Theory of Magnetized Solid Helium Three

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Experiments on the thermodynamic properties of magnetized solid He³ at accessible temperatures and fields are shown to be a conveneint and powerful method of studying the nature and form of the nuclear spin interaction in solid He³. In particular, it should be possible to determine whether these properties are described in a satisfactory way by an exchange-interaction model assumed to be proportional to the scalar product of nuclear spin vectors of nearest-neighbor atoms of the solid. Independently of this model, it is shown that measurements of the magnetic modification of the melting pressure, at low but readily accessible temperatures and moderate field strengths, correspond to almost absolute measurements, of good accuracy, of the nuclear paramagnetic susceptibility of the solid at melting. The entropy diagram of the magnetized model solid enables one to describe with precision the isothermal magnetization and adiabatic demagnetization processes, which should contribute to the verification of the model through their experimental investigation. The increase in the heat capacity on magnetization is large enough to be maesured accurately and to test the validity of the model. An interesting effect is that the thermally anomalous unmagnetized solid becomes normal on magnetization, over a finite and low but accessible temperature range, and at intermediate field strengths. Experimental investigations of various equilibrium thermal properites of magnetized solid He³ could thus contribute significantly toward the elucidation of the behavior of its nuclear spin system which alone determines its thermal properties over the currently accessible low temperatures.

1. INTRODUCTION

ONE of the outstanding problems in the theory of the solid phases of He³ refers to the behavior of their nuclear spin systems at low and very low temperatures. In the liquid phase, the nuclear spin system achieves increasing degrees of order at low temperatures under the influence of the Pauli exclusion principle and Fermi statistics.[1,2] Here, the spin system was expected to exhibit paramagnetic behavior down to the temperature of absolute zero.[1,2] This prediction is well substantiated down to the lowest temperatures reached so far in magnetic susceptibility determinations on the saturated and compressed liquid.[3-5] In contrast with the statistical spin ordering in the liquid phase, the localization in coordinate space of the atoms of the solid phases raises the problem of a different kind of spin ordering resembling that in normal solids. One is led to invoke here magnetic ordering, nuclear ferromagnetism, or antiferromagnetism. In recent experimental work of significant importance on the low-pressure body-centered-cubic (bcc) solid He³, down to quite low temperatures, $T \sim 20m°K$, Adams and his collaborators,[6] at the University of Florida, have succeeded in proving that this solid must approach, at $T \ll 20m°K$, a transition into a spin-ordered phase. The Florida work

is also in agreement with the prediction that the entropy would decrease on isothermal volume increase, discussed by us earlier[7] in a semiqualitative way, and rigorously in recent work.[8] The latter work is based on the antiferromagnetic exchange coupling model, characterized by a single energy parameter J, and the scalar product of nearest-neighbor spin vectors, the Heisenberg model. Inasmuch as the temperature range explored by the Florida workers was the asymptotic or high-temperature range, $T \gg T_x = |J|/k$, only the $(J/kT)^2$ term of the indicated entropy variations was ascertained, proving the impending transformation of the spin system. However, the sign of the exchange-energy parameter, which determines, within the formalism of the model, the ferromagnetic or antiferromagnetic character of the transformation, according as $J \gtrless 0$, was not obtained so far. Direct experimental investigations of the transformation across the transition temperature of about $3(|J|/k)$, or 2m°K, according to the value of $|J|$ determined by the Florida group,[6] appear to be beyond current technical accessibility. However, the experimental problem raised by the nature of the transformation can still be approached in the paramagnetic range, $T > T_{0,x} \cong 3|J|/k$. Paramagnetic susceptibility measurements enable one to determine the sign of J unambiguously, provided the temperature range of these magnetic measurements is extended below 20m°K. But in addition to the nature of the magnetic transformation and the determination of the exchange-energy parameter and its volume or pressure dependence, $J(V)$ or $J(p)$, we wish to call attention to the following outstanding problem.

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[6] M. F. Panczyk, R. A. Scribner, G. C. Straty, and E. D. Adams, Phys. Rev. Letters **19**, 1102 (1967). I should like to thank the members of the University of Florida group for a copy of their paper prior to publication.

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[8] L. Goldstein, Phys. Rev. **159**, 120 (1967).

Assuming that both $J(V)$ and $J(p)$ have been obtained, what is the exact form of the interspin potential energy? Considering, for instance, the derivative thermal properties arising from the spin system, and exhibiting the asymptotic $1/T^2$ behavior, one must keep in mind that the specific form of the spin-spin interaction affects only the terms in $1/T^3$ and those of higher power of $1/T$ appearing in the theoretical representation of these properties in the paramagnetic range. Similarly, the specific form of the interspin coupling affects only the term in the paramagnetic susceptibility formula which is beyond the so-called molecular field theoretical term. The latter term, however, determines the sign of the exchange-energy parameter J. In order to reach, with satisfactory precision and accuracy, the specific interaction terms which appear only at temperatures below the asymptotic range, two possible ways of approach may be considered. One may hope to investigate the thermal and low magnetic field strength properties of solid He³ at very low temperatures which are currently very difficult to obtain in this phase. Or, one may magnetize the solid in constant and uniform fields of increasing though available strengths. In the latter case, the equilibrium thermal properties, at presently accessible temperatures, of the magnetized solid should enable one to attack, in a promising way, the problem of the specific form of the mutual spin-dependent exchange interaction. Accordingly, the subject matter of the present paper is the theoretical elaboration and analysis of the thermal properties of the low-pressure magnetized solid He³ on the basis of the Heisenberg exchange coupling model. The direct and quantitative comparison of the experimentally determined properties with those given and discussed in the present work should be capable of ascertaining whether or not the exchange-interaction model used here is valid in solid He³.

2. SOLID He³ IN MAGNETIC FIELDS OF LOW OR MODERATE STRENGTH

A. Magnetic Modification of the Melting Process

In the bcc modification of solid He³, the exchange coupling

$$V_{ij} = -2J\mathbf{s}_i \cdot \mathbf{s}_j, \qquad (1)$$

of two nearest-neighbor atoms i and j, of spin quantum number $\frac{1}{2}$, with spin vectors $\mathbf{s}_i$, in units of $\hbar$, leads to the magnetic Hamiltonian H_M in the presence of a constant uniform magnetic field of strength H,

$$H_M = \sum_{\langle i,j \rangle} V_{ij} - g\mu_0 \sum_i \mathbf{s}_i \cdot \mathbf{H}. \qquad (2)$$

The $\langle i,j \rangle$ summation refers to nearest neighbors, and the i summation of the second term on the right-hand side extends over the whole system of N atoms. The two types of terms of H_M commute, with $g\mu_0\mathbf{s}_i$ or $\mathbf{u}_i$ being the elementary magnetic dipole moment vector of atom

i, μ_0 being the relevant unit moment and g the splitting factor. The exact high-temperature series representations of the low-field-induced paramagnetic moment, $M(J/kT,\mu H/kT)$, and exchange heat capaicty $C(J/kT, H=0)$ have been extended[9] to about six terms, for a number of lattice structures and a number of spin values. These result, according to statistical thermodynamics, from the partition function $Z(x,y)$ of the Hamiltonian H_M,

$$Z(x,y) = \mathrm{Tr}\left[\exp(-H_M/kT)\right], \qquad (3)$$

through

$$M(x,y)/N\mu = (\partial/\partial y)[N^{-1}\ln Z(x,y)] \qquad (4)$$

and

$$C(x)/Nk = (\partial/\partial T)\{T^2(\partial/\partial T)[N^{-1}\ln Z(x, y=0)]\}. \qquad (5)$$

Here, in the relevant parameters,

$$x = (J/kT), \quad y = (\mu H/kT), \qquad (5')$$

J is the numerical value of the assumed antiferromagnetic exchange coupling energy, the negative sign of which will be taken into account through the powers of -1 in the exact if incomplete series representations of the thermal properties arising with H_M, Eq. (2). At low field strengths H, only terms containing y, that is H, to lowest power will appear in the field-dependent thermal properties.

For our purposes it is appropriate to consider the entropy of the assumed model of the antiferromagnetic solid He³, without and with the field. In the former case one finds, as given earlier[8]

$$S(x)/Nk = \ln 2 - 3x^2 \sum_{n=0} (-)^n[a_n^{(0)}/(n+2)]x^n, \qquad (6)$$

where the coefficients $a_n^{(0)}$ have been evaluated up to $a_8^{(0)}$ recently, in several cubic structures.[10] From the low-field-strength magnetic-moment series, or

$$M(x,y)/N\mu = y \sum_{n=0} (-)^n a_n^{(1)}x^n, \quad y \ll 1 \qquad (7)$$

one obtains, from thermodynamics, the entropy change on magnetization at low field strengths,

$$[S^{(\bullet)}(x,y)/Nk]_{y \ll 1}$$

$$= \int_0^{\mu H} \{\partial[M(x,y)/N\mu]/\partial(kT)\}d(\mu H)$$

$$= \frac{1}{2}y^2[1 + \sum_{n=1} (-)^n(n+1)a_n^{(1)}x^n], \qquad (8)$$

where the $a_n^{(1)}$ coefficients are now available up to $a_{10}^{(1)}$ for several cubic structures.[10] In small fields, the

[9] G. S. Rushbrooke and P. J. Wood, Mol. Phys. **1**, 257 (1958); **6**, 409 (1963). These papers contain references to the earlier literature.

