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## THERMODYNAMIC STUDY OF MAGNETICALLY ORDERED SOLID $^3\text{He}$

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**Résumé.** — Les résultats expérimentaux de Kummer, Mueller et Adams sur les pressions de liquéfaction de  $^3\text{He}$  dans des champs magnétiques, sont analysés de façon à obtenir des informations sur la susceptibilité. Au-dessus de la température d'ordre, la susceptibilité est supérieure à la valeur prévue avec la constante de Curie-Weiss par un facteur supérieur à 2. A bas champ, la susceptibilité décroît d'un facteur environ 3 dans la phase ordonnée. Au contraire, à haut champ  $B \geq 0,42$  T, la susceptibilité s'accroît brusquement, indiquant la possibilité d'une transition du second ordre.

**Abstract.** — The data of Kummer, Mueller and Adams on  $^3\text{He}$  melting pressures in applied fields are analysed to provide information on the susceptibility. Above the ordering temperature the susceptibility is greater than the Curie-Weiss value by more than a factor of two. For low fields, a decrease in susceptibility by about a factor of 3 occurs in the ordered phase. However, at high fields,  $B \geq 0.42$  T, the susceptibility increases abruptly on ordering, indicating the possibility of a second-order transition.

A magnetic phase transition has been observed in solid  $^3\text{He}$  near 1 mK [1-3], which for low fields is very abrupt, possibly first order. Kummer, Mueller and Adams (KMA) have reported measurements of the latent heat of melting in applied magnetic fields [2]. From these measurements, they find that the transition temperature decreases slightly with field for  $B \leq 0.42$  T. For higher fields the ordering region broadened and increased in temperature with increasing magnetic field. KMA suggested that the ordering for  $B \geq 0.42$  T might not be a phase transition but alignment of the spins by the field in the paramagnetic phase.

The purpose of this paper is to report new conclusions deduced from a thermodynamic analysis of the KMA data. The thermodynamic analysis to follow uses pressure data in different fields at the same temperature. Thus to make use of the high resolution in the pressure, the precision of the temperatures must be examined. Temperatures were obtained by KMA from the thermodynamic relation [1]

$$T = T_0 \int_{P_0}^P \frac{\Delta V}{\Delta Q} dP, \quad (1)$$

where  $(P_0, T_0)$  is a suitable reference point and  $\Delta V$  is the volume change in the Pomeranchuk cell when heat  $\Delta Q$  is added at constant temperature.

Using the data of Halperin *et al.* [1], KMA picked  $(P_0, T_0)$  near 17 mK for  $B = 0$ . Because of the lack of any detectable dependence of  $\Delta V/\Delta Q$  on the field at higher temperatures,  $(P_0, T_0)$  was taken near 8 mK, independent of field for  $B \neq 0$ . This procedure provided a severe test of the data and gave absolute temperatures with an estimated accuracy of 8%. Because of the depression of the melting temperature by the field for a given pressure, the temperatures of KMA in a field would be somewhat high through the error in  $T_0$ . A more accurate temperature scale is possible if a lower  $T_0$  with a known field dependence is available. Such a reference point is provided by the A transition in liquid  $^3\text{He}$ . Osheroff and Anderson [4] found that the variation of  $T_{A1}$  and  $T_{A2}$  with field is such that

$$0.375 T_{A1} + 0.625 T_{A2} = T_A(B = 0) \quad (2)$$

independent of field. Osheroff and Anderson found the variation of pressure with field at  $T_A$  (as defined by eq. (2)) to be

$$\delta P = P(T_A, 0) - P(T_A, B) = 530 B^2 T^{-2} \text{ Pa}. \quad (3)$$

The data of KMA give  $\delta P = 564 B^2$  for this quantity. Thus the pressure data of KMA have sufficient precision to permit refinement of the temperature scale by using  $T_A$  of equation (2) for the fixed point. As can be seen from equation (1), all that is necessary to make this correction is to multiply the temperatures of KMA for each field by  $T_A/T_{A,KMA}(B)$ , where  $T_{A,KMA}(B)$  is the value for  $T_A(B)$  reported by KMA. Taking  $T_A = 2.68$  mK gives a reduction in the temperature of KMA of 2, 3 and 7 % for the fields of 0.6, 0.9 and 1.2 T, respectively.

