A Model for Solid $^3$He: II

J. Dubois and P. Kumar

Department of Physics and National High Magnetic Field Laboratory

University of Florida

Gainesville, FL 32611

Abstract

We propose a simple Ginzburg-Landau free energy to describe the magnetic phase transition in solid $^3$He. The free energy is analyzed with due consideration of the hard first order transitions at low magnetic fields. The resulting phase diagram contains all of the important features of the experimentally observed phase diagram. The free energy also yields a critical field at which the transition from the disordered state to the high field state changes from a first order to a second order one.

PACS No. 67.80.Jd, 75.10.Hk, 75.30.Kz
I. Introduction

Solid $^3$He undergoes a phase transition\(^{(1)}\) at $T_N = 1\, mK$ into an ordered state that consists of ferromagnetic (100) planes and two successive planes with parallel spins followed by the next pair of planes where the spins are antiparallel. This state, as the magnetic field is increased to approximately 0.5 T, is replaced by a normal canted antiferromagnet. The transition to the low field uudd (up-up-down-down) phase is first order. It is also first order to the canted antiferromagnet phase for fields $B < 0.6\, T$. It becomes second order\(^{(2)}\) for $B > 0.6\, T$. The point $B = 0.6\, T$ is similar to a critical point.

The standard model\(^{(3)}\) for solid $^3$He consists of pair, triple and four spin exchanges analyzed within a mean field theory. The model has been successful in deriving a large number of experimental observations. The multiple spin exchange model provides important physical insights. It shows that because of the competing interactions (even spin permutations lead to antiferromagnetic and odd permutations to ferromagnetic exchange interactions, see for example D. J. Thouless, ref. (3)) both the Curie-Weiss constant and the bare Neel temperature are small. This feature for example resolves a long standing disagreement between exchange constants measured from $T_1$ measurements and from Curie-Weiss temperature. While the former is an average of squares of the exchange constant, the latter is a linear sum. Exchange constants of different signs contribute differently to the two observables. It also shows that solid $^3$He is a frustrated antiferromagnet. There are some difficulties. For example the molar volume dependence of various observables is almost identical, giving rise to suspicion\(^{(4)}\) that a much smaller number of energy scales are involved. Quantum Monte Carlo calculations\(^{(5)}\) show a weak convergence in the magnitudes of exchanges involving larger number of spins. Finally, multiple spin exchange models are sufficiently complex so that it often becomes difficult to obtain a qualitative understanding of processes in solid $^3$He, something that lately has become essential as more complex phenomena are discovered. For example, in thermal conductivity measurements\(^{(6)}\) there is evidence for magnetic defects and we hope to be able to calculate the energies of these defects through a Ginzburg-Landau model.

Our aim in this paper is to propose a Ginzburg-Landau type free energy. The free energy is not—at least yet—derived from any microscopic Hamiltonian. Its terms are those allowed by the various ground states, satisfying lattice symmetries, and interactions between them. We study thermodynamic consequences with the aim of fine tuning its terms to produce a coherent, albeit phenomenological model for solid $^3$He. Eventually, one task will be a microscopic calculation of the parameters.
Our starting point is a proposal by Guyer and Kumar\(^{(7)}\) (GK). Noting that the spin susceptibility was close to Curie law just above the transition at low fields, they argued that this implied a special cancellation of the interaction energies. They further argued that the nature of the first order transition, hard in the sense that the entropy discontinuity is a major fraction of \(R \ln 2\), further implied that the free energy for solid \(^3\)He is given by

\[
F = Jm_2^4 - m_0 B - T \Sigma(m)
\]  

(1)

Here \(B\) is the magnetic field, \(T\) is the temperature and \(\Sigma\), the entropy given by

\[
\Sigma(S) = \ln 2 - \frac{1}{2}[(1 + S) \ln(1 + S) + (1 - S) \ln(1 - S)]
\]  

(2)