[10] G. A. Baker, Jr., H. E. Gilbert, J. Eve, and G. S. Rushbrooke, Phys. Letters **20**, 146 (1966); **22**, 269 (1966); Phys. Rev. **164**, 800 (1967).

entropy is then given by

$$S(x,y)_{x<1,\ y\ll1}=S(x)+S^{(\epsilon)}(x,y),\qquad(9)$$

which, by (6), (7), and (8) reduces in the limit of $x \to 0$ to

$$\lim_{x\to0} S(x,y)/Nk\sim\ln2-\tfrac12 y^2,\quad y\ll1\qquad(10)$$

the entropy of an ideal paramagnetic system at low fields and high temperatures. With (6), (8), (9), and (10), and the statistical thermodynamic relation

$$S(x,y)/Nk=(\partial/\partial T)[TN^{-1}\ln Z(x,y)],\qquad(11)$$

one obtains

$$N^{-1}\ln Z(x,y)$$
$$=\ln2+3x^2\sum_{n=0}(-)^n[a_n^{(0)}/(n+1)(n+2)]x^n$$
$$+\tfrac12 y^2\sum_{n=0}(-)^n a_n^{(1)}x^n,\qquad(12)$$

the logarithm of the partition function per atom or the negative of the free energy in units of kT, to lowest order in the applied field strength. This can be re-written as

$$N^{-1}\ln Z(x,y)_{x<1,\ y\ll1}=N^{-1}\ln[Z(x)Z(y)Z^{(i)}(x,y)],\qquad(13)$$

with

$$N^{-1}\ln Z(x)=3x^2\sum_{n=0}(-)^n[a_n^{(0)}/(n+1)(n+2)]x^n,$$

$$N^{-1}\ln Z(y)=\ln2+\tfrac12 y^2,\qquad(14)$$

$$N^{-1}\ln Z^{(i)}(x,y)=\tfrac12 y^2\sum_{n=1}(-)^n a_n^{(1)}x^n.$$

The partition function is thus decomposed into that of the exchange-coupled system in absence of the field $Z(x)$, with the spin multiplicity factor omitted, the partition function of the asymptotic ideal system $Z(y)$, and that arising from the interactions of the x and y fields, the exchange field, and the applied external field, $Z^{(i)}(x,y)$. This product representation of the partition function, to lowest order in the applied field strength, satisfies the requirement that in the two limits, $x \to 0$ and $y \to 0$, it should reduce, respectively, to that of an ideal system of spin-$\tfrac12$ particles, or that of the exchange-coupled system in absence of the external field. Let us assume now that the two relevant parameters of the system, x and y, can be varied independently, through modifying J and H. It is then justified to consider the situation where y increases and x is reduced so that the interaction term $Z^{(i)}(x,y)$ may still be represented as in (14) above, while $Z(y)$ has to be completed by additional powers in the still moderate but increasing y. Now with $x \to 0$, $Z(y)$ must reduce to $(2\cosh y)^N$, the exact partition function of the ideal system. At small enough x values, it is justified to retain only the first term of

$Z^{(i)}$, and write, on expanding $\cosh y$ in the just indicated exact form of $Z(y)$,

$$N^{-1}\ln Z(x\ll1,\ y\lesssim1)$$
$$=\ln2+\ln\left(1+\sum_{n=1}^{\infty}\frac{y^{2n}}{(2n)!}\right)-\tfrac12 a_1^{(1)}y^2 x\qquad(15)$$

neglecting $Z(x\ll1)$ whose lowest-order term is quadratic in the very small x. Here one must have

$$\sum_{n=1}^{\infty}\frac{y^{2n}}{(2n)!}\le(\cosh y-1)\le1,\qquad(16)$$

or y must be such that

$$\cosh y\le2,$$
$$y\lesssim1.32.\qquad(16')$$

At increasing x values, the $Z(x)$ part of the partition function becomes important and, in addition, the inter-action term $Z^{(i)}(x,y)$ becomes more complicated, involving increasing powers of both x and y. These latter terms will be considered in a later section devoted to the effect of increasingly large magnetic fields on solid He[3].

The foregoing discussion of the total partition function of our system, based on the exact derivation of its components, shows that the limiting ideal spin system is an integral part of the magnetized system. While the no-field antiferromagnetic component partition function $Z(x)$, as well as the interaction term $Z^{(i)}(x,y)$, though exact up to the number of terms known to date, are described in an incomplete fashion through a finite number of terms in their infinite-series representation, the component ideal system partition function $Z(y)$ can at all times be given in its exact form. Clearly, at low enough y values the use of limiting forms of the expansion of $Z(y)$ is fully justified.

To the approximation (15) of the partition function, the fractional magnetic moment or polarization is

$$\lim_{x\ll1,\ y\le1} M(x,y)/N\mu=(\tanh y)-a_1^{(1)}xy.\qquad(17)$$

The coefficient $a_1^{(1)}$ being positive,[9] the second term on the right-hand side is an approximate measure of the resistance which the internal antiferromagnetic x coupling develops against the ordering effect of the external field. The first term on the right-hand side is the fractional moment induced in the ideal spin-$\tfrac12$ component of the system at hand. In the limit $y\ll1$, one obtains

$$\lim_{x\ll1,\ y\ll1} M(x,y)=N\mu y(1-a_1^{(1)}x)$$

$$\simeq N\mu y/(1+a_1^{(1)}x),\qquad(18)$$

the latter fractional form being valid only to terms linear in x. This is the high-temperature molecular field

approximation to the induced moment, with

$$a_1^{(1)}x = a_1^{(1)}J/kT$$
$$= T_N/T, \qquad (19)$$

T_N being the molecular field theoretical approximation of the antiferromagnetic spin ordering or Néel temperature.

Another aspect of the competition between the antiferromagnetic exchange coupling within the system and the applied external field clearly appears in the expression of the entropy of the magnetized system. To the approximation of the partition function (15), one obtains

$$\lim_{x \ll 1, y \sim 1} S(x,y)/Nk = \partial/\partial T[TN^{-1}\ln Z(x,y)]$$
$$= [\ln(2\,\cosh y)] - y(\tanh y) + a_1^{(1)}y^2x. \qquad (20)$$

The interaction between the x and y couplings is seen to increase the entropy, the coefficient of the interaction term $a_1^{(1)}$ being positive, as noted already. The two terms preceding the entropy of interaction represent $S_{id}(y)$, the entropy of the asymptotic ideal system in presence of the external field.

In relaxing the limitations on x used in Eqs. (15)–(20), $Z_{id}(y)$ can replace $Z(y)$ in the partition function (13), as justified above. By limiting the interaction function $Z^{(i)}(x,y)$ to lowest order in y, as in (14), we want to rewrite the total entropy of the system, to this approximation, as

$$S(x,y) = S(x) + [S_{id}(y) - Nk\ln 2] + S^{(i)}(x,y),$$

$$S^{(i)}(x,y)/Nk = -\tfrac{1}{2}y^2\sum_{n=1}(-)^n(n+1)a_n^{(1)}x^n, \qquad (21)$$

where $S(x)$ was given above [Eq. (6)]; the middle terms have been defined through Eq. (20), and $S^{(i)}(x,y)$ is seen to be $S^{(i)}(x,y)$ defined by (8) augmented by $\tfrac{1}{2}y^2$.

We are now prepared to discuss the modifications of various thermal properties of antiferromagnetic solid He³ on magnetization in low and moderate fields, in the paramagnetic region. At the present time, the measurements of the changes in the melting pressure of magnetized solid He³, as well as the changes in the temperature derivatives of the melting line, should enable one to verify experimentally the entropy decrease on magnetization, at accessible temperatures and field strengths. To show this we have to derive the melting pressure of the solid in the presence of a constant and uniform magnetic field, where the entropy derived above represents with good approximation its field dependence.

The consistent omission of all shape-dependent demagnetization effects in the magnetized solid will be adhered to throughout the present work.

