Solid ^{3}He magnetism: A review of experiments*

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The experimental data on the solid ^{3}He magnetic system are reviewed with an eye toward establishing the constraints placed by the data on models of the system. We find that certain of the data are consistent with no conventional-model magnet and that the present models (Heisenberg near-neighbor antiferromagnet and triple exchange) are inconsistent with the data.

I. INTRODUCTION

Until recently the magnetic properties of solid ^{3}He were believed to be described by a Heisenberg near-neighbor antiferromagnet (HNNA) model.[1,2] But the high external field experiment of Kirk and Adams[3] and the failure of the magnetic-ordering transition to occur at the anticipated temperature[4] have cast doubt on the applicability of a HNNA model. The purpose of this note is to review the data and discuss the constraints placed by it on models for the magnetic properties of solid ^{3}He.

The large body of NMR and thermodynamic data on solid ^{3}He is reviewed by Guyer, Richardson, and Zane[1] (GRZ) and by Trickey, Kirk, and Adams[2] (TKA). These data are supplemented by the recent specific-heat experiment of Castles and Adams[5] and by recent intensive work along the melting curve.[6] Generally for a given magnetic Hamiltonian the calculation of NMR quantities T_1, T_2, $D_z, \ldots$ is relatively complex and involves one or more approximations. For this reason we do not regard NMR data as providing a primary test of a magnetic Hamiltonian. Thus we discuss only thermodynamic data below.

II. HIGH-TEMPERATURE THERMODYNAMIC DATA

Thermodynamic data exist on solid ^{3}He over most of the bcc phase ($30 \leqslant P \leqslant 100$ atm) in many cases down to reasonably low temperatures and in a few cases in finite magnetic field.[2] One has careful measurements of (i) the specific heat $C(T, 0)$, (ii) the magnetic pressure in zero external field $P(T, 0)$, (iii) the magnetic pressure in finite external field $P(T,H)$, (iv) the magnetic susceptibility $\chi(T)$, and (v) indirect evidence about the location of the magnetic phase transition.

To discuss these data we consider the predictions for $C(T, 0)$, $P(T, 0)$, $P(T,H)$, and $\chi(T)$ that follow from a general magnetic Hamiltonian at high temperatures. For this general magnetic Hamiltonian we take

$$\mathcal{H} = \mathcal{H}_Z + \mathcal{H}_2 + \mathcal{H}_3 + \mathcal{H}_4 + \cdots , \tag{1}$$

where

$$\mathcal{H}_2 = -2 \sum_{i<j} \Lambda(ij)_{\alpha\beta}\sigma_i^\alpha \sigma_j^\beta ,$$

$$\mathcal{H}_3 = -2 \sum_{i<j<k} \Lambda(ijk)_{\alpha\beta\gamma}\sigma_i^\alpha \sigma_j^\beta \sigma_k^\gamma , \quad \text{etc.,}$$

$$\mathcal{H}_Z = -2\mu H_0 \sum_i \sigma_i^z .$$

H_0 is the external magnetic field and σ_i^α is the α Cartesian component of $\vec{\sigma}_i$, $\sigma^z = \pm \frac{1}{2}$. We write Eq. (1) in the form

$$\mathcal{H} = \mathcal{H}_S + \mathcal{H}_Z . \tag{2}$$

The results of a high-temperature expansion calculation of $C(T, 0)$, $P(T, 0)$, $\ldots$ are shown in Table I. These results depend only upon counting spins ($\langle \mathcal{H}_n \mathcal{H}_Z \rangle = 0$, $n > 1$) and knowing the external field and volume dependence of each term in $\mathcal{H}_S$ ($\partial \mathcal{H}_S / \partial H_0 = \partial \mathcal{H}_Z / \partial V = 0$). In Table I and here we use the notation

$$\langle A \rangle = \mathrm{Tr}_{(\mathrm{spin})} A . \tag{3}$$

In Table I we also show the results of the specialization of $\mathcal{H}_S$ to a HNNA model and to Zane's triple-exchange model[7] ($X3$ model). For the HNNA model we take $\mathcal{H}_n = 0$ for $n > 2$:

$$\mathcal{H}_2 \equiv \mathcal{H}_A = -2 \sum_{i<j}^{nn} J_1(ij) \vec{\sigma}_i \cdot \vec{\sigma}_j , \tag{4}$$

with $J_1 < 0$.

