

Solid ^3He as a Heisenberg antiferromagnet

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A detailed analysis with a localized magnetic exchange Hamiltonian has been made of existing thermodynamic data on bcc ^3He with the bulk of the analysis performed at the molar volumes $v = 23.34, 23.88, 24.0,$ and $24.25 \text{ cm}^3/\text{mole}$ for $T < 50 \text{ mK}$. The data can be described at all v equally well by four qualitatively different sets of exchange constants. This is true with the possible exception of one set of specific-heat data for which it is not entirely clear that they can be described by a purely magnetic Hamiltonian alone. Except on the melting curve at $v = 24.25$ and possibly for the just-mentioned specific-heat data, one of these four sets is the conventional set, which is supported by exchange calculations. All this points to the possibility of a description of bcc ^3He different from the conventional picture. Experiments that could clarify the situation are discussed.

The generally accepted description of the thermodynamic properties of bcc solid ^3He below a temperature of $T = 50 \text{ mK}$ is based on the localized exchange Hamiltonian

$$\frac{H}{k_B} = -2\Lambda_1 \sum_{i,j}^{\text{nn}} \vec{I}_i \cdot \vec{I}_j - 2\Lambda_2 \sum_{i,j}^{\text{nnn}} \vec{I}_i \cdot \vec{I}_j - \gamma H_0 \sum_i I_i^z. \quad (1)$$

Here \sum^{nn} (\sum^{nnn}) indicates a sum over nearest-neighbor (next-nearest-neighbor) pairs, and \sum_i is a sum over all lattice sites; $I_i^\mu = \frac{1}{2}\sigma_i^\mu$, where σ_i^μ , $\mu = x, y, z$, are Pauli matrices; H_0 is the external magnetic field; $\gamma = 1.556 \times 10^{-7} \text{ }^\circ\text{K/G}$; and k_B is Boltzmann's constant. For the temperature range of interest the exchange constants Λ_1 and Λ_2 are taken to be functions only of the molar volume v .¹⁻⁶

Λ_1 is usually considered to be antiferromagnetic, $\Lambda_1 < 0$, and Λ_2 , ferromagnetic, $\Lambda_2 > 0$, with Λ_1 being the larger in magnitude. Exchange constants with only these signs have been obtained from exchange calculations based on a generalization of Herring's theory of electron exchange in insulators to the case of atom exchange in solid ^3He .⁷ The basic problem in these calculations is the proper selection of a localized (home-based) wave function for a single ^3He atom around its lattice site and the correct description of the correlations introduced by the hard-core repulsion of the ^3He atoms. Theoretical calculations of the Λ 's have been made with a variety of home-based wave functions and correlation functions, resulting, however, in Λ 's that vary by orders of magnitude.^{5,6}

At first a nearest-neighbor Heisenberg model ($\Lambda_2 = 0$) was quite adequate to analyze the data. Since the experiment of Kirk and Adams⁸ could not be explained on the basis of such a model, Zane proposed a Hamiltonian of the form (1) with $\Lambda_2 \neq 0$.³ Stimulated by the possibility to explain the data of Kirk and Adams with Hamiltonian (1), McMahan and Guyer incorporated, more systematically, the effects of multiple-exchange processes into an exchange Hamiltonian.⁶ However, their calculations still suffered from the above-mentioned uncertainty in the choices of the home-based wave function and the correlation function with the attendant variation in the predicted magnitudes of the Λ 's.

Within the present experimental accuracy and with the possible exception of the Dundon and Goodkind (DG) experiment⁹ discussed below, all known thermodynamic data for $T < 50 \text{ mK}$ can be described on the basis of the Hamiltonian (1) with $\Lambda_1 < 0$ and $\Lambda_2 > 0$ for all v , except for $v = 24.25$, where the data force $\Lambda_2 < 0$.¹⁰ (v is always in units of cm^3/mole .) However, the data can *equally* well be described by a variety of sets of Λ_1 and Λ_2 that not only have other signs but also a great variation in absolute values (cf. sets I-IV in Table I). The Dundon-Goodkind experiment can also be made consistent with the Hamiltonian (1) and the sets I-IV of Table I. However, if one accepts DG's analysis of their data, only two sets, one essentially set III of Table I and the other a new set with Λ_1 and Λ_2 both negative for all v , lead to consistency with all thermodynamic data.