The magnetization \(m(r)\) is a vector of length \(\mu_N S\) and is given by

\[
m(r) = m_0 + \tilde{m}_2 \cos(k_2 \cdot r + \pi/4)
\]  

(3)

The vectors \(\tilde{m}_0 \cdot \tilde{m}_2 = 0\) and \(|m|^2 = m_0^2 + m_2^2\). In our notation, when the vector sign is omitted, the quantity refers to the amplitude of the vector. Here \(k_2\) is half of the reciprocal lattice vector in (100) direction and leads to the uudd state. The phase angle \(\pi/4\) ensures that all spins are of the same length. The entropy \(\Sigma(S)\) is the entropy of a spin \(1/2\) system. In general the entropy is a consequence of proper configuration counting subject to the constraints on the length and the discrete nature of the spins. The expression in Eq. (2) reproduces the mean field equation of state. In the following we shall use the entropy as in Eq. (2). Note however that \(S\) is scaled so that its two possible values are \(S^2 = 1\). The effect of a quadratic term in \(m_2\) has been discussed in some detail in ref. (8).

In a recent preprint\(^{(8)}\) we have argued that a Ginzburg-Landau theory as an expansion in powers of the order parameter is inappropriate (and inadequate) to describe a first order phase transition. In a hard first order phase transition, the order parameter, at the phase transition jumps to a value which is easily outside the range of validity of Ginzburg-Landau theory. We cannot expand the entropy in Eq. (1) in powers of the order parameter and Eq. (1), subject to the constraint, \(m^2 = m_0^2 + m_1^2 + m_2^2\) must be solved, if necessary, numerically as was already done in ref. (8). While Eq. (1) was applicable only at low fields and near the phase transition, the corresponding free energy for all fields is described in the next section.

This paper is organized as follows: in section II, we introduce the free energy involving leading contributions of all of the possible ground states. Section III contains a solution
of the phase diagram for $B = T = 0$ in the parameter space of the free energy. Section IV describes the results and we conclude with a summary and a discussion of possible improvements.

II. The Model

The minimal model should contain the three order parameters $m_0$, $m_1$, and $m_2$ such that local magnetization is given by $m(r) = m_0 + m_1 \cos(k_1 r) + m_2 \cos(k_2 r)$. Here $k_1$ and $k_2$ are respectively the wavevectors for the simple antiferromagnetic and the uudd states. The transition in $m_2$ is hard first order and we consider only terms quartic in $m_2$. The order parameter $m_1$ represents the amplitude of a simple antiferromagnetic state, present in the high field part of the phase diagram. Since the phase boundary between high field phase and the paramagnetic phase has a positive slope ($\frac{dB}{dT} > 0$) in the high field phase, $m_0$ must have an attractive interaction with the mode $m_1$. In general, $\sim m_0$, $\sim m_1$ and $\sim m_2$ are all vectors. We will omit this and also any anisotropy energy terms; they are much smaller energy scales. We have for the free energy

$$F = -Jm_2^4 - J_1m_1^2 - J_2m_0^2m_1^2 - m_0B - k_BT \Sigma (S)$$

(3)

The various $m_i = m_0 S_i$ where $m_0 = g \mu_B |S|$. Thus the amplitudes $S_0$, $S_1$ and $S_2$, at $T = 0$ can be represented by points on a unit circle in 3 dimensions. In scaled variables

$$f = F/Jm_0^4 = -S_2^4 - g_1 S_1^2 - g_2 S_0^2 S_1^2 - b S_0 - t \Sigma (S)$$

(4)

Here $g_1 = J_1/Jm_0^2$, $g_2 = J_2/J$, $b = B/Jm_0^3$ and $t = k_BT/Jm_0^4$. The amplitude $S$ is given by $S^2 = S_0^2 + S_1^2 + S_2^2$.

