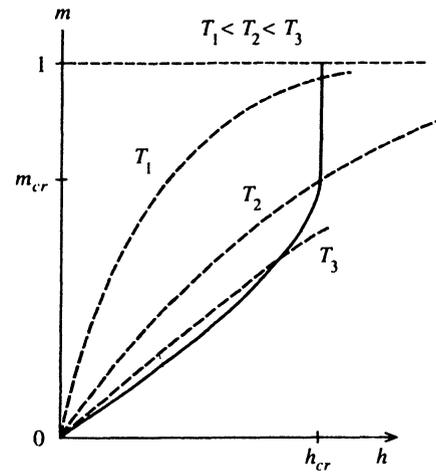


FIG. 3. States of the almost-localized Fermi liquid (solid line) and the paramagnetic solid phase at various temperatures (dashed lines).

It was shown in Ref. 8 that the radius of curvature of the phase boundary is greater than the spin diffusion depth and that in this case the magnetic moment of the liquid equals the magnetic moment  $m_s$  of the bulk solid. However, this condition holds only when the initial magnetic moment is small, and thus the magnetic moment of an almost-localized Fermi liquid is bounded by the critical value  $m_{cr}$ . Therefore, when  $m_s > m_{cr}$ , the magnetic moment of the liquid  $m_l$  equals  $m_{cr}$ . Plots of  $m(h)$  for a paramagnetic solid at various temperatures (dashed lines) and the line of state of the liquid (solid line) are schematically depicted in Fig. 3. When the temperature is low and the initial magnetic moment of the solid phase  $m_0 > m_{cr}$ , equilibrium is established at the critical value of the chemical potential of the almost-localized Fermi liquid  $h_{cr}$ .

If melting occurs at constant temperature, as in Ref. 2, the curves for both phases are fixed and only  $h$  varies. At high temperatures the magnetic moment relaxation rate increases sharply,<sup>4</sup> and the magnetic moment can relax even more rapidly in the solid phase than in the liquid.<sup>4,6</sup> Under these conditions the magnetic moment of the solid phase will be considerably lower than the initial magnetic moment, and there will be some uncertainty in the value of the magnetic moment of the liquid phase. Unfortunately, the magnetic moment was not measured in Ref. 4; therefore, as we analyze its results, we assume that the condition  $m_{ss} = m_l$  holds at high temperatures. The displacement of the line of state of the liquid as a function of the temperature is not shown in Fig. 3.

The equality between the chemical potentials of the liquid and the surface layer leads to a relationship between the pressure  $P$  and the magnetic field  $H$  (Ref. 3) in the form

$$\frac{\partial P}{\partial H} = \frac{\delta M}{\delta V},$$

where  $\delta M$  is the difference between the specific magnetic moments of the solid and liquid phases and  $\delta V$  is the difference between the molar volumes of these two phases. Integration of this equation leads to an expression for the pressure shift<sup>4</sup>

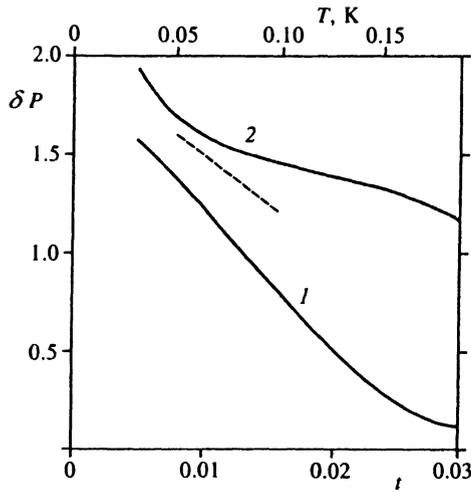


FIG. 4. Pressure depression for rapid melting: 1—experimental value from Ref. 4 for melting over the course of 2 min; dashed line—most rapid melting from Ref. 4; 2—calculated curve without consideration of the relaxation of the magnetic moment of the bulk solid.

$$\delta P = \frac{N_A \Delta}{2 \delta V} \left[ t \ln \cosh \left( \frac{h}{t} \right) - \int_0^h m_L dh' \right], \quad (15)$$

where  $N_A$  is Avogadro's number. Thus, the pressure shift is proportional to the area of the figure bounded by the dashed and solid lines in Fig. 3.

#### 4. RESULTS AND DISCUSSION

When  $m$  and  $t$  are small parameters, the melting pressure shift can easily be evaluated. In (13) we leave only linear terms in the small parameter. Then we have

$$h = \left( \frac{\pi}{4} - \frac{4}{3\pi} \frac{1+2I}{1+I} \right) q(0,0)m, \quad (16)$$

where  $I = u/8f_0(0,0)$ , and  $f_0(0,0)$  and  $q(0,0)$  are the values  $f_0$  and  $q$  when  $m=0$  and  $q=0$ . We represent the first term in the square brackets in (15) in the form  $h^2/32t$ . Then it turns out that  $\delta P \propto m^2$ , and when  $u=3.1$  and  $t=0.016$  ( $T=0.1$  K), we obtain  $\delta P/m^2=4$  bar. This estimate is close to the value obtained in Ref. 2, where  ${}^3\text{He}$  melted at essentially constant temperature ( $T \approx 0$ ).1 K for  $m_L < 0.3$ ). The pressure shift and the magnetic moment were monitored simultaneously. At the end of the melting process, i.e., at small  $m_L$ , it may be assumed that the fraction of the solid phase is small and that the measured magnetic moment equals  $m_L$ . The value  $\delta P/m^2 = 5.5 \pm 1.0$  bar was obtained experimentally. We also note that the value obtained in the thermodynamic model used in Ref. 2 is greatly underestimated (1.9 bar).