The present thermodynamic analysis begins with the equality of changes in the Gibbs free energy in the liquid and solid phases in equilibrium [5].

$$-s_S dT + v_S dP - M_S v_S dB = -s_L dT + v_L dP - M_L v_L dB. \quad (4)$$

Here the subscripts S and L refer to the solid and liquid, respectively, and the other symbols denote the usual standard thermodynamic variables. In this case, we are interested in the change in melting pressure with magnetic field at constant  $T$ , so  $dT = 0$ . Rearranging equation (4), integrating over  $B$ , and assuming  $M_L \ll M_S$  for  $T < 2.6$  mK, we have

$$\Delta P_m \equiv P_m(T, 0) - P_m(T, B) = \frac{1}{v_L - v_S} \int_0^B M_S v_S dB, \quad (5)$$

where  $P_m$  is the melting pressure.

In this analysis, we use the KMA data for  $\Delta P_m$  and the known values [1, 6] or  $v_S = 24.25$  cm<sup>3</sup>/mole and  $\Delta v = v_L - v_S = 1.314$  cm<sup>3</sup>/mole in equation (5)

to calculate  $\int_0^B M_S dB$ . Using the melting pressures of KMA, with temperatures revised as indicated above, we have computed the pressure differences  $\Delta P_m$  of equation (5) for  $T \leq 2.6$  mK. (Because of increasingly smaller pressure differences at higher temperatures, this procedure has insufficient accuracy for application at  $T > 2.6$  mK.) The results are plotted in figure 1 for various magnetic fields. For the three higher fields, arrows indicate the temperatures where a rapid (possibly discontinuous) increase of the specific heat is observed, which we discuss below. For  $B = 0.3$  and  $0.4$  T, the curves show a sharp decrease at  $T_c$ . (Since the quantity plotted involves  $\int_0^B M dB$ , the decrease at low fields is also reflected in the curves of higher fields.) For the three higher fields,  $\int M dB$  continues to increase to temperatures much below the abrupt increase of specific heat. This indicates a different type ordering in high fields than in low fields.

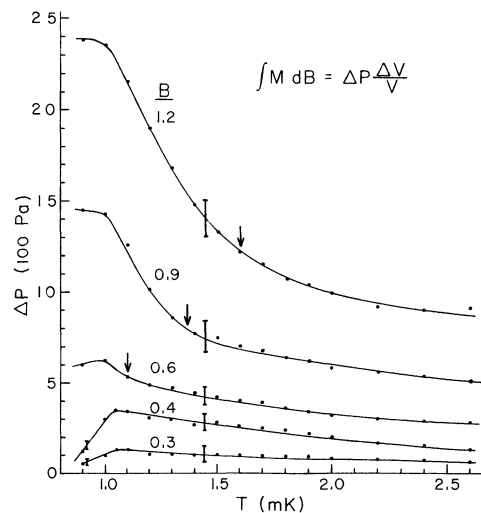


FIG. 1. — Melting pressure differences,  $P(T, 0) - P(T, B)$ , versus temperature for various magnetic fields. For the three higher fields, the arrows show the temperatures  $T(B)$  of the probable specific-heat discontinuity. An estimate of the experimental errors is given by the bars.