At $b = t = 0$, we can draw a phase diagram (see Fig. 1) in the plane $(g_1, g_2)$ for the free energy in Eq. (4). At $t = 0$, we have $S^2 = 1$. For small $g_1$ and $g_2$, only $S_2$ can be non-zero. Since $f$ is monotonic in $S_2$, the minimum for $f$ must lie at $S_2 = 1$ so that $f = -1$. We call this region I. In region II, $g_2$ is small and so we take $S_0 = S_2 = 0$. Again since the minimum of $f$ (at $S_1 = 1$) must be $f_{II} = -g_1$. This minimum is lower than region I when $g_1 > 1$. Thus the phase boundary I–II must be $g_1 = 1$.

For larger values of $g_1$ and $g_2$, we expect $S_2 = 0$. We can write

$$f_{III} = -g_1(1 - S_0^2) - g_2 S_0^2(1 - S_0^2)$$

(5)

Its minimization with respect to $S_0$ leads to

$$S_0^2 = \frac{1}{2} \left( 1 - \frac{g_1}{g_2} \right) , \quad S_1^2 = 1 - S_0^2 = \frac{1}{2} \left( 1 + \frac{g_1}{g_2} \right)$$

(6)
and

\[ f_{III} = -g_1 - \frac{g_2}{4} \left( 1 - \frac{g_1}{g_2} \right)^2 = -\frac{(g_1 + g_2)^2}{4g_2} \] (7)

The boundary II–III, given by \( f_{II} = f_{III} \) is therefore \( g_1 = g_2 \). The boundary I–III is given by

\[ g_2 = 2 - g_1 + 2\sqrt{(1 - g_1)} \sim 4 - 2g_1 + O(g_1^2) \] (8)

The point \((g_1, g_2) = (0, 4)\) can also be derived from a stability analysis. The point \((g_1, g_2) = (1, 1)\) is a confluence of three different phases and may be called a triple point. These results are described in Fig. (1).

At \( t = 0 \), the problem is still solvable, to some extent. For example, we consider the case of the upper critical field. In the ordered state, \( S_2 = 0 \) and \( S_1 \) vanishes at the upper critical field. The transitions are all second order. We have (on substituting \( S^2_0 = 1 - S^2_1 \)), and expanding in powers of \( S_1 \)

\[ f = -b - \left[ (g_1 + g_2) - \frac{b}{2} \right] S_1^2 + \left( g_2 + \frac{b}{8} \right) S_1^4 + O(S_1^6) \] (9)

We see that an \( S_1 \neq 0 \) solution exists only for \( b < 2(g_1 + g_2) = b_{c_2} \). In order that the \( b = 0 \) ground state be \( S_2 \neq 0 \), the \((g_1, g_2)\) parameters have to come from region I. For \( g_2 > 1 \), Eq. (8) leads to an inequality,

\[ b_{c_2} = 2(g_1 + g_2) \leq 4\sqrt{g_2} \] (10)

To calculate the lower critical field, \( b_{c_1} \), we note that the field causes the ground state to change from the one in region I to the one in region II. For a given set of \((g_1, g_2)\) the ground state in region II contains a uniform magnetization \( S_0 \) given by Eq. (6). In region II the free energy is lowered by the Zeeman interaction \(-b \cdot S_0\). To order \( b \) the free energy in region I remains unchanged. Equating the two free energies, we get,

\[ b_{c_1} = \sqrt{\frac{2g_2}{(g_2 - g_1)} \left[ 1 - \frac{(g_1 + g_2)^2}{4g_2} \right]} \] (11)

The lower critical field is negligibly small near the zero field phase boundary between regions I and III. It also diverges at the line \( g_1 = g_2 \). Experimentally the value of the ratio \( b_{c_2}/b_{c_1} \approx 40 \) and the parameters have to be rather close to the I-III phase boundary. While \( g_2 < 4 \), \( g_1 \) must be quite small.
To determine the critical field when the transition to the high field state becomes second order, we have to include the temperature \( t \neq 0 \). Since the transition is second order, at least for \( b > b_t \), a Ginzburg-Landau expansion of the entropy is expected to be valid for \( S_1 \). However the magnetization \( S_0 \) is large and the entropy expansion does not work.