According to thermodynamics, the temperature derivative of the melting pressure is given by

$$dP_M(x,y)/dT = (\Delta V_M)^{-1}[S_{L,M}(T) - S_{s,M}(x,y)], \qquad (22)$$

with

$$\Delta V_M(T) = V_{L,M}(T) - V_{s,M}(T) \qquad (22')$$

standing for the volume change at melting of the unmagnetized system, and where the subscripts L and s refer to the liquid and solid, respectively. The very small volume changes of these dense phases on magnetization are assumed to be negligible. The entropy of the solid at melting is taken to be given by (21), all component entropies and the parameters defining them referring to melting conditions. As discussed by us recently[8] the empirical exchange energy parameter J_M, at melting, may be assumed to remain constant approximately over the small temperature and melting volume ranges relevant for the calculation of the thermal properties. This assumption may not be as restrictive as would appear, at first sight, because the volume changes of the solid at melting, over the pressure or temperature intervals of particular interest here, are expected to be moderate. This then would tend to keep the variations of $J_M(V)$ within a correspondingly limited range. An improved calculation of the entropy of the solid at melting can be performed as soon as the function $V_{s,M}(T)$ becomes available, together with $V_{L,M}(T)$, at the lowest accessible temperatures. However, the loss of numerical accuracy of the calculated thermal properties arising from these various sources will have to be kept in mind.

On substituting $S_{s,M}(x,y)$ given by (21) into (22) and making use of the relations (6) and (8), one obtains the melting-pressure derivative as a sum of two terms:

$$\frac{dP_M(x,y)}{dT} = \frac{dP_M(x)}{dT} + \frac{dP_M^{(i)}(x,y)}{dT}, \qquad (23)$$

with

$$dP_M(x)/dT = (\Delta V_M)^{-1}[S_{L,M}(T) - S_{s,M}(x)] \qquad (24)$$

being the melting-pressure derivative of the free or unmagnetized solid, and

$$dP_M^{(i)}(x,y)/dT = (-)(R/\Delta V_M)[\ln(\cosh y) - y(\tanh y) + S^{(i)}(x,y)/R], \qquad (25)$$

being the interaction derivative (where R is the gas constant) which is positive since the entropy changes on magnetization and the quantity inside the square brackets is negative, even though $S^{(i)}$ itself, arising from the interactions of the x and y fields, is positive. Hence,

$$dP_M(x,y)/dT > dP_M(x)/dT < 0, \qquad (26)$$

the melting-pressure derivative $dP_M(x)/dT$ of free He³ being anomalous or negative throughout the temperature range of interest here, i.e., at $T_\mu' \ll T \le T_\mu$, T_μ and T_μ' being the temperatures of the melting-pressure minimum and maximum, and the latter being at the very low temperature of a few millidegrees, as discussed recently.[8] It is seen that both (22) and (25) carry the

following additional approximation: The modification of the entropy of the liquid at melting due to the presence of the magnetic field has been neglected. This will be justified over the relevant temperature range on the basis of a quantitative argument to be advanced below.

On integrating (23) between T_μ and $T < T_\mu$, one obtains the melting pressure difference

$$P_M(x,y) - P_M(x_\mu,y_\mu) = [P_M(x) - P_M(x_\mu)]$$
$$+ [P_M^{(i)}(x,y) - P_M^{(i)}(x_\mu,y_\mu)], \quad (27)$$

with

$$P_M(x) - P_M(x_\mu) = \frac{R}{\Delta V_M}\left\{\int_{T_\mu}^{T} \frac{S_{L,M}(T)}{R} dT\right.$$
$$\left. + (T_\mu - T)\ln 2 - [Q(x) - Q(x_\mu)]\right\}, \quad (28)$$

where

$$Q(z) = 3(J_M/k)z\sum_{n=0} (-)^n[a_n^{(0)}/(n+1)(n+2)]z^n. \quad (29)$$

Equation (28) is thus the melting-pressure equation of He^3 at $T_\mu' \ll T \leq T_\mu$, in absence of the magnetic field. This melting pressure has been obtained by us recently[8] through combining the numerically evaluated entropies of the liquid and solid at melting and integrating their difference numerically. Since an analytic expression of $S_{L,M}(T)$ valid throughout the range $T_\mu' \ll T < T_\mu$ is not available at the present time, a more explicit form than (28) of the melting line cannot be given. It should be noted that the very small phonon entropy of the solid has been neglected consistently both in the earlier calculation and in the present one.

The field-dependent part of the melting pressure is, from (25),

$$P_M^{(i)}(x,y) - P_M^{(i)}(x_\mu,y_\mu) = -(R/\Delta V_M)$$
$$\times [Q^{(i)}(T,x,y) - Q^{(i)}(T_\mu,x_\mu,y_\mu)], \quad (30)$$

with

$$Q^{(i)}(T,x,y) = T[\ln(\cosh y) + \tfrac{1}{2}y^2\sum_{n=1} (-)^n a_n^{(1)}x^n]. \quad (31)$$

This achieves the determination of the melting pressure of He^3 at temperatures $T < T_\mu$, to the stated approximation, in the presence of a small or moderate magnetic field. The melting pressure is reduced on magnetization as a consequence of the entropy decrease of the magnetized solid when compared to the free solid.

Over the ranges of T, x, and y of particular interest here, one may, in a first approximation, neglect $Q^{(i)}(T_\mu,x_\mu,y_\mu)$ in comparison with $Q^{(i)}(T,x,y)$. The melting-pressure decrease, in the state (x,y), is then

$$\lim_{x \ll 1, \; y \ll 1} P_M^{(i)}(x,y)$$
$$= -(RT/\Delta V_M)(\tfrac{1}{2}y^2)(1 - a_1^{(1)}x), \quad (32)$$

keeping only the term linear in x in the series on the right-hand side of (31) and writing $\tfrac{1}{2}y^2$ for $\ln(\cosh y)$, at small y. To terms linear in x, one has

$$\lim_{x \ll 1, \; y \ll 1} P_M^{(i)}(x,y) = -\frac{(R/2\Delta V_M)(\mu H/k)(\mu H/kT)}{1 + a_1^{(1)}x}$$
$$= -\frac{(1/2\Delta V_M)(N\mu H)(\mu H/kT)}{1 + a_1^{(1)}x}$$
$$= -M(x,y)H/(2\Delta V_M), \quad (33)$$

where use was made of the limiting small x- and small y-value expression of the induced magnetic moment of the solid, Eq. (18). Now $-\tfrac{1}{2}M(x,y)H$ is the magnetic potential energy acquired by the solid in the course of raising the applied field strength from 0 to its value H, i.e., on magnetization to this field strength. Hence, to the stated approximation, the melting-pressure decrease is seen to be given by the magnetic potential-energy density of the solid defined over the volume change at melting ΔV_M.

On keeping both Q functions on the right-hand side of (30) and using the indicated lowest-order x and y approximations to these functions, one finds

$$P_M^{(i)}(x,y) - P_M^{(i)}(x_\mu,y_\mu)$$
$$= -(H/2\Delta V_M)[M(x,y) - M(x_\mu,y_\mu)]. \quad (34)$$

To the order indicated,

$$M(x,y) = \chi(x)HV_{s,M}(T), \quad M(x_\mu,y_\mu) = \chi(x_\mu)HV_{s,M}(T_\mu),$$
$$(35)$$

with the susceptibility

$$\chi(x) = (N\mu^2/V_{s,M}kT)(1 - a_1^{(1)}x)$$
$$\cong (N\mu^2/V_{s,M}kT)/(1 + a_1^{(1)}x), \quad (36)$$

to terms linear in x. The change in the melting-pressure variation from the zero-field case, over the interval (T,T_μ), is then

$$P_M^{(i)}(x,y) - P_M^{(i)}(x_\mu,y_\mu) = -(H^2/2\Delta V_M)$$
$$\times [\chi(x)V_{s,M}(T) - \chi(x_\mu)V_{s,M}(T_\mu)]. \quad (37)$$

Since the variations of the volume of the solid at melting are quite small at $T < T_\mu$, one may write, approximately,

$$P_M^{(i)}(x,y) - P_M^{(i)}(x_\mu,y_\mu)$$
$$= -(H^2 V_{s,M}(T_\mu)/2\Delta V_M)[\chi(x) - \chi(x_\mu)]. \quad (37')$$

To the indicated approximation, the Eqs. (34) through (37') show that the measurements of melting pressure changes of the magnetized solid over the temperature interval (T,T_μ) are equivalent to the measurements of the changes of its induced paramagnetic moment over that interval.

With the moment $M(x,y)$ or the susceptibility $\chi(x)$ being decreasing functions of the temperature, the lower T is the better it is justified to neglect the pressure change $P_M^{(i)}(x_\mu,y_\mu)$ in comparison with $P_M^{(i)}(x,y)$. To the approximation of neglecting $P_M^{(i)}(x_\mu,y_\mu)$, the measurement of the change in the melting pressure

$P_M^{(i)}(x,y)$ yields an approximate value of the induced moment $M(x,y)$ and the associated susceptibility, Eqs. (33) and (35). This approximate value of the induced moment may be said to be measured in a way which is close to being absolute. Indeed, Eq. (33) yields $M(x,y)$ in terms of the directly measurable melting pressure change $P_M^{(i)}$, the volume change at melting ΔV_M, and the strength H of the applied external field, both of these being directly measurable also. If $P_M^{(i)}$ were to be measured through a pressure measuring device, then $M(x,y)$, defined by (33) would correspond to an absolute, if approximate, induced paramagnetic moment, and the same remark applies to the associated susceptibility. Since, however, neither $P_M^{(i)}$ nor H are measured directly, in the sense that the latter, for instance, is not accessible to a magnetic balance, the absolute character of the determination of $M(x,y)$ or $X(x)$ is lost. It is, however, realized that the paramagnetic moment or susceptibility of solid He³ at melting, and only at melting, can thus be reached directly with possibly good accuracy through measurements of melting-pressure variations. The technique of measuring small pressure changes with great accuracy, developed and used recently by the University of Florida workers,[6] could find ready application in the investigation of the melting process of magnetized solid He³.

