# COMMENTS AND ADDENDA

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## Nuclear spin ordering in solid $^3$He

J. D. Johnson*
*Theoretical Division, Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico 87544*

E. G. D. Cohen
*The Rockefeller University, New York, New York 10021*

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The recent experiments of Kummer *et al.*, as well as Collan and Halperin *et al.*, on the melting curve of $^3$He are analyzed with a localized Heisenberg Hamiltonian. They can be described consistently only with ferromagnetic nearest-neighbor and antiferromagnetic next-nearest-neighbor exchange constants that imply a positive Weiss constant, $\Theta = 1.04$ mK. These exchange constants can only with difficulty be made consistent with those derived previously with the same Hamiltonian from experiments at smaller molar volumes.

In a recent Letter[1] Kummer *et al.* reported a magnetic field–temperature phase diagram of solid $^3$He on the melting curve. We have fitted this phase diagram with a Heisenberg Hamiltonian

$$H/k_B = H_1 + H_2 \ , \tag{1a}$$

where

$$H_1 = -2\Lambda_1 \sum_{\rm nn} \vec{I}_i \cdot \vec{I}_j - 2\Lambda_2 \sum_{\rm nnn} \vec{I}_i \cdot \vec{I}_j \tag{1b}$$

and

$$H_2 = -\gamma H_0 \sum_{i=1}^{N} I_i^z \ . \tag{1c}$$

Here, $\gamma = 1.556 \times 10^{-7}$ °K/G, $I^\mu = \frac{1}{2}\sigma^\mu$ with $\sigma^\mu$ being Pauli matrices, $H_0$ is the external magnetic field, $T$ is the absolute temperature, and $k_B$ is Boltzmann's constant. $\sum_{\rm nn}$ ($\sum_{\rm nnn}$) is a sum over nearest-neighbor (next-nearest-neighbor) pairs of lattice sites $i,j$ in a bcc lattice. The fit could only be made with $\Lambda_1 > 0$ and $\Lambda_2 < 0$; i.e., ferromagnetic nn and antiferromagnetic nnn exchange constants. The resulting Weiss constant is positive. All other known data of $^3$He on the melting curve can be represented within the experimental accuracy by the same values of $\Lambda_1$ and $\Lambda_2$. In a previous publication[2] we have analyzed all data on solid $^3$He at smaller molar volumes ($21 \le v \le 24$ cm$^3$/mole) by the same Hamiltonian (1). There it was found that these data could be represented within their experimental uncertainties by four independent sets of $\Lambda_1$ and $\Lambda_2$ among which were the conventional one (set I: $\Lambda_1 < 0$, $\Lambda_2 > 0$) supported by exchange calculations and a set similar to the one quoted here (set III: $\Lambda_1 > 0$, $\Lambda_2 < 0$), although an entirely different representation of the data, not based on the Hamiltonian (1), could not be ruled out. In view of this unclear situation, it is important that the analysis to be presented here of the new data of Kummer *et al.* permits one to correlate all existing data on the melting curve of solid $^3$He consistently on the basis of the Hamiltonian (1). The values found for $\Lambda_1$ and $\Lambda_2$, which would point to an antiferromagnetic class-2 ($AF_2$) type of magnetic structure at least on the melting curve, are not inconsistent with those of set III of Ref. 2. However, to make them actually consistent in a reasonable manner presents such difficulties that either some experimental data are incorrect or have been interpreted incorrectly, or the Hamiltonian (1) cannot describe the magnetic behavior of solid $^3$He over the entire volume range that has been studied.

We now present further details concerning the above statements.

We shall use, wherever possible, results from high-temperature expansions for our theoretical

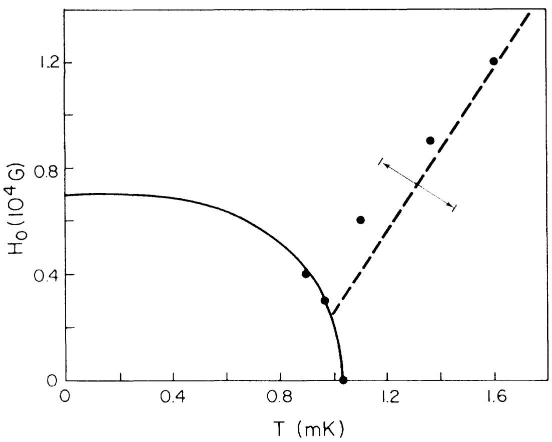


FIG. 1. $H_0$-$T$ phase diagram for solid $^3$He. The points are the experimental data of Ref. 1. The dashed line is computed on the basis of a high-temperature expansion using the Hamiltonian (1). The solid line is the phase boundary between a paramagnetic and an $AF_2$ spin-flop phase, computed on the basis of mean-field theory with the $T$ values of the phase boundary rescaled to give the correct zero-field transition temperature.