A more useful quantity to examine than  $\int M dB$  would be the magnetization  $M$  or the susceptibility,  $\chi = \partial M / \partial H$  (for convenience we now drop the subscript S). We note that if  $\chi$  were independent of  $H$ , then  $M = \chi H = \chi B / \mu$  and

$$\int M dB = \chi B^2 / (2 \mu) = \frac{\Delta v}{v} \Delta P_m, \quad (\text{we note } v_S = v, s_S = s). \quad (6)$$

Thus it is useful to examine a plot of  $\Delta P_m$  versus  $B^2$ , as shown in figure 2. If  $\chi$  were independent of  $H$ , the curve for a given temperature would be a straight line with slope proportional to  $\chi$ . In view of the phase transition near 1 mK and the phase diagram reported by KMA, it is not surprising that a linear relations is not found. Thus, instead of using the approximation of equation (6) with a constant  $\chi$ , we define three different susceptibilities which are functions of  $B$ : the differential susceptibility:

$$\chi = \mu \frac{\partial M}{\partial B} = - \frac{\Delta v}{v} \frac{d^2 P_m}{dB^2}, \quad (7)$$

the estimated susceptibility:

$$\chi_{B_1, B_2} = \mu \frac{2 \Delta v}{v} \frac{P_m(B_1) - P_m(B_2)}{B_2^2 - B_1^2}$$

which gives an estimate of the apparent susceptibility:

$$\frac{\chi^a}{\mu} = \frac{M}{B} = - \frac{2 \Delta v}{v} \frac{dP_m(B)}{dB^2} = \lim_{B_1 \rightarrow B} \chi_{B_1, B}. \quad (8)$$

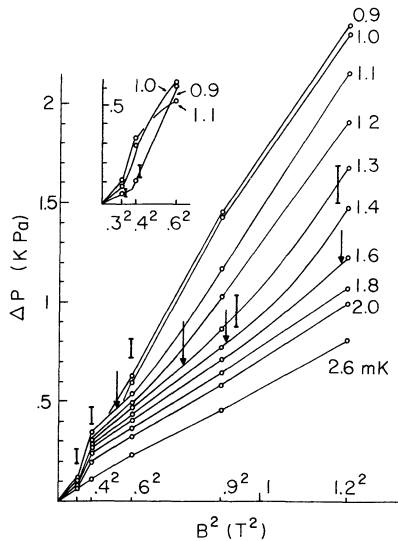


FIG. 2. — Melting pressure differences,  $P(T, 0) - P(T, B)$ , versus  $B^2$  for various temperatures. The inset gives the details of the region near the origin. The interpolating lines connecting the points have a discontinuity in their second derivative given by  $\Delta\chi$  at  $T(B)$  as shown by the arrows (see text). An estimate of the experimental errors is given by the bars.

The experimental values of  $P_m$  being given in figures 1 and 2 at discrete values of  $B$ , only  $\chi_{B_1, B_2}$  can be calculated. The first or second derivative ( $\chi^a$  or  $\chi$ ) need a large number of values of  $B$  in order to draw a continuous curve through the points.

The inverse susceptibility,  $\chi_{B_1, B_2}^{-1}$  obtained from  $\Delta P_m$  through equation (8) is shown in figure 3 for various values of  $B_1, B_2$ . The line marked C-W shows the value of  $\chi^{-1}$  given by a Curie-Weiss law  $\chi = C/(T - \theta)$  with  $\theta = -3$  mK. At  $T = 2.5$  mK the susceptibility for all fields is greater than the Curie-Weiss value by about a factor of 1.7. In normal antiferromagnets, the susceptibility falls below the Curie-Weiss value as the transition is approached. In figure 2 the bump around 0.4 T for  $T > 1.1$  mK is probably connected to experimental errors, so that no definite conclusion can be drawn from this bump; for this reason the values of  $B_1, B_2$  (0.3, 0.6) or (0.4, 0.3) have not been shown in figure 3. The errors on  $\chi_{0, 0.3}$  are quite large, except for  $T < 1.1$  mK where the uncertainties are smaller, especially at low fields where the pressure becomes nearly independent of temperature. This can be seen on the data curve in figure 4. As the temperature drops below  $T_c^0 = 1.05$  mK, the low-field susceptibility ( $B_1 = 0.3, B_2 = 0$ ) undergoes a rapid decrease to about 1/3 of its value just above  $T_c^0$ . The decrease in susceptibility at low fields indicates some type of antiferromagnetic ordering, although such a rapid decrease is unexpected for the usual antiferromagnet. The high-field behaviour of  $\chi$  is discussed later.