With the melting-pressure variations in presence of a magnetic field being proportional to the variations of the induced moment [Eq. (34)] it is seen that their measurements can yield the sign of the exchange energy parameter J, and determine thereby the nature of the magnetic transformation of the solid. Indeed, considering the melting-pressure changes given by Eqs. (30) and (31), one can write these also as

$$P_M^{(i)}(x,y) - P_M^{(i)}(x_\mu,y_\mu) = \Delta P_{id}(y) + \Delta P_M^{(i)}(x,y), \quad (38)$$

with

$$\Delta P_{id}(y) = -(R/\Delta V_M)[T\ln(\cosh y)$$
$$- T_\mu \ln(\cosh y_\mu), \quad (38')$$

and

$$\Delta P_M^{(i)}(x,y) = -(R/\Delta V_M)[q^{(i)}(x,y) - q^{(i)}(x_\mu,y_\mu)],$$

$$q^{(i)}(x,y) = T(\tfrac{1}{2}y^2)\sum_{n=1}(-)^n a_n^{(1)}x^n. \quad (38'')$$

The pressure change $\Delta P_{id}(y)$ arising from the magnetization is the one which would be observed if the solid were an ideal paramagnet. As a consequence of the entropy decrease of the ideal system of spins, the melting pressure decreases. This decrease is then compensated in part by the interaction term which is a measure of the resistance developed by the internal antiferromagnetic couplings against the entropy decrease imposed by the ordering effect of the applied field. The infinite series in $q^{(i)}(x,y)$ being convergent, the sign of $q^{(i)}$ is that of its first term which is negative, the same as the sign of J, and $\Delta P_M^{(i)}$ is positive. In antiferromagnetic systems the component variations $\Delta P_{id}(y)$ and $\Delta P_M^{(i)}(x,y)$ are of opposite sign, in the assumed exchange-coupling model of these systems. Accordingly, the experimental determination of the opposing signs of $\Delta P_{id}(y)$ and $\Delta P_M^{(i)}(x,y)$ is equivalent, within the framework of the exchange model, Eqs. (1) and (2), to the verification of the antiferromagnetic character of solid He³.

Measurements of the melting pressure in presence of an applied external magnetic field involve the two-phase liquid-solid system at equilibrium. Both phases are magnetized and the entropy of the liquid will be decreased from its value in the absence of the field, as is the case with the solid phase shown above. However, within the framework of the theory of the spin system of the liquid[2] at $T \ll T_0(P_M)$, $T_0(P_M)$ being the characteristic temperature of this system at the melting pressure P_M, one has,[2,7] for the fractional induced magnetic moment,

$$M(T,P_M,H)/N\mu$$

$$= \frac{F[\alpha_M - (\mu H/kT)] - F[\alpha_M + (\mu H/kT)]}{F[\alpha_M - (\mu H/kT)] + F[\alpha_M + (\mu H/kT)]}, \quad (39)$$

where at the low temperatures of interest here

$$(-\alpha_M) = -\alpha(P_M) \sim T_0(P_M)/T \gg 1, \quad (\mu H/kT) = y \lesssim 1, \quad (40)$$

and $F(x)$ is a characteristic function of the formalism[2,7] which need not be given here explicitly. Inasmuch as at all field strengths considered here, $\mu H \ll kT_0$, one obtains, expanding numerator and denominator in (39) to terms linear in y,

$$\lim_{\mu H \ll kT_0(P_M)} M(T,P_M,H)/N\mu$$
$$= (\mu H/kT)[-F'(\alpha_M)/F(\alpha_M)], \quad (41)$$

$$F'(x) = dF/dx,$$

and using the low-temperature asymptotic series expansion[2,7,11] of the logarithmic derivative of $F(\alpha)$, one obtains

$$\lim_{\substack{\mu H \ll kT_0(P_M) \\ T \ll T_0(P_M)}} M(T,P_M,H)/N\mu = \tfrac{3}{2}(\mu H/kT_0)$$

$$\times\left[1 - \tfrac{1}{12}\pi^2(T/T_0)^2 - \frac{11\pi^4}{360}(T/T_0)^4 - \cdots\right]. \quad (42)$$

Using now the thermodynamic relation included in the first of Eqs. (8), one finds the entropy decrease of the

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[11] The expansion of $M(T,H)$ up to $(T/T_0)^6$ was first given by E. C. Stoner, Proc. Leeds Phil. Lit. Soc. Sci. Sec. 7, 403 (1938). Additional terms up to $(T/T_0)^{12}$ are to be found in the second paper of Ref. 7.

magnetized liquid, to the above approximation,

$$\lim_{\substack{\mu H \ll kT_0(P_M) \\ T \ll T_0(P_M)}} \Delta S_{L,M}(T,P_M,H)/Nk$$

$$= -\tfrac{1}{8}\pi^2(\mu H/kT_0)^2(T/T_0)$$

$$\times\left[1+\frac{11\pi^2}{15}(T/T_0)^2+\cdots\right], \quad (43)$$

since only the temperature-dependent part of the induced moment (42) can contribute. At all field strengths of interest here, as well as over the temperature range relevant here, one obtains, to lowest order in $\mu H/kT$ and $\mu H/kT_0$,

$$\Delta S_{L,M}/S(x,y) \sim (T/T_0)^3, \quad (44)$$

keeping only the term in y^2 in $S(x,y)$, arising from $S_{id}(y)-\ln2$, in Eq. (21) above. Now $T_0(P_M)$ is about 250m°K, over the temperature range of interest here which extends below 100 or 80m°K. Hence, the correction to the temperature derivative of the melting pressure amounts here at most to 3 or 4% of the melting-pressure derivative due to the magnetization of the solid, since

$$d\Delta P_{L,M}(T,H)/dT = \Delta S_{L,M}(T,P_M,H)/\Delta V_M. \quad (45)$$

To the approximation of a constant ΔV_M, the melting-pressure change over the temperature range (T_1,T_2) arising from the magnetization of the liquid is

$$\Delta P_{L,M}(T_2,H) - \Delta P_{L,M}(T_1,H)$$

$$= (1/\Delta V_M)\int_{T_1}^{T_2} \Delta S_{L,M}(T,P_M,H)dT, \quad (46)$$

whose relative magnitude is seen, with (43), to be again the fraction $(T_i/T_0)^3$ of the melting-pressure changes arising with the magnetization of the solid at T_i $(i=1, 2)$, to order y^2.

At the low temperatures of interest here, the corrections $\Delta P_{L,M}$ can be computed to a fair degree of accuracy and can be taken into account accordingly. At higher temperatures, $T \sim T_0(P_M)$, which include the reference temperature T_μ of the melting-line minimum in absence of the field, the susceptibility of the liquid approaches its classical or ideal limit. Neglecting the relatively small volume difference ΔV_M in comparison with the volumes themselves of the liquid and solid at melting, their respective paramagnetic susceptibilities are fairly close at or around T_μ. As emphasized already, the relevant temperature range for the investigations of the magnetic modification of the melting line is at most at $T < \sim 0.1°K$. Neglecting here, by (44), the effects arising from the magnetization of the liquid, the quantity to be measured should be taken, at $T_2 < T_1$,

to be

$$P_M(x_2,y_2) - P_M(x_1,y_1) = \Delta P_{id}(y_2,y_1)$$
$$+ \Delta P_M^{(i)}(x_2,y_2; x_1,y_1), \quad (47)$$

with

$$\Delta P_{id}(y_2,y_1) = (R/\Delta V_M)[T_1 \ln(\cosh y_1)$$
$$- T_2 \ln(\cosh y_2)] < 0, \quad (47')$$

and

$$\Delta P_M^{(i)}(x_2,y_2; x_1,y_1) = (-)(R/\Delta V_M)$$
$$\times [q^{(i)}(x_2,y_2) - q^{(i)}(x_1,y_1)], \quad (47'')$$

using (38), (38'), and (38''). The quantity (47) is obtained directly through the measurements of the melting-pressure changes between T_1 and T_2, in the absence and in the presence of the field. To lowest order in x and y, (37') becomes here

$$P_M(x_2,y_2) - P_M(x_1,y_1)$$
$$= (-)[H^2 V_M(T_1,T_2)/2\Delta_M][\chi(T_2) - \chi(T_1)], \quad (48)$$

to the approximation of an almost constant volume $V_M(T_1,T_2)$ of the solid over the indicated temperature range. If the volumes $V_M(T_1)$ and $V_M(T_2)$ are available, then a relation resembling (37) replaces (48). Inasmuch as at $T_1 \gg T_{0,x}$, $T_{0,x}$ being the very low spin ordering temperature of about $(3J/k)$, the nuclear susceptibility of solid He³ is practically ideal, it is seen again that the measurements of $P_M(x_2,y_2) - P_M(x_1,y_1)$ are equivalent to an almost absolute measurement of the nuclear susceptibility of the solid, in the state (x_2,y_2), at $T_2 < T_1$, at melting.