The pressure shift for a large initial magnetic moment at various temperatures was investigated in Ref. 4. The results for melting over the course of 2 min are depicted by solid curve 1 in Fig. 4. The dashed curve shows the pressure depression for the most rapid melting. The difference between these lines gives an estimate of the measurement error. Solid curve 2 in this figure shows the pressure obtained by numerically solving (7), (13), and (14). Good correspondence between the theoretical and experimental curves is observed at

low temperatures. Strong divergence appears at higher temperatures due to the uncertainty in the melting regime mentioned above. This divergence shows that our artificial condition  $m_{ss} = m_L$  does not hold. It may also be noted that this condition places an upper limit on the pressure shift. If the real value of  $h$  is lower, the area bounded by the lines of the surface and liquid phases decreases. Otherwise, an additional area appears in the expression (15) with a minus sign. Thus, a correct analysis of the experimental data at high temperatures requires simultaneous measurements of the pressure shift and the magnetic moment of the liquid phase.

In Ref. 5 rapid melting of the solid phase with a very high value of the initial magnetic moment (80%) produced strongly polarized liquid  ${}^3\text{He}$ , which subsequently relaxed at an almost constant temperature ( $\approx 0.08$  K). The magnetic moment of the liquid  $M$  and the heat  $Q$  evolved during relaxation were measured. The effective magnetic field  $B_{\text{eff}}$  was then calculated from the expression

$$\dot{Q} = -\mu_n (B_{\text{eff}} - B_a) \dot{M},$$

where  $\mu_n$  is the nuclear magnetic moment of  ${}^3\text{He}$  and  $B_a$  is the applied magnetic field. This gave the dependence of the magnetic moment on  $B_{\text{eff}} - B_a$ , which was nearly linear. This dependence was taken in Ref. 5 as evidence that the no first-order transition occurs at magnetic moments up to  $m=0.6$ , since if such a transition had occurred, the liquid would have separated into two phases and the plot of  $B(m)$  would have contained a plateau. These arguments, however, totally ignore the latent heat of the transition, and under the calculation method used this heat results in overestimation of the values of  $B(m)$ . The overestimation of  $B(m)$  in Ref. 5 is also supported by the following fact. If the melting pressure shift is calculated from a plot of the dependence of  $B_{\text{eff}} - B_a$  on  $m$  (Ref. 5) (since  $m$  is not bounded here by a critical value, the magnetic moment of the liquid is equal to the initial magnetic moment of the solid phase<sup>8</sup>), we obtain  $\delta P \approx 3$  bar. However,  $\delta P \approx 1$  bar was obtained experimentally for the same conditions.<sup>4</sup>

Comparing the almost-localized Fermi liquid theory with the other theories, we should note the following. Under the present theory the localized state for  ${}^3\text{He}$  is clearly not physically definable in the limit  $m \rightarrow 1$ , and the induced interaction and nearly metamagnetic liquid theories are preferable here. On the other hand, they are mean-field theories; therefore, one would scarcely expect that they will correctly describe the strong short-range interaction at moderate values of  $m$ . Thus, the question of the occurrence of a first-order transition in the strongly magnetized state remains open. An answer to it can be obtained from rapid  ${}^3\text{He}$  melting experiments. We note that the initial magnetic moment in Ref. 4 was appreciably greater than the critical value at low temperatures. Under such circumstances a further increase in the initial magnetic moment does not cause variation of the pressure shift in the almost-localized Fermi liquid theory.

#### 5. CONCLUSIONS

In the present work we generalized the almost-localized Fermi liquid model to arbitrary magnetic fields and low tem-

peratures. It is shown that the critical value of the magnetic moment, at which a first-order transition occurs in an almost-localized Fermi liquid, is strongly dependent on the temperature.

The almost-localized Fermi liquid model is applicable to the analysis of the experimental results obtained in Refs. 2 and 4. When the magnetic moment and the temperature are low, the pressure shift for rapid  $^3\text{He}$  melting is quadratically dependent on  $m$ . The theoretical value of the ratio  $\delta P/m^2$  corresponds satisfactorily with the experimental value. There is good agreement between the almost-localized Fermi liquid model and experiment at large initial magnetic moments and low temperatures. Strong divergence appears at high temperatures, and it is attributed to uncertainties in the experimental data<sup>4</sup> (the magnetic moment of the liquid phase is unknown).

The possibility of experimentally determining the occurrence (or nonoccurrence) of a first-order transition in  $^3\text{He}$  in a strong magnetic field has been demonstrated. If a transition occurs, saturation appears in the dependence of the pressure shift on the initial magnetic moment at large initial magnetic moments and low temperatures.

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