Several groups have recently made measurements of the susceptibility in the vicinity of the ordering temperature, in qualitative agreement with the present

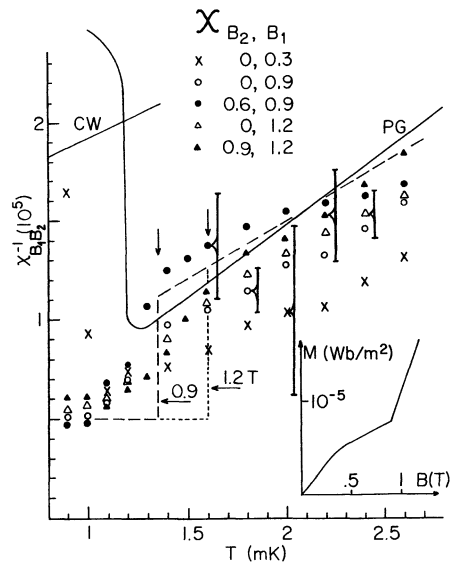


FIG. 3. — Inverse susceptibility  $\chi^{-1}$  deduced from melting pressure differences versus temperature given by relation (8). For  $B = 0.9$  and 1.2 T the arrows show the temperatures of the specific heat discontinuity. The dashed lines are the estimated  $\chi(B)$  for  $B = 1.2$  and 0.9 T with the calculated discontinuity  $\Delta\chi$ . The solid line PG gives the experimental results of Prewitt and Goodkind [3]. The solid line CW gives the Curie-Weiss value with  $\theta = -3$  mK. An estimate of the experimental errors is given by the bars. The inset shows an estimate, through relation (12), of the nonlinear curve  $M(B)$  with  $\Delta\chi = 1.8 \times 10^{-5}$  at  $B = 0.9$  T for  $T = 1.4$  mK. The accuracy for the bump near 0.3 and 0.4 T is poor because of the experimental uncertainties at low field for small  $\Delta P_m$ .

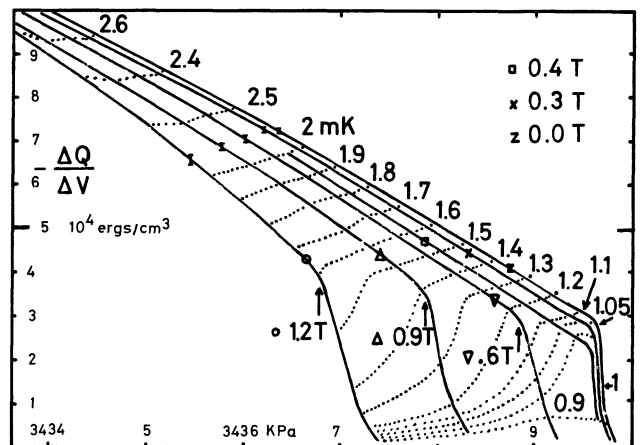


FIG. 4. — Data  $-\Delta Q/\Delta V$  versus pressure of K.M.A. [2]. Solid line : constant  $B$  curves. Dotted line : constant  $T$  lines which have been interpolated near the discontinuity at  $T_c(B)$  (shown by the arrows), with the help of thermodynamic relations. Above  $T_c(B)$ , the deviation of constant  $T$  lines from a straight line are due to the experimental uncertainties on  $-\Delta Q/\Delta V$  as shown by the bars. The relative errors on  $P$  are in comparison negligible.

low-field results. Using NMR techniques, Osheff [7], Bernier and Delrieu [8], Bakalyar *et al.* [9], and Morii *et al.* [10] have observed the increase in susceptibility above the C-W value above  $T_c$ . Prewitt

and Goodkind [3] have succeeded in making static measurements below  $T_c^0$  using a SQUID. They find a 60 % decrease in magnetization on cooling through the transition, in qualitative agreement with the present results for  $B \leq 0.3$  T.

