Near-Metamagnetism of Liquid ^{3}He at High Pressure

Kevin S. Bedell[(a)]
Kamerlingh Onnes Laboratorium der Rijksuniversiteit, 2300 RA Leiden, The Netherlands

and

Carlos Sanchez-Castro
Physics Department, State University of New York at Stony Brook, Stony Brook, New York 11794
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A new magnetic equation of state for liquid ^{3}He is derived. This is obtained from a model for the polarization dependence of the Landau parameters. The parameters of the model are constrained by general thermodynamic relationships and microscopic symmetries. It is shown that the nonlinear field splitting of the A phase in liquid ^{3}He is consistent with this model. We propose that liquid ^{3}He can be viewed as nearly metamagnetic.

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In a number of recent experiments[1-3] it has been possible to produce samples of nuclear-spin–polarized liquid ^{3}He with polarizations of 50% or larger lasting for as long as several minutes. The large depressions in the melting curve observed in these experiments depend in detail on the magnetic equation of state, MEOS, of the polarized liquid.[4] The behavior of liquid ^{3}He as a function of polarization is not known experimentally. Some attempts have been made to determine the properties of polarized ^{3}He by either simple extrapolations from the unpolarized phase[4] or by introduction of model calculations.[5-7]

The simple extrapolation used by Castaing and Nozières[4] gives reasonable estimates for some quantities but it does not provide a detailed picture of the magnetic equation of state. The two previous attempts to calculate the MEOS of ^{3}He were the paramagnon model calculations of Béal-Monod and Daniel[5] and the extension of the Gutzwiller model by Vollhardt.[6] The predictions of these two models at $T=0$ are very different. While the paramagnon model[5] predicts that both the susceptibility and the total density of states decreases with increasing field, the Gutzwiller model predicts that both quantities increase. It is well known that the total density of states decreases with increasing field for liquid ^{3}He; however, the trend in the susceptibility is not known experimentally.

In this Letter we present a model for polarized ^{3}He with a behavior that is quite different from the other models. We predict that while the total density of states decreases, the susceptibility at constant density, χ_n, where n is the density, will initially increase with polarization reaching a maximum value then dropping to zero in the fully polarized phase. From the susceptibility we can obtain the dependence of the energy density, $\mathcal{E}$, on the magnetization density, $\mathcal{M}$. The initial increase in χ_n gives rise to a negative $\mathcal{M}^4$ term in $\mathcal{E}$; the $\mathcal{M}^2$ term and $\mathcal{M}^6$ terms are positive. If the $\mathcal{M}^4$ term is attractive enough it would give rise to a field-induced ferromagnetic transition. A field-induced transition from an itinerant paramagnetic phase to a ferromagnetic one is what Wohlfarth and Rhodes[8] described as a metamagnetic transition. In liquid ^{3}He this term does not appear to be attractive enough; however, it allows for a *new* interpretation of the magnetic properties of liquid ^{3}He, i.e., ^{3}He is nearly metamagnetic. This is in contrast to the usual pictures of ^{3}He as being nearly ferromagnetic[5] or nearly localized.[6] As we will argue, currently available measurements[9,10] on the nonlinear field splitting of the A phase in liquid ^{3}He tend to support our picture.

We begin at the level of the Landau theory for a polarized Fermi liquid. The energy density is given by[11]

$$\mathcal{E}=\mathcal{E}_0+\sum_{\mathbf{p}\sigma}\epsilon^0_{\mathbf{p}\sigma}\delta n_{\mathbf{p}\sigma}+\frac{1}{2}\sum_{p\sigma,p'\sigma'}\tilde{f}^{\sigma\sigma'}_{\mathbf{pp}'}\delta n_{\mathbf{p}\sigma}\delta n_{\mathbf{p}'\sigma'}, \quad (1)$$