The discussion of the magnitude of the magnetic modification in the melting pressures will be postponed to a later section, where the effects of increasing field strengths will be included in the evaluation of the various thermal properties of the magnetized solid.

We have shown recently[8] that the entropy of the liquid at melting $S_{L,M}(T)$ can be evaluated to a fair degree of accuracy. Hence, by (24), melting-pressure derivative determinations can yield the entropy of the solid at melting. Even though the predictions of this model concerning solid He³ require for their experimental verification the development and use of advanced cryogenic techniques, this solid offers conceptually a much simpler realization of the exchange-coupling model than the one attributed to certain metals or nonmetals. In the latter systems, the coupling schemes (1) and (2) refer to electrons or ions, wherein various additional effects, some due to the motion of charges, will always tend to obscure the phenomena arising strictly with the dominant exchange-coupling schemes. The various side effects present in the electronic and ionic cases are absent in solid He³ in thermodynamic equilibrium. This should enable one to verify, in nearly ideal conditions, whether or not the exchange model, Eqs. (1) and (2), is valid in solid He³.

B. Derivative Thermal Properties of Solid He³ in Magnetic Fields of Moderate Strength

We saw in the previous subsection that the exchange-coupling scheme described by Eqs. (1) and (2) leads to definite predictions on the magnetic modification of the melting process of He³. Measurements of the melting pressure in the presence and the absence of the field enable one to obtain the paramagnetic susceptibility of the solid at melting, and determinations of the melting-pressure derivatives should yield the entropy of the solid at melting. Comparison of these measured properties with their theoretical predictions based on the coupling schemes (1) and (2) should show if the latter were obeyed in solid He³ or not. Verification of predictions based on the assumed model can also be performed through direct measurements of derivative thermal properties arising from the entropy, such as heat capacity and isobaric volume-expansion coefficient.

Using the entropy expression (21) one obtains

$$C(x,y) = C(x) + C_{id}(y) + C^{(i)}(x,y), \qquad (49)$$

with

$$C(x)/R = 3x^2 \sum_{n=0} (-)^n a_n^{(0)} x^n, \qquad (49')$$

$$C_{id}(y)/R = (y/\cosh y)^2, \qquad (49'')$$

$$C^{(i)}(x,y)/R = \tfrac{1}{2} y^2 \sum_{n=1} (-)^n (n+1)(n+2) a_n^{(1)} x^n. \qquad (49''')$$

To lowest order in the field strength, the excess heat capacity is, combining the first term of the y expansion of $C_{id}(y)$ with $C^{(i)}(x,y)$,

$$C^{(i)}(x,y)/R = \tfrac{1}{2} y^2 \sum_{n=0} (-)^n (n+1)(n+2) a_n^{(1)} x^n. \qquad (50)$$

This is positive as long as the x series is taken to be physically usable only over the range of x values where its convergence is uniform. In contrast with the entropy decrease of the solid on magnetization, the heat capacity increases. This is well known for paramagnetic systems at $\mu H/kT \ll 1$, or at $y \ll 1$. It is associated with the fact that the temperature rate of variation of the ordering effect of the field is positive even though this increase of order is equivalent to a decrease of entropy. As shown further below, the relative heat-capacity increases are quite large even in fields of moderate strength and at accessible temperatures.

Compared with the melting-pressure, entropy, and heat-capacity modifications due to magnetization, the problem of the magnetic changes of the expansion coefficient is of greater complexity. Namely, $(\partial V/\partial T)_P$ or $(V\alpha_p)$, α_p being the isobaric volume-expansion coefficient, and V the volume of the solid, depends not only on the exchange energy J, as the entropy, heat capacity, and the melting pressure, but on the pressure derivative of J also. One finds with the entropy (21) and the heat capacity $C(x)$, Eq. (49'), along the melting

line,

$$\alpha_{M,p} = -(1/V_M)[\partial S_M(x,y)/\partial p]_T$$

$$= -(R/V_M)|d \ln J_M/dp| \{ [C_M(x)/R] $$

$$+ \tfrac{1}{2} y^2 \sum_{n=1} (-)^n n(n+1) a_n^{(1)} x^n \}, \qquad (51)$$

or

$$\alpha_{M,p} = \alpha_{M,p}(x,J') + \alpha_{M,p}^{(i)}(x,J',y), \qquad (52)$$

where J' stands for (dJ/dp) or (dJ_M/dp). In the preceding relations, use was also made of the empirical result[6,12] that $J(p)$ is a decreasing function of the pressure or an increasing function of the volume. In the absence of the external field, one then has

$$\alpha_{M,p}(x,J') = -(1/V_M)|d \ln J_M/dp| C_M(x), \qquad (53)$$

which we gave recently[8] with $C_M(x)$ expressed through its series representation (49'). This shows that as long as it is justified to neglect the phonon contribution to the expansion coefficient, and over the temperature range where the exchange expansion coefficient $\alpha_{M,p}$-(x,J') is larger than the phonon expansion coefficient, solid He³ at melting and at low pressures is thermally anomalous. That is, it has a negative volume-expansion coefficient or its entropy increases on isothermal compression. We can rewrite (53) as

$$\alpha_{M,p}(x,J') = (-)(d \ln J_M/d \ln V_M) \chi_T C_M(x)/V_M, \qquad (54)$$

χ_T being the isothermal compressibility of the solid. The equivalent relation for (α_p/χ_T), that is, $(\partial p/\partial T)_V$, on the left-hand side was given and used by the Florida workers in connection with their measurements of this latter derivative.[6] In absence of the external field, the two subsystems of degrees of freedom of the solid, the phonon system and the spin system, give rise, at equilibrium, to the total entropy

$$S(T,\Theta,J) = S_\varphi(T/\Theta) + S_x(J/kT), \quad J/k < T < \Theta, \qquad (55)$$

a linear superposition of the component entropies, Θ being the characteristic temperature of the phonon system.[8] In terms of the simple phonon model, one obtains at melting or near melting

$$\alpha_{M,p}(T,\Theta_M,J_M/k)$$

$$= \alpha_{M,\varphi} + \alpha_{M,x}$$

$$= (-)(\chi_T/V_M)[C_{M,\varphi}(T/\Theta_M)(d \ln \Theta_M/d \ln V_M)$$

$$+ C_{M,x}(x)(d \ln J_M/d \ln V_M)]. \qquad (56)$$

The relevant physical characteristics of the two subsystems Θ and J appear in a similar way; and solid He³, in the representation (55) of its entropy, has the Grueneisen parameter

$$\Gamma_\varphi = (-)(d \ln \Theta/d \ln V), \qquad (57)$$

[12] R. C. Richardson, E. Hunt, and H. Meyer, Phys. Rev. 132, A1326 (1965); M. G. Richards, J. Hatton, and R. G. Giffard, ibid. 139, A91 (1965).

and the corresponding exchange parameter

$$\Gamma_x = (d \ln J / d \ln V), \qquad (58)$$

associated with the assumed coupling scheme arising from the spin system. These results cease to be valid in the presence of an external uniform and constant magnetic field. This arises from the entropy of interaction of the external field, or y field, and the internal or exchange field, the x field. The total entropy in presence of the external field is not a linear superposition of the entropies of the magnetic subsystem, or the y system, and of the exchange subsystem, the x system.

Returning now to the expansion coefficient (52), it is seen that this property can be strongly modified by the external field. This modification is such as to change the sign of the expansion coefficient. The excess expansion coefficient $\alpha_{M,p}{}^{(i)}(x,J',y)$, in Eq. (52), arising from $S^{(i)}(x,y)$, tends to compensate the anomalous negative expansion coefficient $\alpha_{M,p}(x,J')$ of the free solid in the absence of the field. To lowest order in the field strength as well as in the exchange-coupling strength, i.e., in y and x, the magnetic increase in the expansion coefficient is, at melting, by (51) and (52),

$$\lim_{T \text{ large}} \alpha_{M,p}{}^{(i)}(x,J',y)$$

$$= (R/V_M) |d \ln J_M / dp| (\mu H / kT)^2 (J_M / kT). \quad (59)$$

The normal or positive asymptotic excess expansion coefficient increases rapidly as $1/T^3$ with decreasing temperatures, while the asymptotic anomalous expansion coefficient decreases algebraically as $1/T^2$, according to the asymptotic $1/T^2$ behavior of the free solid exchange heat capacity $C(x)$ [Eq. (49')] to which it is proportional by Eq. (53). It is thus to be expected that below some temperature, the expansion coefficient changes sign. It is needless to add that the preceding discussion refers also to the temperature coefficient of the pressure, $(\partial p / \partial T)_V$, or $(\partial S / \partial V)_T$, the isothermal volume variations of the entropy. The technique evolved by the Florida group[6] for the very-high-resolution pressure-change measurements in the free solid might likely be used effectively in the qualitative and quantitative investigations of the indicated variations in the above derivative thermal properties of the low-pressure magnetized solid He³.