Some experimental data require changes of sign in Λ_2 while other data imply $\Lambda_2 < 0$ for all v . This is at variance with present exchange calcula-

tions since such calculations have, to date, always predicted one particular set of signs for Λ_1 and Λ_2 ; $\Lambda_1 < 0$ and $\Lambda_2 > 0$. However, more collective pictures of bcc ^3He cannot *a priori* be ruled out and unorthodox values of Λ_1 and Λ_2 , that describe the data for $T \leq 50$ mK, could be viewed as an attempt to mimic a more collective behavior of solid ^3He with an effective Hamiltonian of the form of Eq. (1). In this connective it is interesting to note the contribution, linear in the temperature, to the specific heat at constant volume and zero magnetic field, $C_{v,0}$, around 100 mK.¹¹ Dzyaloshinskii *et al.* have attempted to relate this behavior to a Fermi-liquid model of ^3He .¹² One should also mention the possibility of effects on the exchange constants of the phonon modes that have been considered by Nosanow and Varma.¹³

We stress that we do not want to imply that the Hamiltonian (1) is correct nor that it is incorrect and that a more collective description should be made. We merely want to indicate that on the basis of present experimental data no clear decision can be made as to the correctness and proper interpretation of the Hamiltonian (1) or as to a unique set of Λ 's. In other words, the nature of the proper Hamiltonian to describe solid ^3He at low temperatures is still open.

Sets of ^3He data, more restricted than that we considered, have been previously analyzed by Goldstein¹⁴ and by Guyer.¹⁵ Goldstein used the pressure data of Kirk and Adams to determine the four constants c_i and γ_i , assuming a dependence of the Λ 's on v of the form $\Lambda_i = c_i v^{\gamma_i}$ ($i=1, 2$), where the c_i and γ_i were taken to be volume, temperature, and magnetic field independent. Goldstein obtained essentially our set I, for $v \leq 24.0$, to fit the data with the Hamiltonian (1). Guyer proceeded to determine the compatibility of the Hamiltonian (1) with all thermodynamic data then available but concluded that the data and Eq. (1) were inconsistent. It is not clear that such a conclusion is necessary, because Guyer may have underestimated the uncertainties in the Weiss susceptibility data, as well as, in the magnetic-phase-transition temperature of ^3He when calculated from the Λ 's by mean-field theory.

The data that we analyze can be divided into two classes.¹⁶

(1) *High-temperature data*: thermodynamic quantities in a region of T and H_0 , where they can be calculated by a few terms of a high- T and small- H_0 expansion. In this class belong the following: (i) the pressure data of Kirk and Adams⁸ for the pressure p as a function of T and H_0 at $v = 23.34$, 23.88, and 24.0; (ii) the data of Panczyk and

TABLE I. Results of data fitting. For $v = 24.25$ the γ 's are not known and there are two choices of the Λ 's for sets I and II consistent with smaller volumes. The large γ values (with an asterisk) arise because γ , the relative variation of Λ with v , can become very large when $\Lambda \approx 0$; the α 's are always smooth functions of v . In sets I and II, $\Lambda_1 < 0$ and is dominant, while in sets III and IV, $\Lambda_2 < 0$ and dominant.