where $\delta n_{\mathbf{p}\sigma}=n_{\mathbf{p}\sigma}-n^0_{\mathbf{p}\sigma}$ and $n^0_{\mathbf{p}\sigma}$ is the equilibrium distribution function in the presence of the field $\mathbf{B}_0$. The quasiparticle interaction $\tilde{f}^{\sigma\sigma'}_{\mathbf{pp}'}$ has three distinct components for the spin combinations $\uparrow\uparrow$, $\uparrow\downarrow$, and $\downarrow\downarrow$ ($\neq\uparrow\uparrow$). For our discussion we can ignore the transverse component of the interaction. We can expand $\tilde{f}^{\sigma\sigma'}_{\mathbf{pp}'}$ in the usual way,[12] $\tilde{f}^{\sigma\sigma'}_{\mathbf{pp}'}=\sum_l \tilde{f}^{\sigma\sigma'}_l P_l(\hat{\mathbf{p}}\cdot\hat{\mathbf{p}}')$ where the $\tilde{f}^{\sigma\sigma'}_l$ are functions of the polarization $\Delta=\mathcal{M}/n$ and the density. The Landau parameters $\tilde{f}^{\sigma\sigma'}_1$ are related to the effective masses m^*_σ by Galilean invariance[7]

$$m^*_\sigma/\mathcal{M}=1+\tfrac{1}{3}N_\sigma(0)[\tilde{f}^{\sigma\sigma}_1+(k_F^{-\sigma}/k_F^{\sigma})^2\tilde{f}^{\uparrow\downarrow}_1],$$

where m^*_σ enters into the density of states, $N_\sigma(0)=k_F^\sigma m^*_\sigma/2\pi^2$, and the Fermi momentum is given by $k_F^\sigma=k_F(1+\sigma\Delta)^{1/3}$, $\sigma=+1$ ($\uparrow$), -1 ($\downarrow$).

The model we have constructed consists of an expansion of the Landau parameters in the polarization. In principle, these expansions can contain any number of terms; however, we truncate the expansions at a finite order. The physical arguments are as follows: For the $l=0$ moments we have

$$\tilde{f}_0^{\sigma\sigma} = f_0^{\uparrow\uparrow}(1 - b_0\sigma\Delta + b_1\Delta^2), \tag{2a}$$

$$\tilde{f}_0^{\uparrow\downarrow} = f_0^{\uparrow\downarrow}(1 + c_1\Delta^2). \tag{2b}$$

The moment $\tilde{f}_0^{\uparrow\downarrow}$ arises from the screened short-range potential and the exchange of density fluctuations, whereas $\tilde{f}_0^{\sigma\sigma}$ comes mostly from the exchange of density fluctuations.[7,13] In a polarization-potential picture[14] these correspond to core energies of the order of 10–100 K, depending on pressure. Since the magnetic field energies are of the order of 1 K or less we expect only small changes in $\tilde{f}_0^{\sigma\sigma'}$. Thus, the expansion to order Δ^2 should be reasonable at all Δ for the $l=0$ moments.

For the $l=1$ moments the situation is more complicated. In projecting out an $l=1$ moment from the quasiparticle interaction the longer-range parts of the interaction will contribute, e.g., spin fluctuations.[7,13] These of course are low-energy excitations and they will be more sensitive to the magnetic field energies necessary to produce highly polarized ^{3}He. Moreover, we know from general arguments that $\tilde{f}_1^{\uparrow\downarrow}$ and $\tilde{f}_1^{\downarrow\downarrow}$ will vanish[15] and that $\tilde{f}_1^{\uparrow\uparrow}$ is negative when $\Delta=1$.[7] To account for the more detailed structure anticipated in $\tilde{f}_1^{\sigma\sigma'}$ we include terms to fourth order in Δ, where

$$\tilde{f}_1^{\sigma\sigma} = f_1^{\uparrow\uparrow}(1 - d_0\sigma\Delta + d_1\Delta^2 - d_2\sigma\Delta^3 + d_3\Delta^4), \tag{3a}$$

and

$$\tilde{f}_1^{\uparrow\downarrow} = f_1^{\uparrow\downarrow}(1 + g_1\Delta^2 + g_3\Delta^4). \tag{3b}$$

In Eqs. (2a) and (2b) and (3a) and (3b) the moments $f_l^{\sigma\sigma'}$ refer to the values of the unpolarized Landau parameters.