analysis of the data. However, with the exception of Eq. (2), derived by us, the only relevant high-temperature results available for the Hamiltonian (1) are for $H_0 = 0$. Therefore, we have supplemented the high-temperature results, where necessary, with results derived from mean-field theory. We emphasize that we use mean-field theory only for qualitative, not for quantitative predictions.

The data of Kummer *et al.* are shown in Fig. 1. We interpret the upper three points—as was suggested by Kummer *et al.*—as representing the peak in the specific heat in the paramagnetic phase and not as a phase transition. The lower points are taken to be on the phase boundary of a paramagnetic to spin-flop phase transition. As suggested by the experimental points, we take the upper part of the paramagnetic $C_v$-peak curve to be very close to a linear curve in $T$. It is not possible to use a high-temperature expansion to verify the quasilinear behavior since, owing to a lack of convergence of the series, this expansion predicts a linear behavior only down to about 4 mK; i.e., below 4 mK the high-temperature expansion neither confirms nor contradicts the assumption of quasilinearity. However, mean-field theory, which agrees very well with the high-temperature expansion above 4 mK, predicts a continuation of the linear behavior down to near the phase boundary, including the region of interest to us here.

The assumption of linearity being made, it remains to determine the slope and the intercept of the straight line. For this we can use the high-temperature expansion for the $C_v$ peak in its asymptotic region, where only two terms are needed. To compute these two terms, we make a high $T$, but arbitrary $H_0$, expansion of the $e^{-H/k_B T}$ where $e^{-H_1/T}$ was expanded in a power series in $1/T$ while $e^{-H_2/T}$ was not expanded. Then for large $T$ the position of the specific heat maximum is given in the $H_0$-$T$ plane by

$$\gamma H_0 = 2\lambda T + 2\alpha \quad . \qquad (2)$$

Here, $\lambda$, the slope of the trajectory in the $H_0$-$T$ plane, is given by the equation $\lambda \tanh\lambda = 1$, leading to $\lambda = 1.2$, and $\alpha$, related to the intercept with the $T$ axis is given by $\alpha = -2(3\lambda - 1/\lambda)\, c/\lambda^2 = -3.843c$, where the unknown constant $c = \Lambda_1 + \frac{3}{4}\Lambda_2$ (cf. Ref. 2). With the slope given by $\lambda$, a reasonable representation of the $C_v$-peak curve is given by the dashed line of Fig. 1, which has an intercept with the $T$ axis that yields a value for $c = 0.26$ mK. We estimate the error in $c$, due to experimental as well as theoretical uncertainties, to be about $\pm 0.06$ mK. The arrows in Fig. 1 indicate the shift of the dashed line consistent with this error.

In order to determine the phase boundary, we have to find $\Lambda_1$ and $\Lambda_2$. To do this, a second relation between $\Lambda_1$ and $\Lambda_2$, in addition to that given by $c$, must be found. This can be achieved in a variety of ways, all leading to equivalent results. The easiest way is perhaps to determine, what we have called before in Ref. 2, $d = \Lambda_1^2 + \frac{3}{4}\Lambda_2^2$ from recent experiments on the melting curve by Collan[3] and Halperin *et al.*[4,5] These experiments have yielded two different values of $d$. Collan found $d = 0.64$ mK$^2$, while a value of $d = 1.1$ mK$^2$ is obtained from the experiments of Halperin *et al.*[6] Both values of $d$ are consistent with those found previously at smaller volumes (cf. Ref. 2).