We now discuss the question of whether the behaviour in fields  $B > 0.42$  T indicates a phase transition or simply ordering through alignment of the spins by the fields as suggested by KMA. For  $B \leq 0.42$  T the entropy curves of KMA show a very abrupt decrease at  $T_c$ , possibly indicating a first-order transition. At fields greater than this, the decrease in entropy is smoother but may still have a discontinuous slope characteristic of a second-order transition. The raw data curves  $\frac{\Delta Q}{\Delta V}(P) \approx T \frac{s(P)}{\Delta v}$  in figure 4 (and figure 4 of KMA) show that within about 0.1 mK, a fast change of slope of  $T \frac{s}{\Delta v}$  occurs at  $T_c(B)$  given by the arrows on figures 1 and 4. This change occurs in a temperature interval as small as that which can be resolved experimentally (when one accounts for the experimental errors) and for that reason we call it a *discontinuity*.

To look for a *discontinuity*, it is useful to suppress the high-temperature behaviour of  $s$  by plotting  $(1 - s/R \ln 2) T^2$  versus  $1/T$ . This plot (not shown here because of space limitation) does strongly suggest that there is a *discontinuity* of the specific heat  $C = T \left( \frac{\partial s}{\partial T} \right)_B \frac{1}{v}$  at  $T_c(B)$ . Such a *discontinuity* implies through the thermodynamic relations at a second-order transition [5], a *discontinuity* of the magnetization derivative :

$$\Delta \left( \frac{\partial M}{\partial T} \right)_B = - \frac{dT_c}{dB} \Delta \left( \frac{\partial s}{\partial T} \right)_B \frac{1}{v} \quad (9)$$

and of the susceptibility

$$\Delta \chi = \Delta \left( \frac{\partial M}{\partial B} \right)_T \mu = - \frac{dT_c}{dB} \Delta \left( \frac{\partial M}{\partial T} \right)_B \mu. \quad (10)$$

From the raw data of figure 4 of KMA, this *discontinuity* is seen to occur over approximately 0.1 mK. Thus with the observed  $dT_c/dB \approx 0.6$  mK/T of KMA, the *discontinuity*  $\Delta \chi$  as a function of  $B$  is spread over about 0.16 T. We can calculate  $\Delta \chi$  directly from the raw data curve if we neglect  $s_L$  with respect to  $s_g$  and combine relations (1), (9) and (10) :

$$\begin{aligned} \Delta \chi &= \Delta \left( \frac{\partial M}{\partial B} \right)_T \mu = + \frac{\mu}{T_c v} \left( \frac{dT_c}{dB} \right)^2 \Delta C = \\ &= - \frac{\mu}{T_c^2} \left( \frac{dT_c}{dB} \right)^2 \frac{\Delta v}{v} \frac{\Delta Q}{\Delta V} \Delta \left\{ \frac{\partial}{\partial P} \left( \frac{\Delta Q}{\Delta V} \right) \right\}. \quad (11) \end{aligned}$$

Figure 4 of KMA for  $B = 1.2$  T and  $T = 1.6$  mK gives  $\Delta Q/\Delta V = -4 \times 10^4$  erg/cm<sup>3</sup>,

$$\Delta \left\{ \frac{\partial}{\partial P} \left( \frac{\Delta Q}{\Delta V} \right) \right\} = -60 \text{ erg/cm}^3 \text{ Pa},$$

and  $dT_c/dB = 0.6$  mK/T at the *discontinuity*. This gives a *discontinuity*  $\Delta C = 8 \times 10^3$  erg/mK cm<sup>3</sup> and  $\Delta \chi = 1.8 \times 10^{-5}$ , which is of the same order as the maximum susceptibility plotted in figure 3. We predict also a *discontinuity* of

$$\Delta \left( \frac{\partial M}{\partial T} \right) \mu = - \frac{dB}{dT_c} \Delta \chi = -3 \times 10^{-2} \text{ Wb/m}^2 \text{ K}. \quad (12)$$