| Volume (cm ³ /mole) | Set | Λ_1 (mK) | Λ_2 (mK) | γ_1 | γ_2 |
|-----------------------------------|-----|---------------------|---------------------|------------|------------|
| 23.34 | I | -0.39 | 0.14 | 15.9 | 24.3 |
| | II | -0.40 | -0.014 | 17.5 | -324.1* |
| | III | 0.094 | -0.48 | 20.1 | 15.2 |
| | IV | -0.093 | -0.45 | -33.7 | 20.1 |
| 23.88 | I | -0.55 | 0.25 | 15.3 | 26.1 |
| | II | -0.60 | 0.083 | 14.9 | 83.6 |
| | III | 0.16 | -0.73 | 25.0 | 14.3 |
| | IV | -0.013 | -0.70 | -374.1* | 16.6 |
| 24.0 | I | -0.64 | 0.28 | 15.5 | 17.7 |
| | II | -0.71 | 0.12 | 13.9 | 32.6 |
| | III | 0.18 | -0.77 | 19.8 | 16.0 |
| | IV | 0.014 | -0.83 | 193.2* | 14.2 |
| 24.25 | I | -0.6 | -0.6 | ... | ... |
| | | -0.76 | -0.28 | ... | ... |
| | II | -0.6 | -0.6 | ... | ... |
| | | -0.76 | -0.28 | ... | ... |
| | III | -0.6 | -0.6 | ... | ... |
| | | -0.6 | -0.6 | ... | ... |

Adams¹⁷ for p as a function of T at $H_0 = 0$ for $21.0 \leq v \leq 24.0$; (iii) the zero-field susceptibility per spin χ_0 data of Kirk *et al.*¹⁸ for $21.0 \leq v \leq 24.0$; (iv) the $C_{v,0}$ data of Castles and Adams¹¹ and Dundon and Goodkind⁹ for $T > 5$ mK and for $23.1 \leq v \leq 24.13$.

(2) *Low-temperature data*: the zero-field transition temperature T_c for the magnetic phase transition. This class contains the following T_c data: (i) Dundon and Goodkind⁹ for $T < 5$ mK and for $v = 23.8$ and 23.1; (ii) Halperin *et al.*¹⁰ on the melting curve, $v = 24.25$.

Ignoring for the moment the Dundon and Goodkind data of class 1 which will be discussed later, all the other data of class 1 are analyzed through the high-temperature expansions of Dalton and Wood¹⁹ and Baker *et al.*²⁰ for the free energy and χ_0 . The first few terms of the p , χ_0 , and $C_{v,0}$ expansions are

$$vp/AT = 3a/T^2 + \gamma^2 H_0^2 b/2T^3 + \dots, \quad (2a)$$

$$4T\chi_0/k_B\gamma^2 = 1 + 4c/T + \dots, \quad (2b)$$

and

$$C_{v,0} = 3Nk_B d/T^2 + \dots. \quad (2c)$$

Here p is in atmospheres; $A = 8.206 \times 10^7 \text{ cm}^3 \text{ atm/mole } ^\circ\text{K}$; T is in $^\circ\text{K}$; and N is the number of spins. Furthermore,

$$a = \Lambda_1 \alpha_1 + \frac{3}{4} \Lambda_2 \alpha_2, \quad (3a)$$

$$b = \alpha_1 + \frac{3}{4} \alpha_2, \quad (3b)$$

$$c = \Lambda_1 + \frac{3}{4} \Lambda_2, \quad (3c)$$

and

$$d = \Lambda_1^2 + \frac{3}{4} \Lambda_2^2, \quad (3d)$$

where

$$\alpha_i = \Lambda_i \gamma_i$$

and

$$\gamma_i = \frac{d(\ln |\Lambda_i|)}{d(\ln v)};$$

$i = 1, 2$. a , b , c , and d are functions of v only.²¹

The Λ 's and α 's for $v = 23.34$, 23.88 , and 24.0 are derived from the data of class 1 through a , b , c , and d . Here d can be determined either from $C_{v,0}$ or, for a wider range of v , from the volume dependence of a , since $d(d)/d(\ln v) = 2a$. The two methods give consistent values of d for all v .

The values of a , b , c , and d as a function of v were derived from the experimental data in the following way. First, for each v , a and d were determined from the more reliable $H_0 = 0$ data of class 1 (i) and 1 (ii). Then a value of c , consistent with the data of class 1 (iii) was assumed. Since these χ_0 data were very uncertain, only the sign and order of magnitude of c could be determined.²² Finally, a value of b was sought that would give—the best fit of the data of class 1 (i).²³ a and b then determined α_1 and α_2 once Λ_1 and Λ_2 were found from c and d . The data of class 1 (iv)—still excluding the DG data—appear to be always consistent with the values of a – d found in this way.²⁴