The only other physical assumption we make is that the moments $\tilde{f}_l^{\sigma\sigma'}$ for $l \geqslant 2$ are zero. This is consistent with the short-range nature of the quasiparticle interaction as suggested by the induced-interaction model[13] or the polarization-potential approach.[14] Additionally we will see that this gives good agreement with a number of experiments.

To determine the parameters we make use of thermodynamic and microscopic constraints. It has been shown[11] that magnetostriction and the forward scattering sum rules, $\sum_l \tilde{A}_l^{\sigma\sigma} = 0$, where

$$\tilde{A}_l^{\sigma\sigma} = (\tilde{f}_l^{\sigma\sigma} C_l^{-\sigma-\sigma} - \tilde{f}_l^{\uparrow\downarrow 2}/2l+1)[N_\sigma(0)/D_l]$$

with

$$C_l^{\sigma\sigma} = N_\sigma^{-1}(0) + \tilde{f}_l^{\sigma\sigma}/(2l+1)$$

and

$$D_l = N_\uparrow(0)N_\downarrow(0)[C_l^{\uparrow\uparrow}C_l^{\downarrow\downarrow} - (\tilde{f}_l^{\uparrow\downarrow}/2l+1)^2],$$

determine the linear coefficients. To second order in Δ we have five parameters if we include the expansion of m_σ^*, $m_\sigma^* = m^*(1 - a_0\sigma\Delta + a_1\Delta^2 + \dots)$. The sum rule and Galilean invariance determine two of the parameters. The compressibility at constant magnetization, κ_M^{-1}, with M the magnetization, and the temperature dependence of the zero-field susceptibility,[16] $\chi(T) = \chi(0)(1 - \alpha T^2)$ where $\chi(0)$ is the $T=0$, $B=0$ susceptibility, gives two additional parameters.

To see this we note that[17]

$$\kappa_M^{-1} = n(\partial P/\partial n)_{\mathcal{M}} + \mathcal{M}(\partial P/\partial \mathcal{M})_n$$

$$\simeq \kappa^{-1} + \frac{\mathcal{M}^2}{3n}\frac{\partial}{\partial n}\left(n^2 \frac{\partial T_{\rm SF}}{\partial n}\right), \tag{4}$$

where $\kappa^{-1} = \frac{2}{3} n \epsilon_{\rm F}(1 + F_0^s)$ and $T_{\rm SF} = (1 + F_0^a)\epsilon_{\rm F}$. Here F_0^s, F_0^a, and $\epsilon_{\rm F} = k_{\rm F}^2/2m^*$ are the Landau parameters and Fermi temperature of the unpolarized system. To order $\mathcal{M}^2$, Eq. (4) is exact and from this we can detemine the change in the sound velocity, $\Delta c_M = (c_M - c_1)/c_1$, which at $P=34.36$ bars yields a value of 3.5×10^{-6} for $B=10$ T. If we begin with Eq. (1) we see that the term of order $\mathcal{M}^2$ in Eq. (4) is

$$(\tfrac{2}{3})n\epsilon_{\rm F}[\tfrac{5}{9} + F_0^a + a_0^2 + \tfrac{5}{3}a_0 - F_0^{\uparrow\uparrow}b_0 - a_1 + \tfrac{1}{2}(F_0^{\uparrow\uparrow}b_1 + F_0^{\uparrow\downarrow}c_1)].$$