On the raw data of figure 4, the constant temperature  $T$  curves are straight lines if  $\chi$  is constant with field. For  $T > T_c(B)$  the points are within experimental errors on a straight line, so that  $\chi$  is practically constant ; thus the bump in figure 2 near 0.4 T is within these experimental errors. For  $T < T_c(B)$  the constant  $T$  lines are curved, well behind the experimental errors, so that  $\chi$  is no more constant ; this comes from the *discontinuity* at  $T_c(B)$  in the raw data curve.

Further evidence of a second-order phase transition at high fields can be seen in figures 2 and 3. For  $B > 0.6$  T and  $T \geq 1.8$  mK, the constant  $T$  lines of figure 2 are almost straight. However, at  $T_c(B) \leq 1.6$  mK, an increase in slope of the high- $B$  end of the curve occurs. From equations (7) and (8) this increase in slope shows that around  $T_c(B)$  there is an increase of the susceptibility which is smeared between  $T_c(B_1)$  and  $T_c(B_2)$ . In figure 2 the interpolating lines are drawn taking account of the change in  $\Delta \chi$  calculated above. This gives a *discontinuity* of the second derivative of  $\Delta P_m$  in figures 1 and 2 at the arrows. Through equation (8) we obtain the qualitative curve  $M(B)$  shown in the inset of figure 3 for  $T = 1.4$  mK. Thus we believe that the results of KMA provide good evidence of a second-order transition at  $B(T_c)$ . In this high field ordered phase, there is an abrupt increase in susceptibility rather than a decrease as in an antiferromagnet.

Assuming zero point vacancies which behave like spin polaron, Heritier and Lederer [11] predict a susceptibility larger than the Curie-Weiss value at low field, which saturates for  $B \sim 0.1$  T but cannot explain an increased susceptibility up to 1.2 T. Andreev, Martchenko and Meierovich [12] propose that the number of zero point vacancies is a function of the spin state. They find a phase diagram with a low field second-order transition (from para- to antiferromagnetism with no vacancy) and a high field first-order transition (from para- to ferromagnetism with a concentration  $6 \times 10^{-3}$  of vacancies). This is in contradiction with the experimental results previously analysed in this paper.

The model of Hetherington [13], further analysed by Roger *et al.* [14, 15], assumes different exchange interactions including up to four spins. If we assume the following set of values [16] :

- first neighbour pair exchange  $J_1 = -0.56$  mK,
- second neighbour pair exchange  $J_2 = -0.175$  mK,
- planar four spin exchange  $K_p = -0.32$  mK,

and all other exchanges vanishing, a phase diagram is possible with a low field first-order transition to a low susceptibility scf || phase [14] and a high field second-order transition to a *pseudo-ferromagnetic* phase with an increased susceptibility.

This model also explains other experimental results on solid  $^3\text{He}$  which are in contradiction with the usual antiferromagnetic model : the negative sign of the

$T^{-3}$  term in the specific heat [14, 17] and the increase of the susceptibility with respect to the Curie-Weiss law in the paramagnetic phase [15] presented in this paper for high field. In conclusion, we point out that in solid  $^3\text{He}$  at  $T = 1.6$  mK and  $B = 1.2$  T there is within 0.1 mK a second-order transition to a high field phase with an increased susceptibility, which has some similarity with a ferromagnetic phase. On the contrary, the low field phase with a small susceptibility has some similarity to an anti-ferromagnetic phase. This paper points out the necessity of more direct and precise experiments in particular, at larger fields in order to determine the maximum of  $T_c(B)$  which could be around 2 mK, or around the critical point at 0.4 T and 1 mK.

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