Before entering into a detailed analysis of these derivative properties, we should like to consider the case of strong field magnetizations. Their representation involves the parameter y or $\mu H / kT$ to higher powers than the second which was sufficient at low or moderate fields.

3. SOLID He³ IN MAGNETIC FIELDS OF INCREASING STRENGTH

We saw in Sec. 2 A that an indirect approach allowed one to derive the partition function of the exchange-coupled solid in the presence of magnetic fields of low or moderate strength. At these field strengths, it was justified to take into account the relevant parameter, $(\mu H / kT)$ or y, to lowest order in those terms of the various thermal properties which coupled the external field, or y field, to the internal exchange interactions, the x couplings or x field. At increasing field strengths, the combinatorial problem raised by the derivation of the partition function arising from the Hamiltonian (2) was solved on the basis of its representation as a doubly infinite series in ascending powers of the parameters x and y.[10] According to this formalism, the logarithm of the partition function is given by

$$N^{-1} \ln Z(x,y) = N^{-1} [\ln Z(x) + \ln Z^{(i)}(x,y)], \quad (60)$$

with $\ln Z(x)$ given by Eq. (14), and

$$N^{-1} \ln Z^{(i)}(x,y) = \sum_{s=1} \alpha(s) y^{2s} \sum_{n=0} (-)^n a_n{}^{(s)} x^n, \quad (61)$$

which to lowest order in y, or to order y^2, was included in Eqs. (14). In the limit of $y \to 0$, (60) reduces to (11), the partition function of the free exchange-coupled solid; and in the limit $x \to 0$, the system is an ideal paramagnet of spin $\frac{1}{2}$, whose partition function is $Z_{\text{id}}(y)$, or $(2 \cosh y)^N$, which appeared in (15) and (16) above. It is instructive to rewrite $Z_{\text{id}}(y)$ as

$$N^{-1} \ln Z_{\text{id}}(y) = \ln 2 + \ln \left(1 + \sum_{n=1}^{\infty} \frac{y^{2n}}{(2n)!} \right)$$

$$= \ln 2 + \sum_{s=1}^{\infty} \frac{\varphi(s) y^{2s}}{(2s)!}, \qquad (62)$$

which defines the coefficients $\alpha(s)$ in (61), subject to the requirement of $[N^{-1} \ln Z^{(i)}(x,y)]$ reducing to (62), in the limit $x \to 0$, or that the coefficients $a_0{}^{(s)}$ be equal to unity for all s in (61). Up to $s = 4$, one finds easily the $\varphi(s)$ to be, respectively, 1, (-2), 16, and $(-)$ (16×17). The coefficients $a_n{}^{(s)}$, up to $s = 4$ and $n \leq 10$ have been tabulated for the three cubic structures of spin-$\frac{1}{2}$ systems by Rushbrooke, Baker, and their collaborators.[10] These need not be given here. As noted in Sec. 2 A, the series in ascending powers in x and y are limited to $y \lesssim 1.31$–1.32. For larger y values, and above all for very large y's, it is justified to require that $N^{-1} \ln Z(x,y)$ should reduce, in the limit $x \to 0$, to

$$\ln(2 \cosh y) = y + \ln(1 + e^{-2y}), \quad y \gg 1$$

the large y expansion of $N^{-1} \ln Z_{\text{id}}(y)$.

We have to obtain finally the various strong magnetic field thermal properties of solid He³ to the approximation of the truncated double-series representation of the partition function, Eqs. (60)–(62). The approximate thermal properties will then be evaluated numerically in the next section over temperature and field strength intervals over which their series representations exhibit good convergence. An eventual

direct confrontation of future experimental data with the calculated properties becomes thereby accessible.

It is convenient to rewrite the partition function (61) in terms of the functions

$$\Phi_Z^{(s)}(x) = F^{(s)}(x) - 1$$
$$= \sum_{n=1} (-)^n a_n^{(s)} x^n, \quad s \geq 1 \qquad (63)$$

or

$$N^{-1} \ln Z(x,y)_{y \lesssim 1.31} = \ln(2 \cosh y) + N^{-1} \ln Z(x)$$
$$+ \sum_{s=1} \varphi(s) [y^{2s}/(2s)!] \Phi_Z^{(s)}(x). \qquad (64)$$

With (5), one then at once obtains

$$M(x,y)/N\mu$$
$$= (\partial/\partial y)[N^{-1} \ln Z(x,y)]$$
$$= \tanh y + \sum_{s=1} \varphi(s) [y^{2s-1}/(2s-1)!] \Phi_Z^{(s)}(x). \qquad (65)$$

To lowest order in the external field or y, or with $s=1$, this fractional moment reduces to (7), keeping only the linear term in y in the expansion of $\tanh y$. To lowest order, both in x and in y, the double series in (65) reduces to $-a_1^{(1)} yx$, as found above [Eq. (18)]. In the present case of the body-centered solid He^3, $a_1^{(1)} = 4$.

The strong-field entropy resulting from (64) is then

$$S(x,y)/Nk = (\partial/\partial T)[TN^{-1} \ln Z(x,y)]$$
$$= [S(x)/Nk] + \{[S_{id}(y)/Nk] - \ln 2\}$$
$$+ [S^{(i)}(x,y)/Nk]. \qquad (66)$$

Here, $S(x)$ and $S_{id}(y)$ have been defined through Eqs. (6) and (20), and

$$S^{(i)}(x,y)/Nk = (-) \sum_{s=1} \varphi(s) [y^{2s}/(2s)!] \Phi_S^{(s)}(x),$$
$$\Phi_S^{(s)}(x) = \sum_{n=1} (-)^n (n+2s-1) a_n^{(s)} x^n. \qquad (67)$$

The entropy $S(x)$ of the free or unmagnetized solid will be seen to be reduced on magnetization, as expected, because of the ordering imposed by the applied field. Formally, this reduction arises with the entropy deficit of the asymptotic magnetized ideal system, or $[S_{id}(y) - Nk \ln 2] < 0$, which reduction is then moderated by the increasing disorder of positive entropy of interaction $S^{(i)}(x,y)$, of the two competing x and y fields.

One verifies easily that the entropy change on magnetization $S^{(e)}(x,y)$, the sum of the two y-dependent terms on the right-hand side of (66), is identical with what one obtains through the thermodynamic relation given by the first of Eqs. (8), using the strong-field moment of Eq. (65).

With the more complete entropy, Eq. (66), one is led to an improved form of the temperature derivative of the melting pressure of magnetized solid He^3, at low

enough temperatures where the entropy of the phonon excitations may be neglected. The changes in these temperature derivatives, defined through Eqs. (22)–(25), under the assumption of an approximately temperature-independent volume change at melting ΔV_M, will be obtained on replacing the interaction entropy $S^{(i)}(x,y)$ in those equations by its strong-field form (67). One thus finds that Eq. (23) remains valid here also, together with (24) and (25), wherein $S^{(i)}(x,y)$ is to be taken from (67). On integrating $dP_M(x,y)/dT$, as done above in connection with (23), one obtains the strong-field melting-pressure difference between the states (x,y) and (x_μ,y_μ), the latter referring to the state of the melting-pressure minimum in absence of the field. This state remains unchanged practically on magnetization to all field strengths considered here. Or, rewriting (27),

$$P_M(x,y) - P_M(x_\mu,y_\mu) = [P_M(x) - P_M(x_\mu)]$$
$$+ [P_M^{(e)}(x,y) - P_M^{(e)}(x_\mu,y_\mu)],$$

the change in the melting-pressure difference is, on the indicated temperature integrations,

$$P_M^{(e)}(x,y) - P_M^{(e)}(x_\mu,y_\mu) = (-)(R/\Delta V_M)$$
$$\times [\bar{Q}^{(e)}(T,x,y) - \bar{Q}^{(e)}(T_\mu,x_\mu,y_\mu)] \qquad (68)$$

with

$$\bar{Q}^{(e)}(T,x,y) = T[\ln(\cosh y)$$
$$+ \sum_{s=1} \varphi(s) [y^{2s}/(2s)!] \Phi_Z^{(s)}(x)]. \qquad (69)$$

Again, this result can be obtained through the thermodynamic relation (8), yielding

$$P_M^{(e)}(x,y) - P_M^{(e)}(x_\mu,y_\mu)$$

$$= (-)(\Delta V_M)^{-1} \int_{T_\mu}^T dT \int_0^H [\partial M(x,y)/\partial T] dH$$

$$= (-)(\Delta V_M)^{-1} \int_0^H [M(x,y) - M(x_\mu,y_\mu)] dH$$

$$= (-)(R/\Delta V_M) \left\{ T \int_0^y [M(x,y)/N\mu] dy \right.$$

$$\left. - T_\mu \int_0^{y_\mu} [M(x_\mu,y_\mu)/N\mu] dy_\mu \right\}. \qquad (70)$$

On replacing $M(x,y)/N\mu$ by (65) and integrating over y, one recovers again (68). The two terms on the right-hand side of (68) generalize the expression of the melting-pressure change given above to lowest order in y and x [Eqs. (32) and (33)]. In stronger fields, the melting-pressure change on magnetization is thus equal to the change in the density of the energy of magnetization of the solid over the chosen temperature interval, the density being defined over the volume change at

melting ΔV_M. To lowest order in y and x, (68) and (69) reduce to (30) and (31), respectively.