(a) We first discuss Collan's value of $d$. Using that solution of the equations for $\Lambda_1$ and $\Lambda_2$ which gives a correct zero-field phase transition temperature,[7] we find $\Lambda_1 = 0.656$ mK and $\Lambda_2 = -0.528$ mK. These values are consistent with other measured quantities on the melting curve. Thus, Collan determined $e = \Lambda_1^3 + \frac{3}{4}\Lambda_2^3 - 9\Lambda_1^2\Lambda_2 = 2.65 \pm 0.6$ mK$^3$ while we find $e = 2.22$ mK$^3$ with the above values of $\Lambda_1$ and $\Lambda_2$, which is well within the experimental error. We shall now calculate the entire $H_0$-$T$ phase boundary, using high-temperature results where possible. From the high-temperature work of Pirnie *et al.*,[7] we obtain the zero-field transition temperature $T_c = 1.03$ mK, which agrees very well with that found experimentally. Furthermore, with spin-wave theory we obtain the critical field at $T = 0$, $H_c = 7000$ G. Since there are no high-temperature results available to determine the phase boundary for $H_0 \neq 0$, we use mean-field theory to calculate an approximate phase boundary between $H_c$ and $T_c$. Now mean-field theory gives a phase boundary between an $AF_2$ spin-flop phase[8] and a paramagnetic phase specified by

$$t=\frac{2h}{\ln[1+2h)/(1-2h)]}\,, \qquad (3)$$

where $t=-T/6\Lambda_2$ and $h=-\gamma H_0/(16\Lambda_1+24\Lambda_2)$ for $\Lambda_2<0$ and $\Lambda_2/\Lambda_1<-\frac{2}{3}$. We cannot use Eq. (3) directly to obtain the phase boundary since (3) gives a (mean-field theory) zero field $T_c=1.58$ mK instead of $T_c=1.03$ mK. Thus we rescale the mean-field $T$ axis with the high-temperature value $T_c=1.03$ mK, such that the mean field $T_c$ from Eq. (3) agrees with the high-temperature value. A similar scale change on the $H_0$ axis is not necessary since mean-field theory gives the correct value for the critical field $H_c$. The resulting phase boundary is given by the solid curve in Fig. 1 and can be expected to be a reasonable representation of the actual phase boundary. We stress that the only feature from mean-field theory that is actually needed to supplement the high-temperature results in our comparison with experiment is that the behavior of the lower part of the phase boundary near $T_c$ is very steep. The agreement with experiment is striking but perhaps misleading since there are certainly 4% uncertainties in the quoted experimental temperatures while the $T_c$ value given by the high-temperature expansions is at least that uncertain.

(b) The second value of $d$ leads with $c=0.26$ mK to $\Lambda_1=0.823$ mK and $\Lambda_2=-0.751$ mK. We now find $e=4.82$ mK$^3$ and $H_c=1.56\times10^4$ G. Although the value of $e$ is much larger than the experimental value $e\cong 2$ mK$^3$, it is not necessarily inconsistent with the experimental value due to the large uncertainty in $e$.[9] The phase boundary between the $AF_2$ spin-flop phase and the paramagnetic phase has now been moved upwards so that each $H_0$ value is about twice as large as in Fig. 1. However, the temperature uncertainties quoted above still allow the phase boundary to be consistent with the experimental data, although the agreement is clearly not as good as under (a).

We now turn to the question of the consistency of the $\Lambda$'s and related quantities on the melting curve (molar volume $v=24.25$ cm$^3$/mole) with those determined previously for smaller volumes. The only set of $\Lambda$'s that need be considered is the set III of Ref. 2 with $\Lambda_1>0$ and $\Lambda_2<0$, $AF_2$ ordered structure, and negative Weiss constant. There $\Lambda_1=0.094$, 0.16, and 0.18 mK and $\Lambda_2=-0.48$, $-0.73$, and $-0.77$ mK for $v=23.34$, 23.88, and 24.0 cm$^3$/mole, respectively.[10] The $\Lambda$'s were determined from four parameters, $a$, $b$, $c$, and $d$, that were directly deduced from experiment: $a=\Lambda_1^2\gamma_1+\frac{3}{4}\Lambda_2^2\gamma_2$, $b=\Lambda_1\gamma_1+\frac{3}{4}\Lambda_2\gamma_2$, and $c$ and $d$ have been defined above. Here $\gamma_i=d(\ln|\Lambda_i|)/d(\ln v)$, $d(d)/d(\ln v)=2a$ and $d(c)/d(\ln v)=b$. The values of $a$ and $d$ at smaller volumes are consistent with those at $v=24.25$. Also, the values of $e$ given above are consistent with those found by Dundon and Goodkind at lower molar volumes.[11]