For the $X3$ model we take $\mathcal{H}_n = 0$ for $n > 2$:

$$\mathcal{H}_2 \equiv \mathcal{H}_B = \mathcal{H}_A - 2 \sum_{i<j}^{nnn} J_2(ij) \vec{\sigma}_i \cdot \vec{\sigma}_j , \tag{5}$$

with $J_2 > 0$.

TABLE I. Results of a high-temperature expansion.

Quantity	General Hamiltonian [Eq. (1)]	$\mathcal{H}_A$ (HNNA) [Eq. (4)] bcc lattice	$\mathcal{H}_B$ ($X3$) [Eq. (5)] bcc lattice
$C(T,H=0)$	$k_B\beta^2[\langle\mathcal{H}_S^2\rangle+O(\beta)]$	$3k_B\beta^2J_1^2$	$3k_B\beta^2(J_1^2+\frac{3}{4}J_2^2)$
$P(T,H=0)$	$\frac{\beta}{2}\frac{\partial}{\partial V}[\langle\mathcal{H}_S^2\rangle+O(\beta)]$	$\frac{3}{2}k_B\beta\frac{\partial J_1^2}{\partial V}$	$\frac{3}{2}k_B\beta\frac{\partial}{\partial V}(J_1^2+\frac{3}{4}J_2^2)$
$\chi(T)_{H\to 0}$	$\mu^2[1-\beta k_B\Theta_N+O(\beta^2)]$	$\mu^2(1-4\beta J_1)$	$\mu^2[1-4\beta(J_1+\frac{3}{4}J_2)]$
$P(T,H)-P(T,0)$	$-y^2\frac{N}{2}\frac{\partial}{\partial V}k_B\Theta_N$	$-2y^2N\frac{\partial J_1}{\partial V}$	$-2y^2N\frac{\partial(J_1+\frac{3}{4}J_2)}{\partial V}$
Remarks	$y=\beta\mu H$ $k_B\Theta_N=\langle\mathcal{H}_2S_Z{}^2\rangle/\langle S_Z^2\rangle$	$k_B\Theta_N=4J_1$	$k_B\Theta_N=4J_1+3J_2$

From Table I we see results whose essential features are displayed in Table II. Succinctly these are (i) a high-temperature specific-heat experiment in which $C(T,0)\propto T^{-2}$ determines $\langle\mathcal{H}_S^2\rangle$, i.e., *the square of the entire magnetic Hamiltonian;* (ii) a high-temperature, zero-field, magnetic-pressure experiment in which $P(T,0)\propto T^{-1}$ determines $(\partial/\partial V)\langle\mathcal{H}_S^2\rangle$; (iii) a high-temperature susceptibility measurement in which $\chi(T)^{-1}\propto T$ determines the Curie-Weiss constant,

$$k_B\Theta_N\equiv\frac{\langle\mathcal{H}_S S_Z{}^2\rangle}{\langle S_Z{}^2\rangle}=\frac{\langle\mathcal{H}_2 S_Z{}^2\rangle}{\langle S_Z{}^2\rangle},$$

$$(S_Z=\sum_i\sigma_i{}^z),$$

i.e., *the second ferromagnetic moment of* $\mathcal{H}_2$; (iv) a high-temperature, high-field, magnetic-pressure experiment in which $P(T,H)-P(T,0)\propto\beta^2H^2$ determines $(\partial/\partial V)(k_B\Theta_N)$.

In Table II we have also listed the experiments in which these quantities have been measured. In principle, there is no simple relation between experiments of type I (1 and 2) that measure $\langle\mathcal{H}_S^2\rangle$ and experiments of type II (3 and 4) that measure $\langle\mathcal{H}_2S_Z{}^2\rangle/\langle S_Z{}^2\rangle$. For either $\mathcal{H}_S=\mathcal{H}_A$ or $\mathcal{H}_S=\mathcal{H}_B$ all four experiments are simply related (see Tables I and II).