For the specific heat we have

$$C_v/T = (\pi^2/3)[N_\uparrow(0) + N_\downarrow(0)]$$

$$\simeq (\pi^2/3)N(0)[1 + (a_1 - a_0/3 - \tfrac{1}{9})\delta^2]. \tag{5}$$

From the Maxwell relation $\partial c_v/\partial B = T\partial^2\mathcal{M}/\partial T^2$ we have that

$$a_1 - a_0/3 - \tfrac{1}{9} = -\tfrac{4}{3}(\alpha/\pi^2)(1 + F_0^a)\epsilon_{\rm F}^2.$$

The four remaining parameters are constrained by the sum rules and the vanishing of $\tilde{f}_1^{\uparrow\downarrow}$ and $\tilde{f}_1^{\downarrow\downarrow}$ for $\Delta=1$. In this limit ($\Delta=1$) the sum rule $\sum_{l=0}^{1}\tilde{A}_l^{\downarrow\downarrow}=0$ with $\tilde{f}_1^{\uparrow\downarrow}$ and $\tilde{f}_1^{\downarrow\downarrow}$ equal to zero gives the additional constraint[15]

$$\tilde{f}_0^{\downarrow\downarrow} = \tilde{f}_0^{\uparrow\downarrow 2}/[N_\uparrow^{-1}(0) + \tilde{f}_0^{\uparrow\uparrow}]. \tag{6}$$

We note that Eq. (6) gives a relationship between the effective mass $m_\uparrow^*$ and the parameters $\tilde{f}_0^{\sigma\sigma'}$.[15]

With all of the parameters of the theory determined we can calculate several other properties of the system. The most significant result is for the MEOS, which we express as the polarization dependence of χ_n. From Eq. (1) we obtain $\chi_n^{-1} = \partial^2 E/\partial\mathcal{M}^2$, where [11]

$$\chi_n^{-1} = \tfrac{1}{4}[N_\uparrow^{-1}(0) + N_\downarrow^{-1}(0) + \tilde{f}_0^{\uparrow\uparrow} + \tilde{f}_0^{\downarrow\downarrow} - 2\tilde{f}_0^{\uparrow\downarrow}]. \tag{7}$$

To obtain the results shown in Fig. 1 we have used the expansions Eqs. (2a) and (2b) and (3a) and (3b) in the exact expression, Eq. (7), for χ_n. For the m_σ^*'s

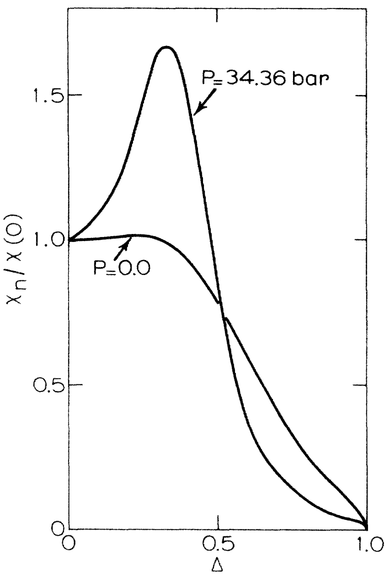

FIG. 1. The ratio of the susceptibility to the zero-field susceptibility at two pressures.

that appear in χ_n we used Eqs. (3a) and (3b) in the Galilean-invariant expression for the effective mass. It is useful to study the leading correction, of order Δ^2, in χ_n, where $\chi_n \simeq \chi(0)(1+\beta\Delta^2)$. The coefficient β is given by

$$(1+F_0^a)\beta = -[\tfrac{2}{9} + a_0(a_0 - \tfrac{1}{3}) - a_1 + \tfrac{1}{2}(F_0^{\uparrow\uparrow} b_1 - F_0^{\uparrow\downarrow} c_1)].$$

What is most significant here is that we find $\beta > 0$. Clearly, if ^{3}He remains paramagnetic up to $\Delta = 1$ and $\beta > 0$ then χ_n must have a maximum since $\chi_n \rightarrow 0$ as $\Delta \rightarrow 1$. The coefficient β could be obtained by measurement of χ_n at high pressure. At the melting pressure there would be a 5% increase in a 20-T field and only an 0.1% increase at saturated vapor pressure.