III. Phase Diagram

We have chosen \((g_1, g_2) = (0.5, 1.5)\) for illustration. The ratio of upper critical field to lower critical field for this choice is 7. This is far from the experimental (see e.g. Godfrin and Osheroff in ref. 3) value 47. However, the qualitative features are essentially independent of the exact value of \((g_1, g_2)\) in a given region in Fig. (1).

It is not possible to determine \((g_1, g_2)\) from the \( T = 0 \) critical fields alone. For a given ratio of \( B_{c2}/B_{c1} \), we can determine a contour in the \((g_1, g_2)\) plane. If we take the experimental value of the \( B_{c1} = 0.45T \), then \( b_{c1}/t_c = 0.37 \) using the experimental constants for the nuclear Bohr magneton. Thus \( b_{c1} = 0.53 \), (for \( t_c = 1.45 \)). Using the ratio for \( B_{c2}/B_{c1} = 47 \), we get \( b_{c2} = 25 \). The model has an upper limit of \( b_{c2} < 8 \) to guarantee that the ground state at \( T = 0 \) is the uudd state. There is an easy resolution of this problem, namely a g shift, all \( b \)'s are replaced by \( g_3b \). This however leaves us unable to determine the parameters based on the phase diagram alone. In any case there needs to be some accommodation of the finite temperature renormalizations which in turn involves the mean field assumption on the entropy. We therefore are reluctant to use finite temperature observables. We hope to calculate temperature independent properties such as spin wave velocities and the energies of defects to fix the parameters at a later date.

At \( b = 0 \), the transition occurs at \( t_c = 1.45 \) into the uudd state. The entropy discontinuity is almost full in accord with ref. (7). Note that if the entropy \( \Sigma(S) \) had been expanded in powers of \( S \), as noted in ref. (8), the transition temperature would have been 2.93 \( (t_c^{-1} = \frac{1}{12} + \frac{1}{\sqrt{15}}) \) and the change in \( S_2, \Delta S_2 = 1.97 \). In the present calculations \( \Delta S_2 = 1 \). Since \( S_0 \) and \( S_1 \) are zero in the ground state, the transition temperature is independent of \((g_1, g_2)\). However the lower critical field does depend on these parameters.

As the magnetic field increases, \( t_c \) slowly decreases. There is a point at which all three phases \((S_2, (S_0, S_1)\) and paramagnetic) coexist, a triple point. The transition between low field phase \((S_2)\) and high field phase \((S_0, S_1)\) is first order, as is the transition between the paramagnetic and high field state. These experimental features appear to be reproduced by the model. It came as a surprise that the model also produces a critical point. This is
the point \((b_t, t_t)\) at which the first order phase transition between disordered paramagnetic phase \(S_0 \neq 0, S_1 = 0\) and the high field phase \((S_0, S_1) \neq 0\) becomes a second order phase transition.

We thus see in Fig. (2) a complete phase diagram for solid \(^3\)He magnetism. In region I, the ground state is the well known up-up-down-down state. In region III, the ground state is \(S_0 \neq 0, S_1 \neq 0\). Region II represents the disordered state. Again the choice of parameters determines the ratio \(b_{c2}/b_{c1} \approx 7\). The transition at \(b = 0\) is independent of the parameters \((g_1, g_2)\) and is at 1.4. The precise value of \(b_{c1}\) (at .6) clearly depends on \((g_1, g_2)\).

In Fig. (2), the solid lines describe a first order phase transition while the dashed line represents a second order transition. Fig. (3) shows the discontinuity in \(S_1(\Delta S_1)\) as a function of magnetic field along the phase boundary. We see that \(\Delta S_1\) vanishes at \(b = 1.25\). For \(b > 1.25\), the phase transition is second order. The transition temperature increases with magnetic field until \(t \approx 2.2\) and \(b = 2.7\). For \(b > 2.7\), the transition temperature decreases with increasing magnetic field, reaching \(t = 0\) at \(b_{c2} = 2(g_1 + g_2) = 4\).