It is now a simple matter to rewrite the melting-pressure change on magnetization over the here relevant low-temperature range indicated above, where it becomes justified to neglect the magnetization of the liquid phase of the two-phase system. One has, with (68) and (69),

$$P_M(x_2,y_2) - P_M(x_1,y_1)$$
$$= \Delta P_{\mathrm{id}}(y_2,y_1) + \Delta P_M^{(i)}(x_2 y_{2d} x_1,y_1), \quad (68')$$

$$\Delta P_{\mathrm{id}}(y_2,y_1) = -(R/\Delta V_M)$$
$$\times [T_2 \ln(\cosh y_2) - T_1 \ln(\cosh y_1)], \quad (69')$$

$$\Delta P_M^{(i)}(x_2,y_{2d};x_1,y_1)$$
$$= -(R/\Delta V_M)\{T_2 \sum_{s=1} \varphi(s)[y_2^{2s}/(2s)!]\Phi_Z^{(s)}(x_2)$$
$$- T_1 \sum_{s=1} \varphi(s)[y_1^{2s}/(2s)!]\Phi_Z^{(s)}(x_1)\}. \quad (69'')$$

We turn now to the heat capacity and expansion coefficient of solid He³ in stronger fields. One finds with the strong-field entropy (66), using Eqs. (6) and (67), along or near the melting line as well as throughout the paramagnetic range of the low-pressure solid,

$$C(x,y)/Nk = [C(x)/Nk] + (y/\cosh y)^2 + [C^{(i)}(x,y)/Nk]. \quad (71)$$

Here, $C(x)$ was given by Eq. (49'), and

$$C^{(i)}(x,y)/Nk = \sum_{s=1} \varphi(s)[y^{2s}/(2s)!]\Phi_C^{(s)}(x),$$

$$\Phi_C^{(s)}(x) = \sum_{n=1} (-)^n(n+2s)(n+2s-1)a_n^{(s)}x^n \quad (72)$$

is the contribution arising from the entropy of interaction $S^{(i)}(x,y)$ [Eq. (67)]. The second term on the right-hand side of (71) is $C_{\mathrm{id}}(y)$ encountered already above [Eq. (49')].

The strong-field isobaric volume-expansion coefficient is, with the corresponding entropy (66), at or near melting,

$$-V_M \alpha_{M,p}(x,y) = [\partial S_M(x,y)/\partial p]_T$$
$$= R|d \ln J_M/dp|\{[C_M(x)/R]$$
$$+ \sum_{s=1} \varphi(s)[y^{2s}/(2s)!]\Phi_\alpha^{(s)}(x)\},$$

$$\Phi_\alpha^{(s)}(x) = \sum_{n=1} (-)^n n(n+2s-1)a_n^{(s)}x^n. \quad (73)$$

The expansion coefficient in absence of the field, $\alpha_{M,p}(x)$ or $V_M \alpha_{M,p}(x)$, is the negative of the first term on the right-hand side. It was given above [Eq. (53)] and derived earlier.[8] The second term is the excess

expansion coefficient

$$\alpha_{M,p}^{(i)}(x,y) = -(R/V_M)|d \ln J_M/dp|$$
$$\times \sum_{s=1} \varphi(s)[y^{2s}/(2s)!]\Phi_\alpha^{(s)}(x). \quad (74)$$

In the region of convergence of the truncated double series, $\alpha_{M,p}^{(i)} > 0$. Or the anomalous negative expansion coefficient of the unmagnetized solid, at low temperatures, is increased on magnetization and the anomaly is reduced and even eliminated as discussed in the previous section to lowest order of approximation in y. The form (73), as well as (51), of the expansion coefficient clearly shows that the reduction or elimination of the anomalous free solid derivative properties, such as $\alpha_{M,p}(x)$ and $[\partial p(x)/\partial T]_V$, is accelerated at increasing field strengths or at increasing y values. A more complete discussion of these properties will be given in the next section.

4. VARIOUS THERMAL PROPERTIES OF MAGNETIZED SOLID He³

We would like to review finally some of the detailed features of several equilibrium properties of the magnetized solid. One of the most important of these is the entropy $S(T)$, at or near melting. We give in Fig. 1 the (S,T)-entropy diagram at a series of field strengths between 0 and 100 kG, and between 5 and 50m°K. The upper abscissa refers to the x variable with J_M or J taken to be[6] 0.7m°K. The increasingly large depression of the free-solid entropy at melting (top curve) at increasing field strengths is clearly displayed on the diagram. The five field strengths of the magnetized-solid entropy curves refer, respectively, to 20, 40, 60, 80, and 100 kG, in succession below the top no-field entropy curve.

The quantitative evolution of two processes can be directly read off this diagram. One of these refers to the
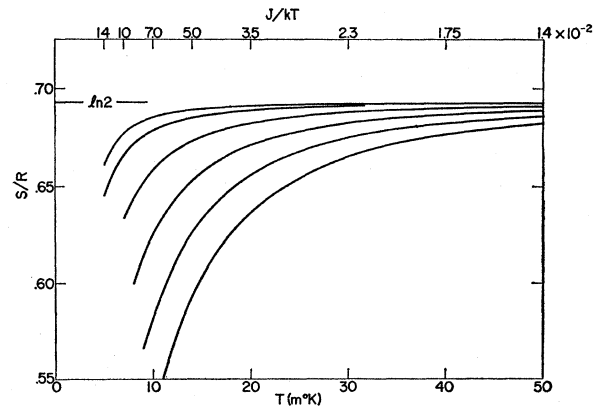

Fig. 1. The entropy of solid He³ at melting as a function of the temperature (m°K) (lower scale) and the parameter J/kT (upper scale). The top curve is the entropy of the unmagnetized solid; the others refer, from the second curve downward, to field strengths of 20, 40, 60, 80, and 100 kG, respectively.

energy or heat to be removed from the solid on isothermal magnetization. This process is where H_i and H_f are the initial- and final-state field strengths, y_i and y_f the corresponding y values, and

$$\Delta Q(x, y_f, y_i) = T[S(x, y_f) - S(x, y_i)]. \quad (75)$$

The relatively small values of the heat of magnetization at the higher temperatures suggest the following procedure. In order to explore the thermal behavior of magnetized solid He³ at low temperatures, the solid should preferably be magnetized at higher temperatures, and cooled afterwards in presence of the uniform constant external field reached in the magnetization process. It should be noted that the presence of the latter field does not need to interfere with the cooling of the magnetized solid, by conduction, with a cold paramagnetic salt which was itself cooled, in an independent process through adiabatic demagnetization in a separate magnetic field arrangement. Clearly, liquid He³-He⁴ dilution refrigerators, when capable of cooling samples of solid He³ down to the relevant temperatures indicated here, $T \ll 80\text{m°K}$, for instance, could contribute significantly to the experimental investigations of magnetized solid He³.