TABLE II. Quantities measured in various experiments.

Thermodynamic quantity	Determines	Experiment (Ref.)
1. $C(T,0)$	$\langle\mathcal{H}_S^2\rangle$	5
2. $P(T,0)$	$\frac{\partial}{\partial V}\langle\mathcal{H}_S^2\rangle$	9
3. $\chi(T)_{H\to 0}$	$\frac{\langle\mathcal{H}_2S_Z^2\rangle}{\langle S_Z^2\rangle}$	10, 13
4. $P(T,H)-P(T,0)$	$\frac{\partial}{\partial V}\frac{\langle\mathcal{H}_2S_Z^2\rangle}{\langle S_Z^2\rangle}$	3, 8

The following empirical observations can be made about the experiments listed in Table II.

(a) Panczyk and Adams[9] find $(\partial/\partial V)\langle\mathcal{H}_S^2\rangle$ as a function of V for several V. They fit this quantity to the HNNA model and find J_P defined by

$$\frac{1}{3}\frac{\partial}{\partial V}\langle\mathcal{H}_S^2\rangle=2J_P(V)^2\frac{d[\ln J_P(V)]}{d(\ln V)}. \quad (6)$$

(b) Castles and Adams find $\langle\mathcal{H}_S^2\rangle$ at two values of V. Furthermore they find

$$\langle\mathcal{H}_S^2\rangle=3J_P(V)^2 \quad (7)$$

to better than 10%. Thus zero-field, magnetic-pressure, and specific-heat measurements are in good agreement with one another. They verify the relation $\langle\mathcal{H}_S^2\rangle=3J_P(V)^2$.

(c) A large number of susceptibility measurements have determined $k_B\Theta_N$ throughout the bcc phase.[10] These measurements (often subject to substantial uncertainty) are consistent with[11]

$$k_B\Theta_N^{(X)}=-4J_P. \quad (8)$$

Furthermore this agreement at all volumes means that we have

$$\frac{d[\ln|\Theta_N^{(X)}(V)|]}{d(\ln V)}=\frac{d[\ln J_P(V)]}{d(\ln V)}. \quad (9)$$

(d) Kirk and Adams[3] measured the magnetic pressure in finite external field and found $P(T,H)$ in disagreement with the HNNA model [$\mathcal{H}_S$ given by $\mathcal{H}_A$ in Eq. (4)] with the value of J_1 taken to be $-J_P$ from Panczyk and Adams.

Zane[7] showed that a pair Hamiltonian slightly more complex than the HNNA model could account for the observations of Kirk and Adams. The choice $\mathcal{H}_S=\mathcal{H}_B$ with $J_2>0$ can explain the observations of Kirk and Adams and not change in an essential way the consistency of specific-heat and zero-field pressure measurements. Zane supported his suggestion with a microscopic model.

Zane's suggestion made it possible to resolve a

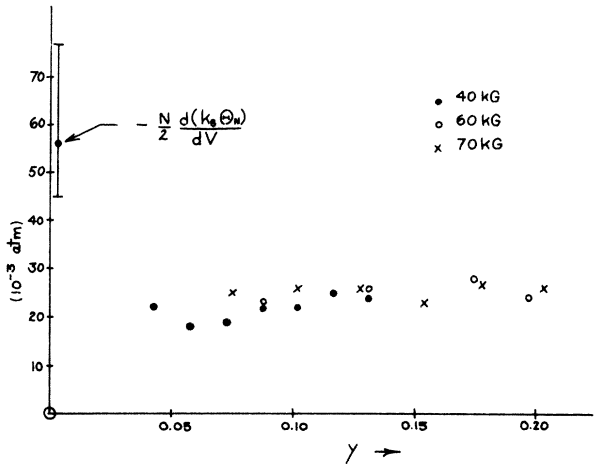

FIG. 1. Data of Kirk and Adams at 23.34 cm^3/mole are plotted as in Eq. (10). The data at three values of the external field fall on a universal curve that is approximately horizontal. Thus the pressure is quadratic in the field at least for $y < 0.20$. The intercept of the horizontal line with the vertical axis has a value of $-\frac{1}{2}N \times (\partial/\partial V) k_B\Theta_N$ determined by this experiment. The direct measurement of $-\frac{1}{2}N(\partial/\partial V)k_B\Theta_N$ by Kirk, Osgood, and Garber and by Bernat and Cohen is shown on the vertical axis at 56 with the experimental uncertainty indicated.