This procedure leads to four qualitatively different sets of Λ 's and α 's (or γ 's) that describe all the experiments of class 1 equally well (DG to be discussed later). Two sets are obtained because the equation for d is quadratic in the Λ 's, leading with the equation for c to two possible solutions for the Λ 's,

$$\Lambda_1 = \frac{4}{7} c \mp \frac{3}{7} \left[\frac{1}{3} (7d - 4c^2) \right]^{1/2} \quad (4a)$$

and

$$\Lambda_2 = \frac{4}{7} c \pm \frac{3}{7} \left[\frac{1}{3} (7d - 4c^2) \right]^{1/2}, \quad (4b)$$

where either both upper or both lower signs have to be taken in these equations. These two sets of Λ 's lead to four qualitatively different sets of Λ 's because of the freedom in the choice of $|c|$ ($c < 0$

always) as follows from the uncertain χ_0 data.

Once the Λ 's are known and the α 's (or γ 's) are obtained, an important check is that the γ 's should be consistent with their definitions as volume derivatives of the Λ 's. In view of the small volume variations a linear volume dependence of the Λ 's on v can be assumed. In Table I we list four typical sets for each of the volumes $v = 23.34$, 23.88 , and 24.0 . Sets I and II use the upper signs and have $\Lambda_1 < 0$ and dominant, while the sets III and IV use the lower signs in Eqs. (4) and have $\Lambda_2 < 0$ and dominant. For these volumes the set I is the conventional one with $\Lambda_1 < 0$ and $\Lambda_2 > 0$. One can go continuously from set I to set II and from set III to set IV by varying c consistent with the χ_0 experiment.

We make two remarks:

(a) If the upper signs are chosen in Eqs. (4), then $\Lambda_2 \leq 0$ when $c^2 \geq d$. Taking the χ_0 data at face value, this would imply that Λ_2 as a function of v changes sign, with $\Lambda_2 > 0$ for $v \geq 22.0$ and $\Lambda_2 < 0$ for $v \leq 22.0$. However, a Λ_2 changing sign is at variance with present exchange theory results.

(b) For the Λ 's to be real, one must have $\frac{1}{4} d \geq c^2$. This condition would be violated at $v = 21.0$ if the χ_0 data are taken at face value. We stress that in view of the insufficient accuracy of the χ_0 data and the ensuing uncertainty in $|c|$, these two points should be considered with great caution.

Once the Λ 's and γ 's are known from the class-1 data—the DG experiment excluded—we will determine their consistency with the low-temperature data of class 2. We will use below the results of Pirnie *et al.*²⁵ which relate $\alpha = \Lambda_2/\Lambda_1$ to T_c/Λ_1 for a system with Hamiltonian (1). Such a relation can be obtained by using Padé approximants on high-temperature expansions of χ_0 and the staggered susceptibility to determine T_c/Λ_1 .

(a) Dundon and Goodkind suggest the existence of (magnetic) phase transitions at $T \cong 2$ mK and $T \cong 1.4$ mK for $v \cong 23.8$ and at $T \cong 1.5$ mK for $v \cong 23.1$. Using the results of Pirnie *et al.*, we obtain from sets I–IV of Table I $T_c = 2.06$, 1.87 , 1.46 , and 1.36 mK, respectively, for $v = 23.88$. Each of these T_c agrees with one of the measured T_c ; those obtained from sets I and II with $T_c \cong 2$ mK and those from sets III and IV with $T_c \cong 1.4$ mK. However, linear extrapolation down to $v = 23.1$ yields T_c that do not exceed about 1 mK, which is much smaller than the observed $T_c \cong 1.5$ mK at $v = 23.1$. In view of the difficulty of identifying a phase transition at this volume, this should not be construed as a serious inconsistency.