A direct measurement of β would clearly distinguish between our nearly metamagnetic model and the other models.[5,6] However, recent measurements of the nonlinear field splitting of the A phase of ^{3}He may already give us the information.[9] The transition temperature $T_c^{A_1}$ is given by[7]

$$(T_c^{A_1}/T_c) = (\epsilon_F^{\uparrow}/\epsilon_F)\exp(1/\tilde{g}_1^{\uparrow\uparrow} - 1/g_1^{\uparrow\uparrow}), \quad (8)$$

where $\tilde{g}_1^{\uparrow\uparrow} = \frac{1}{3}N_{\uparrow}(0)\tilde{a}_0^{\uparrow\uparrow}$ and $g_1^{\uparrow\uparrow} = \frac{1}{6}(A_0^s + A_0^a)$ with $A_0^{s,a} = F_0^{s,a}/(1+F_0^{s,a})$. If we expand this in the field we have $(T_c^{A_1} - T_c)/B \simeq t_{c1} + t_{c2}B$, where the coefficients are given in Table I. The most significant feature of this result is that $t_{c2} > 0$. This is determined largely by the parameter combination $F_0^{\uparrow\uparrow}b_1 - F_0^{\uparrow\downarrow}c_1$ which also is the largest contributor to β. At the higher pressures if $\beta > 0$ then $t_{c2} > 0$, and for $\beta < 0$ we have $t_{c2} < 0$.

The experimental measurement of the nonlinear field splitting is only known for $T_c^{A_1} - T_c^{A_2}$.[10] At $P = 29$ bars Osheroff[9] finds the nonlinear term in $T_c^{A_1} - T_c^{A_2}$ to be approximately 0.6 μK/T^2 while our calculated t_{c2} is 1.8 μK/T^2. That our nonlinear term in $T_c^{A_1} - T_c$ is larger than the $A_1 - A_2$ splitting is as it should be. The reason is that the terms quadratic in B must come with the same sign in $T_c^{A_1}$ and $T_c^{A_2}$. For example, in the weak-coupling limit the quadratic term in $T_c^{A_1} - T_c^{A_2} = T_c^{\uparrow} - T_c^{\downarrow}$ which is odd in B (or Δ). The strong-coupling corrections of course change this. Without a measurement or calculation of the strong-coupling corrections it is difficult to determine how they will affect $T_c^{A_2}$. However, we believe the following arguments are most plausible: A decrease of the spin fluctuations, i.e., a decrease in χ_n, would reduce the strong-coupling corrections. This will drive the system closer to the weak-coupling limit, i.e., $T_c^{A_2} \rightarrow T_c^{\downarrow}$. In this case the Δ^2 term in $T_c^{A_2}$ would be more negative than in $T_c^{A_1}$ giving a positive Δ^2 term in $T_c^{A_1} - T_c^{A_2}$. On the other hand, increasing the spin fluctuations with increasing Δ would increase the strong-coupling corrections. This would tend to push $T_c^{A_2}$ away from $T_c^{\downarrow}$ making the Δ^2 term, which is positive, smaller in $T_c^{A_2}$ than in $T_c^{A_1}$. This would also give a positive coefficient to the Δ^2 term in $T_c^{A_1} - T_c^{A_2}$. We believe that the latter possibility holds in liquid ^{3}He. This allows us to make an estimate of the strong-coupling corrections by the assumption that the corrections to the Δ^2 term are the same as the linear term; this will probably underestimate the effect. For example, in linear order at 29 bars,[10] $T_c^{A_1} - T_c = 1.7(T_c - T_c^{A_2})$ and by use of this for the quadratic order we would get a value of 0.74 μK/T^2 for the B^2 ($B = M/\chi$) coefficient in $T_c^{A_1} - T_c^{A_2}$, which is in good agreement with experiment.[9] This of course is only a crude estimate. What is needed is a direct mea-

TABLE I. Some of the predictions of the model. The parameters are defined in the text. The experimental values for t_{c1} are from Ref. 10.