There are difficulties, however, with the $b$'s and $c$'s. The $b$'s have only been determined from pressure measurements by Kirk and Adams[12] over a limited range of volumes, viz., $23.34\le v\le 24.0$, and found to be negative and decreasing: $b=-3.6$, $-3.8$, and $-5.7$ mK for $v=23.34$, 23.88, and 24.0 mK, respectively. The $c$'s, as deduced by Kirk *et al.*[13] from a susceptibility experiment over the range $21\le v\le 24$, are negative.[14] Since for $v=24.25$, $c=0.26$ mK, $c$ must make a very considerable increase and $b$ must become large and positive—$b\cong 30$ mK or larger—over a relatively small volume range in order to allow a change from a negative value at $v=24$ to $c=0.26$ mK at $v=24.25$. (Since $c$ is directly related to the paramagnetic Weiss constant, $\Theta=4c$, a change of sign in $\Theta$ should occur in the interval $24<v<24.25$.) Although such large increases in $b$ and $c$ in the interval $24<v<24.25$ cannot be ruled out *a priori*, they do not seem too likely.[15] If these difficulties with $b$ and $c$ are taken seriously, one is inclined to doubt the possibility of describing the magnetic properties of solid $^3$He over the entire volume range on the basis of the Hamiltonian (1).

Alternatively, one could doubt the accuracy of the $c$ values and, to a lesser extent, of the $b$ values for $v\le 24$ and stress the consistency of the other quantities $a$, $d$, and $e$. Then one could still hope that the behavior of solid $^3$He can be described consistently by the Hamiltonian (1) with one, albeit unconventional, set of $\Lambda$'s: $\Lambda_1>0$ and $\Lambda_2<0$ for at least $23.34\le v\le 24.25$ cm$^3$/mole. Even if one takes this point of view, it is unclear to what extent the Hamiltonian (1) is not merely an effective Hamiltonian, and the $AF_2$ spin-flop structure is actually the structure of solid $^3$He at low $T$. Clearly more experiments, especially pressure measurements in the region $v\ge 24$ and susceptibility measurements for all $v$, would be helpful to clarify further the present situation.

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[6]We are indebted to Dr. A. K. McMahan for furnishing us with an unpublished value of $d$ obtained from the experimental work of Halperin *et al.* Halperin *et al.* in Ref. 5 give the same value for $d$.

[7]K. Pirnie, P. J. Wood, and J. Eve, Mol. Phys. 11, 551 (1966). We use the high-temperature work of Pirnie *et al.* to connect $\Lambda_1$ and $\Lambda_2$ to $T_c$. For the values of $\Lambda_1$ and $\Lambda_2$ on the melting curve, it is difficult to deduce reliable values of $T_c$ from their graph; therefore, the estimated uncertainty in $T_c$ is easily 15 to 20%.

[8]Within mean-field theory and on a bcc lattice, an $AF_2$ spin-flop system in zero field consists of two interpenetrating cubic sublattices which independently exhibit a simple antiferromagnetic order. See, for instance, J. S. Smart, *Effective Field Theories of Magnetism* (Saunders, Philadelphia, 1966). If $H_0 \neq 0$, one has a simple spin-flop system on each sublattice with interaction between sublattices via their net magnetization.

[9]We feel that the uncertainty on $e$ is probably larger than the $\pm 0.6$ $mK^3$ quoted earlier since $e$ is found by fitting low-temperature data with a truncated high-temperature expansion.

[10]These values of $\Lambda_1$ and $\Lambda_2$ are offered as a typical example of a set of $\Lambda$'s. Because there are large uncertainties in the measured values of $c$ for these molar volumes, one can easily obtain a range of values for $\Lambda_1$ and $\Lambda_2$, all equally consistent with the existing data.

[11]J. M. Dundon and J. M. Goodkind, Phys. Rev. Lett. 32, 1343 (1974). As explained in a previous paper (see Ref. 2), the precise significance of the Dundon and Goodkind experiment is not clear to us. Besides $e$ quoted above, they find $d$'s larger than those of other experiments and a possible anomolous contribution to the specific heat.

[12]W. P. Kirk and E. D. Adams, Phys. Rev. Lett. 27, 392 (1971).

[13]W. P. Kirk, E. B. Osgood, and M. Garber, Phys. Rev. Lett. 23, 833 (1969).

[14]We stress that there are, in the $c$ values obtained from the susceptibility experiment of Kirk *et al.*, large uncertainties. For instance, apart from the values of $c$ found by Kirk *et al.*, a different, but equally valid, analysis gives $c$'s different by factors of 2 or 3.

[15]It should be remarked that, although the experiments of Dundon and Goodkind, Halperin *et al.*, and Kummer *et al.* can be described to varying degrees by (the bulk) Hamiltonain (1) with an unconventional set of $\Lambda$'s, we cannot rule out the possibility that important surface effects are involved in these experiments, as has already been suggested by Kummer *et al.*