Dundon and Goodkind suggest the possibility of more than one phase transition at $v = 23.8$. If true, this would seem to be in disagreement with the sets I and II of Table I since these sets of Λ 's

give for $H_0=0$ —at least according to mean field theory—only one phase transition from a paramagnetic to an antiferromagnetic phase. The sets III and IV, on the other hand, could, using the reasoning of Swendsen,²⁶ possibly lead to two phase transitions for $H_0=0$. Swendsen argued his point in the following way. He found that the critical temperature as a function of $\alpha = \Lambda_2/\Lambda_1$ exhibited an anomaly at a certain value of α . This anomaly was characterized by a change in sign of the difference between the Padé approximant estimate and the random-phase estimate of the critical temperature. Concurrently, he noted that instabilities occur for approximately the same value of α in the mean-field and random-phase calculations of the magnetic phases of the system. He concluded that at this value of α a new type of ordered state occurs, where the spins are aligned in a canted or perpendicular fashion. Then upon lowering the temperature, the system could first make a transition from the paramagnetic to this canted or perpendicularly aligned state before it makes a second transition to the antiferromagnetic state.

If one is not prepared to identify (magnetic) phase transitions in the $T < 5$ mK specific heat data of DG, then these data have no bearing on the validity of using Hamiltonian (1) and our sets I–IV to describe the magnetic properties of solid ^3He .

(b) We found it possible to obtain consistency between the experiments of Halperin *et al.* and all the sets I–IV in Table I. Halperin *et al.* find $T_c = 1.17$ mK at $v = 24.25$. Since this volume lies outside the range of volumes used above in the determination of sets I–IV, we have to extrapolate in order to compare. Because straightforward linear extrapolation of the Λ 's as a function of v leads at best (set IV) to the too large value of $T_c = 1.72$ mK, we investigated the possible consistency of Halperin's data with the previous results in a different fashion. Using Pirnie's results for T_c/Λ_1 as a function of α , one can derive from $T_c = 1.17$ mK and a value of $d = 0.64$ (mK)² (obtained by a linear extrapolation of $\ln d$), two sets of Λ 's: (i) $\Lambda_1 = \Lambda_2 = -0.6$ mK or (ii) $\Lambda_1 = -0.76$ mK and $\Lambda_2 = -0.28$ mK. All four sets I–IV can be made internally consistent, as far as volume dependence is concerned, with (i), if one uses a rapid decrease in b between $v = 24.0$ and $v = 24.25$ that seems already implied by the variation of b between $v = 23.34$ and $v = 24.0$.²⁷ However, only sets I and II can be made consistent with (ii) as far as volume dependence is concerned. We remark that the values of (i) and (ii) are both inconsistent with present exchange calculations.²⁸

Finally, we discuss Dundon and Goodkind's $T > 5$ mK specific-heat data of class 1. It is not

entirely clear that these data represent a purely magnetic contribution to the specific heat and that, therefore, an analysis with Eq. (2c) on the basis of the Hamiltonian (1) is meaningful. In fact, these data can only be described on the basis of an expansion of the form:

$$C_{v,0}/Nk_B = f + 3d/T^2 - 3e/T^3 + \dots, \quad (5)$$

where

$$e = \Lambda_1^3 + \frac{3}{4}\Lambda_2^3 - 9\Lambda_1^2\Lambda_2.$$

However, f cannot consistently be assumed to be zero for all v , as should be the case for the Hamiltonian (1) [cf. Eq. (2c)]. In their least-square analysis DG find values for d whose magnitudes and v dependence are not quite consistent with other experiments,¹ while their values for f do not always include zero in their respectively quoted ranges of uncertainty. In addition, a value of $e \geq 0$ is derived that would only be consistent with two sets of Λ 's, one essentially our set III and the other a set with both Λ_1 and Λ_2 negative for all v ($\Lambda_1 = -0.35, -0.57,$ and -0.69 mK and $\Lambda_2 = -0.25, -0.12,$ and -0.09 mK for $v = 23.34, 23.88,$ and $24.$, respectively). In view of the difficulties with d and f , it is not clear to what extent $e \geq 0$ and its consequences should be taken seriously.

Alternatively, we can try to fit the specific heat data with Eq. (5) with d and e determined by the sets I–IV of Table I, using f as a free parameter. Then curves are obtained which are either indistinguishable or only slightly deviating, well within the experimental accuracy, from the least-square fits of DG.²⁹ The values of f then obtained are larger than those obtained by DG and would again indicate the possible presence of an anomalous nonmagnetic contribution to the specific heat.