P (bars)	β	t_{c1} (μK/T) Theor.	Expt.	t_{c2} (μK/T^2)	$c_M^{\downarrow}$ (m/s)	$m_{\uparrow}^*/m$
0.0	0.57	6.66	8.0	0.075	202.2	0.84
34.36	7.16	28.0	39.0	2.3	438.6	0.77

surement of $T_c^{A_1} - T_c$ to make a comparison with our theory.

The key result of this paper is that the possibility χ_n initially increases with increasing field, i.e., $\beta > 0$. This result is not very sensitive to the approximations we have used. The largest contributor to β is the $l=0$ parameters in the combination $F_0^{\uparrow\uparrow} b_1 - F_0^{\uparrow\downarrow} c_1$. At the melting pressure this is five times larger than the $l=1$ contribution coming from the density of states. Setting $\tilde{f}_l^{\sigma\sigma'} = 0$ for $l \geqslant 2$ would have very little effect on this, unless there were an unusually strong polarization dependence in these parameters. The expansions to order Δ^2 in the $\tilde{f}_0^{\sigma\sigma'}$'s are based on sound physical arguments. On the other hand, the expansions for the $\tilde{f}_1^{\sigma\sigma'}$'s were chosen for their simplicity since there are no simple physical arguments for these expansions. Fortunately, this will have *no* effect on the value of β. The reason is that the two sum rules in the fully polarized phase determine the values of $m_\uparrow^*/m$ and $F_0^{\uparrow\uparrow} b_1 - F_0^{\uparrow\downarrow} c_1$. Thus, the parametrization of the $\tilde{f}_1^{\sigma\sigma'}$'s will only determine how $m_\uparrow^*/m$ reaches its fully polarized value, the final value being fixed by the sum rules. Of course if a metamagnetic transition did occur we would expect that the two main assumptions, i.e., truncation at $l=1$ and retention of only terms of order Δ^2 in $\tilde{f}_0^{\sigma\sigma'}$, break down. However, since this does not seem to be the case we believe that these assumptions are good. Finally, the good agreement between our theoretical predictions for the field dependence of $T_c^{A_1}$ and the experiments[9,10] is further confirmation of the soundness of our approximations, since any errors in the field dependence would be exponentially magnified in the $T_a^{A_1}$ equation.

We can summarize our main results as follows.

(1) We have obtained a new MEOS for liquid ^{3}He, i.e., ^{3}He is nearly metamagnetic. An increasing χ_n as a function of Δ was also predicted by Vollhardt.[6] This, however, resulted from the increase of the total density of states in his calculation, which is the opposite of the experimental behavior (see Quader and Bedell[7]). Thus, the origin of the metamagnetic behavior seen by Vollhardt is quite different from what we have found.

(2) This model gets good results for the linear field splitting (see Table I) and properly accounts for the magnetostriction. We have also made the predictions for the sign and magnitude of the nonlinear field splitting of the A_1 phase of ^{3}He.

(3) We can also make some qualitative predictions regarding the transport coefficients: The transport properties at $T \ll \epsilon_F$ are dominated by the spin fluctuations. We would expect an initial decrease with increasing Δ in the transport coefficients since the spin-fluctuation scattering is initially increasing. There should be a minimum (near the maximum in χ_n), for example, in the viscosity since in the fully polarized phase it has increased by a factor of 200 or more.[7,18] A recent measurement of the viscosity[19] as a function of the polarization is consistent with our prediction.

(4) Recent experiments[20] suggest that χ_n increases, reaching a maximum at some finite polarization. Unfortunately, this is not conclusive since it is difficult to extract the liquid susceptibility from the measurements. Moreover, the interpretation of the data requires a detailed model of melting which is currently not understood.

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(a)Permanent address: Los Alamos National Laboratory, T-11, MS-B262, Los Alamos, NM 87545.

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