basic inconsistency between an experiment of type II and experiments of type I. But the two experiments of type II are not consistent with one another.

(e) The high-field, magnetic-pressure data of Kirk and Adams can be viewed in the form

$$\lim_{y\to 0}\frac{P(T,H)-P(T,0)}{y^2}=-\frac{N}{2}\frac{\partial(k_B\Theta_N)}{\partial V}, \tag{10}$$

where $y=\beta\mu H_0$. For the data at $V=23.34$ cm^3/mole one finds the result shown in Fig. 1. Data for three values of the external field ($H=40$, 60, 70 kG) lie on a universal curve so that the $y\to 0$ limit is valid and represented by the raw data. The resulting value of $-\frac{1}{2}N(\partial k_B\Theta_N/\partial V)$ is given in Table III as well as that at $V=24.0$ cm^3/mole. These numbers are to be compared with the direct measurement of $-\frac{1}{2}N(\partial k_B\Theta_N/\partial V)$ by Kirk, Osgood, and Garber[12] and by Bernat and Cohen[13] reported in Table III. Recall these susceptibility measurements are made in low fields and correspond to $y\to 0$. The difference of a factor greater than 2 is beyond the limits of the experimental uncertainties. Thus two independent measurements of $(\partial/\partial V) \times (k_B\Theta_N)$ are in disagreement with one another.

This observation is independent of the details of the magnetic Hamiltonian.

TABLE III. Result of measurements of $N\left(\frac{1}{2}\right)\frac{\partial(k_B\Theta_N)}{\partial V}$

Volume (cm^3/mole)	$\frac{P(T,H)-P(T,0)}{y^2}$ (10^{-3} atm)	Direct measurement (10^{-3} atm)
24.0	35 ± 15	104^{+28}_{-11}
23.34	20 ± 10	56^{+21}_{-11}

III. MAGNETIC PHASE TRANSITION

Consider a magnetic system described by a general two-spin Heisenberg Hamiltonian

$$N=-2\sum_{i<j}\Lambda(ij)\vec{\sigma}_i\cdot\vec{\sigma}_j\,. \tag{11}$$

Let us look at the magnetic phase transition in mean-field theory. To do this we consider the inequality

$$\hat{\mathcal{G}}=\hat{F}+\hat{\mathcal{H}}_H-\hat{\mathcal{H}}\geq F_H\,, \tag{12}$$

where F_H is the true free energy of $\mathcal{H}_H$, $\hat{F}$ and $\hat{\mathcal{H}}$ are the free energy and energy, respectively, of a trial Hamiltonian

$$\hat{\mathcal{H}}=-2\mu\sum_i\sigma_i^z h_i\,, \tag{13}$$

and

$$\hat{\mathcal{H}}_H=\frac{\mathrm{Tr}\,e^{-\beta\hat{\mathcal{H}}}\mathcal{H}_H}{\mathrm{Tr}\,e^{-\beta\hat{\mathcal{H}}}}\,. \tag{14}$$

We calculate $\hat{\mathcal{G}}$ with $\mathcal{H}_H$ and $\hat{\mathcal{H}}$ given by Eqs. (11) and (13) and vary the result with respect to h_i. We find

$$\mu h_i=\sum_j\Lambda(ij)\hat{\sigma}_j\,, \tag{15}$$

and

$$\hat{\sigma}_i=\tfrac{1}{2}\tanh\sum_j\beta\Lambda(ij)\hat{\sigma}_j\,. \tag{16}$$