Although the DG data of class 1 might therefore exclude sets I, II, and IV of Table I, it seems premature to conclude this on the basis of present evidence.

Summarizing, one can say, that there is no contradiction—but no unique way either—in relating the known experimental data to a Hamiltonian of the form (1), except possibly for the χ_0 data at $v = 21.0$ and possibly for the Dundon and Goodkind $C_{v,0}$ data for $T > 5$ mK, if one takes $f \neq 0$ seriously. There is an inconsistency ($\Lambda_2 < 0$) with present exchange calculations for $v = 24.25$, and there are possible inconsistencies for $v \leq 24.0$, since the χ_0 data might imply a $\Lambda_2 < 0$ and there might be more than one phase transition at $v = 23.8$. The analysis by DG of their own experiment requires either Λ 's essentially the same as set III of Table I or both Λ_1 and Λ_2 negative for all v .

While other experiments would also give valuable information, the above discussion emphasizes, in particular, the importance of the following: (a) new measurements of χ_0 as a function of T which would lead to a better determination of c and, therefore, of the Λ 's and γ 's; (b) measurements of T_c as a function of v to verify and extend the results of Halperin *et al.* and to ascertain whether more than one phase transition occurs for $H_0=0$ at $v=23.8$. These measurements in combination with the χ_0 and the p data would allow a decision between the + and - signs in Eqs. (4); (c) measurements of p as a function of T and H_0 for more

v would yield the v dependence of b for $v \geq 24.0$ and thus check the consistence of the Λ 's and α 's for $v \leq 24.0$ and those at $v=24.25$; (d) an improvement in the specific-heat data is needed to clarify the situation with respect to the Dundon and Goodkind experiment for $T > 5$ mK.

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²²The uncertainty in c is shown by two different data reductions. If the data are plotted as χ_0^{-1} vs T and as $\chi_0 T$ vs T^{-1} and c obtained from the T intercept and initial slope, respectively, values of c differing by factors of 2 or 3 are found.

²³One has to keep in mind that for the pressure data one is allowed to shift the p scale since the quantity actually measured is the change in bcc ^3He pressure, not p , as a function of T . We perform such shifts if our fit is improved.

²⁴While our most detailed work was done at $v=23.34$, 23.88, 24.0, and 24.25, we did check the consistency of all available data among themselves and with the Hamiltonian (1) whenever possible and found all data to be compatible with what is described in the body of the paper.

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²⁷To achieve compatibility among Λ 's at $v=24.0$ and 24.25, as a function of v the nondominant Λ has to change sign, while the dominant Λ decreases in absolute value. That this may be so is argued by looking at $\alpha_1 = (a - b\Lambda_2)/(\Lambda_1 - \Lambda_2)$ and $\alpha_2 = 4(a - b\Lambda_1)/3(\Lambda_2 - \Lambda_1)$. If, for increasing v , a continues its moderate increase while $|b|$ increases more rapidly, first the α for the nondominant Λ , then the α for the dominant Λ , will change sign. This will cause the desired behavior of the Λ 's.

²⁸Dr. A. K. McMahan has kindly provided us with unpublished entropy data of Halperin *et al.* in the range 2

$< T < 8$ mK at $\nu = 24.25$. Analysis of these data with the high-temperature expansion of the entropy leads to a value of $d \cong 1$ (mK)² which is considerably larger than one finds by extrapolation from lower values of ν . This value of d together with $T_c = 1.17$ mK implies values of Λ_1 and Λ_2 (viz., $\Lambda_1 = -0.93$ mK and $\Lambda_2 = -0.42$ mK or $\Lambda_1 = -0.9$ mK and $\Lambda_2 = -0.5$ mK) that can still be reconciled with the sets I-IV of Table I, albeit with some

difficulty. However, it is not clear to what extent convergence difficulties of the high-temperature series and the location on the melting curve are responsible for the large value of d in this case.

²³In fact, the greatest deviations from DG occur at low temperature where the high-temperature series does not seem to converge well.