IV. Summary

We have shown that the phase diagram of solid \(^3\)He can be derived from rather simple free energy considerations. These are extensions of a model proposed by Guyer and Kumar for the ordering transition at low magnetic field. The present results reproduce the essential features of the phase diagram including the critical point in the high field-paramagnetic phase boundary.

There are two possible directions for further work, both involving the introduction of a space gradient dependent term in the free energy. These are (1) the derivation of magnon (spin wave) dispersion and (2) analysis of defects and their interaction with the magnons. The dynamics of spins can be easily written down using the Bloch equations for the motion of spins subjected to a local field derived from Eq. (4). The defects are most likely domain walls between metastable states frozen into the true ground state. We will return to these questions later.

We acknowledge useful discussions with E. D. Adams, R. Guyer, M. Roger and N. S. Sullivan. This work was partially supported by the National High Magnetic Field Laboratory and the US Department of Energy, grant DEF G05-91-ER45462.
1. There are several reviews of experiments on solid $^3$He with references to original experiments. See for example E.D. Adams, Can. J. Phys. 65, 1336 (1987) and D.D. Osheroff, J. Low Temp. Phys. 87, 297 (1992). Also see D.E. Greywall and P. Busch, Phys. Rev. B36, 6853 (1987).

2. J.S. Xia, W. Ni and E.D. Adams, Phys. Rev. Lett. 70, 1481 (1993).

3. The multiple spin exchange model has been reviewed by M. Roger, J. H. Hetherington and J.M. Delrieu, Rev. Mod. Phys. 55, 1 (1983). A more recent discussion can be found in D. D. Osheroff, H. Godfrin R. Ruel, Phys. Rev. Lett. 58, 2458 (1987) and H. Godfrin and D.D. Osheroff, Phys. Rev. B38, 4492 (1988). An early discussion can be found in D. J. Thouless, Proc. Phys. Soc. London 86, 893 (1965); 86, 905 (1965).

4. See for example Osheroff (ref.(1)) and Greywall and Busch (ref. (1)).

5. D. M. Ceperley and G. Jacucci, Phys. Rev. Lett. 58, 1648 (1987), also see Godfrin and Osheroff op. cit. ref. (3).

6. Y.P. Feng, P. Schiffer, J. Mihalism and D. D. Osheroff, Phys. Rev. Lett., 65, 1450 (1990); Y.P. Feng, P. Schiffer and D. D. Osheroff, Phys. Rev. B49, 8790 (1994). These papers report thermal conductivity due to scattering of magnons from umklapp scattering and defects. The properties of defects are one objective in setting up the simple formalism reported here.

7. R.A. Guyer and P. Kumar, J. Low Temp. Phys. 47, 321 (1982).

8. J. Dubois, B. Nelson and P. Kumar, UF preprint (1994).
Figure Captions

1. The phase diagram in the \((g_1, g_2)\) parameter space at \(b = t = 0\), zero magnetic field and temperature. In region I, the only non-zero order parameter is \(S_2\), representing the up-up-down-down state. All other amplitudes \((S_0, S_1)\) are zero. Similarly in region II only \(S_1 \neq 0, S_0 = S_2 = 0\). In region III in contrast, \(S_2 = 0\) and \(S_0\) and \(S_1\) are finite.

2. The phase diagram in the \((b, t)\) plane for \((g_1, g_2) = (0.5, 1.5)\). The solid lines represent a first order phase transition while the dashed line represents a second order transition.

3. The discontinuity in \(S_1(\Delta S_1)\) at the phase transition. At small fields \((b \gtrsim 0.5)\), the transition is clearly hard second order since \(\Delta S_1 \sim 0.6\). However for \(b > 1.25\), the transition is second order.