From Eq. (16) we find that the Fourier component $\hat{\sigma}(\vec{q})$ of the spin density has a critical point at

$$k_BT_C(\vec{q})=\tfrac{1}{2}\Lambda(\vec{q})\,, \tag{17}$$

where

$$\Lambda(\vec{q})=\sum_i e^{i\vec{q}\cdot\vec{R}_i}\Lambda(\vec{R}_i)\,.$$

We associate the magnetic system critical point with $k_BT_C(\vec{q}_C)$, where $\vec{q}_C$ is that value of $\vec{q}$ for which $\Lambda(\vec{q})$ is a maximum;

$$k_BT_C=\tfrac{1}{2}\Lambda(\vec{q}_C)\,. \tag{18}$$

At the same time the Curie-Weiss temperature $k_B\Theta_N$ is simply the $q=0$ component of Λ,

$$k_B\Theta_N = \frac{\langle \mathcal{H}_H S_Z{}^2\rangle}{\langle S_Z{}^2\rangle} = \tfrac{1}{2}\Lambda(0)\,. \tag{19}$$

Since $\Lambda = 0$ $(\vec{R} = 0)$ we have

$$\frac{1}{N}\sum_q \Lambda(\vec{q}) = 0\,. \tag{20}$$

From Eq. (11) we have

$$\langle \mathcal{H}_S^2\rangle = \frac{3}{8}\,\frac{1}{N}\sum_q \Lambda(\vec{q})^2\,. \tag{21}$$

Combined with the data on solid ^{3}He, Eqs. (18)–(21) are four separate constraints on the form of a two-spin model [as in Eq. (11)] for solid ^{3}He. From the present data we have

$$\tfrac{1}{2}\Lambda(0) = 4J_P\,, \tag{22}$$

$$\frac{1}{N}\sum_q \Lambda(\vec{q})^2 = 8J_P^2\,, \tag{23}$$

$$\frac{1}{N}\sum_q \Lambda(\vec{q}) = 0\,, \tag{24}$$

$$\Lambda(\vec{q}_C) \lesssim \tfrac{1}{2}|\Lambda(0)|\,. \tag{25}$$

Equations (22) and (23) follow from the discussion above; Eq. (24) is trivial. Equation (25) follows from the failure of recent efforts to observe the magnetic phase transition down to temperatures of order 1 mK. It is a conservative statement of this failure.

A HNNA model yields $\Lambda(\vec{q}_C) = |\Lambda(0)|$ and is inconsistent with the data. Zane's $X3$ model yields $\Lambda(\vec{q}_C) > |\Lambda(0)|$ and is also inconsistent with the data. We are able to conclude that the HNNA and $X3$ model are ruled out by the experimental constraints. One has enormous freedom to choose magnetic Hamiltonians that are consistent with the four constraints.

IV. CONCLUSION

We have examined the existing thermodynamic data on solid ^{3}He to learn what constraints it places on model Hamiltonians with which one might attempt to describe the system. We find that the data of Kirk and Adams on the finite-field magnetic pressure is inconsistent with the susceptibility data that yields $k_B\Theta_N$ within the context of the Hamiltonian in Eq. (1). Thus we must conclude either that the data of Kirk and Adams and the susceptibility data should be viewed with skepticism or that no magnetic Hamiltonian of the form in Eq. (1) will correctly describe solid ^{3}He magnetism. In the case that the explanation of the inconsistency is looked for in the data, the good agreement of the Kirk, Osgood, and Garber experiment with the Bernat and Cohen experiment suggests that it is the Kirk and Adam's experiment that is in doubt. But this is by no means the only possibility. We urge that both the Kirk and Adam's measurements and susceptibility measurements be repeated. If the explanation of the inconsistency is in the theory, it appears that there is nothing to be added to the magnetic Hamiltonian that will supply the answer. An alternate to the conventional approach to the statistical mechanics of a compressible magnet will be necessary.

We also find that the near-neighbor Heisenberg antiferromagnet and the triple-exchange model of Zane are inconsistent with the data regardless of the resolution of the experimental difficulty discussed above.

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