The entropy diagram of Fig. 1 also defines the temperatures reached on adiabatic demagnetization of the solid from an initial state (H_i, T_i, p_i), $T_i \leq 50\text{m°K}$ to a final state (H_f, T_f, p_f), through

$$S(H_f, T_f, p_f) = S(H_i, T_i, p_i),$$
or $\qquad\qquad\qquad\qquad\qquad\qquad\qquad (76)$
$$S(H_f, T_f, V_f) = S(H_i, T_i, V_i).$$

As discussed earlier[8] and seen above, [Eq. (6)], at the low temperatures of interest here, the entropy $S(x)$ of the free solid, arising from its nuclear spin system, on neglecting its very small phonon entropy, depends only on the variable x, that is, $J(p)/kT$ or $J(V)/kT$. In presence of a constant and uniform external field, the entropy becomes a function of both x and y, or $\mu H/kT$, or of T, $J(V)$, and H, where $J(V)$ is independent of H, to the approximation which neglects magnetostrictive or magnetoelastic effects. If, in addition, one neglects the variations of J with volume or pressure, over the melting or near-melting temperature range of Fig. 1, $T < 50\text{m°K}$, by using an approximate average J value, then the entropy diagram yields a correspondingly approximate solution $T_f(S, H_i, H_f)$ of Eq. (76), with $S(H, T, p)$ or $S(H, T, V)$ defined by (66), (67), and (6). The diagram of Fig. 1 refers to the intermediate empirical J value indicated above. In the limiting case of vanishing final field, $H_f \to 0$, or H_f very small, one has, to the stated approximation,

$$S(x_f) = S(x_i, y_i). \quad (77)$$

If T_f, the final temperature reached in this adiabatic demagnetization process, is such that the asymptotic large T approximation of $S(x)$ is still justified, then,

by (6),

$$\lim_{T_f \gg J/k} S(J/kT_f)/R = \ln 2 - \tfrac{3}{2}(J/kT_f)^2, \quad (78)$$

where J refers to the numerical value of the negative exchange energy. Using the exact expression of the entropy of the magnetized solid, given by (6), (66), and (67), which is used in Fig. 1, one obtains, with (78),

$$T_f(x_i, y_i, y_f = 0) = (\tfrac{3}{2}\ln 2)(J/k)$$
$$\times\{1 - [S(J/kT_i; \mu H_i/kT_i)/R\ln 2]\}^{-1/2}. \quad (79)$$

This shows, and the entropy diagram displays it clearly, that the smaller the initial-state entropy $S(x_i, y_i)$ is in comparison with the complete spin entropy of $R\ln 2$, the closer one approaches the lowest temperature allowed by the asymptotic representation of S_f, or

$$\lim\inf T_f = (\tfrac{3}{2}\ln 2)(J/k). \quad (80)$$

The asymptotic relation (79) has been obtained earlier[8] in connection with the approximate final temperature reached in the adiabatic solidification process of He³, starting with the entropy $S_{L,M}(T_i)$ of the liquid at melting and producing the solid of entropy $S_{s,M}(T_f)$ equal to $S_{L,M}(T_i)$. Here, $T_i < T_\mu$, where T_μ is the temperature of the melting-pressure minimum. Clearly, the entropy $S(x_i, y_i)$ of the magnetized solid appearing in (79) and given by (66), (67), and (6) above, has no connection with the entropy $S_{L,M}(T)$ of the liquid at melting, in the present formalism.

We turn now to the discussion of the experimentally accessible variations in the melting-pressure change over some relevant temperature interval, variations arising from the magnetization of the solid at melting. We will neglect here the small effect due to the magnetization of the liquid. This was justified in the previous section over the low temperatures and field strengths of interest here. We give in Fig. 2 the vari-
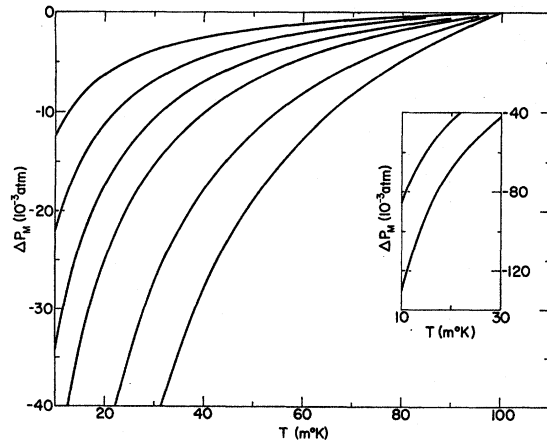

FIG. 2. The magnetic change in the melting-pressure difference $[P_M(T_1, H) - P_M(T < T_1, H)]$ or $\Delta P_M(T, T_1, H)$, in units of 10^{-3} atm, as a function of the temperature at various field strengths H. The reference temperature T_1 is 100m°K; the field strengths are, starting with the top curve, 30, 40, 50, 60, 80, and 100 kG, respectively. The insert gives extended ΔP_M arcs at 80 and 100 kG, down to 10m°K.

ations in the melting-pressure change between 10 and 100m°K, as defined by Eqs. (68'), (69'), and (69''). The graphs give the differences

$$\Delta P_M(x_2, y_2; x_1, y_1) = P_M(x_2, y_2) - P_M(x_1, y_1),$$

$$x = J/kT, \quad y = \mu H/kT, \quad 10 \leq T \leq T_1 = 100\text{m}°\text{K}$$

at constant field strengths H, between 30 and 100 kG. The curves refer, respectively, from the top curve downward, to 30, 40, 50, 60, 80, and 100 kG. The two ΔP_M arcs of the insert are continuations of the 80- and 100-kG-field ΔP_M curves. The large magnetic variations of the melting pressure are clearly displayed by these curves. In terms of the pressure resolution achieved by the Florida group,[6] and provided that some similar if poorer resolution can be attained with the magnetized solid at melting, the measurements of ΔP_M even at field strengths lower than 30 kG should be feasible with good precision even at $T \geq 20$m°K, or at currently accessible temperatures.

The measured ΔP_M values have to be confronted with their theoretical values available through (68') and (69), some of which are given in Fig. 2. At low and moderate field strengths, it is justified to use the y^2 approximation to $\Delta P_M^{(i)}$ [Eq. (69'')] and to write with (31)

$$\Delta P_M(x_2, y_2; x_1, y_1)$$

$$= -(R/\Delta V_M)\left\{[T_2 \ln(\cosh y_2) - T_1 \ln(\cosh y_1)]\right.$$

$$\left. + \left(\frac{\mu H}{k}\right)^2 \sum_{n=1} (-)^n a_n^{(1)}\left(\frac{x_2^n}{T_2} - \frac{x_1^n}{T_1}\right)\right\}. \quad (81)$$

The measured ΔP_M values can be analyzed so as to extract from them the component $\Delta P_M^{(i)}$, the inter-action term on the right-hand side of (81). This term represents the interaction between the ordering imposed by the external field and the opposite ordering arising from the antiferromagnetic spin ordering. This term contributes a positive $\Delta P_M^{(i)}$ component which tends to balance the negative ΔP_{id} term associated with the asymptotic limiting ideal spin system, the square bracket on the right-hand side of (81). It is seen that if T_2 is sufficiently small in comparison with the reference temperature T_1, the high precision reached in the measurements of ΔP_M leads to fairly accurate values of $\Delta P_M^{(i)}$ representable now by

$$\lim_{T \ll T_1} \Delta P_M^{(i)}(T, J, H) = (RT/\Delta V_M)(\mu H/kT)^2$$

$$\times [a_1^{(1)}(J/kT) - a_2^{(1)}(J/kT)^2 + \cdots], \quad (82)$$

where the subscript 2 of T has been omitted. This analysis is thus capable of yielding the value of the coefficient $a_2^{(1)}$, characteristic of the exchange-coupling model, at accessible temperatures. The empirical derivation of $a_2^{(1)}$ is necessary for the determination of

whether or not the assumed exchange-coupling model defined through (1) and (2) is valid in the low-pressure solid He³.

As discussed in detail in Secs. 2A and 3, the ΔP_M measurements are equivalent at low and moderate field strengths to the measurements of the differences in the induced magnetic moments, as given by Eq. (34). The numerical evaluation of the induced magnetic moment of the solid at melting, with the indicated approximate averaged low-temperature value of the exchange energy parameter J, shows that at 100m°K, and even down to about 70m°K, the solid may be said to be an ideal paramagnet. Using (34), one obtains

$$\lim_{y2 \leq 1/4} \Delta P_M(x_2, y_2; x_1, y_1) - M_{id}(y_1)H/2\Delta V_M$$

$$= -M(x_2, y_2)H/2\Delta V_M, \quad (83)$$

with

$$M_{id}(y_1) = N\mu(\mu H/kT_1),$$

which defines at T_2, or $T < T_1$, the moment $M(x, y)$ given by Eq. (7). The experimental determination of $M(x, y)$ at a series of temperatures and field strengths should again enable one to obtain the coefficient $a_2^{(1)}$ of the series representation (7) and to compare it with its exact theoretical value tied to the assumed exchange-coupling model. This method of analysis or the one based directly on the derived $\Delta P_M^{(i)}$ values should be justified up to y values of about $\frac{1}{4}$. At larger y values one must use the more complete formalism of the melting-pressure changes contained in Eqs. (68'), (69'), and (69''), with the series $\Phi_Z^{(s)}(x)$ defined by (63). However, even up to field strengths of 100 kG, if T_1 is taken to be 100 to 70m°K, the T_1 term inside the curly brackets on the right-hand side of (69'') reduces to its ideal limit of $\frac{1}{2}T_1 y_1^2$, allowing one to analyze the ΔP_M data with the series representation referring to T_2. In principle, there appear to be no difficulties in this more elaborate confrontation of the melting-pressure variations with the double-series formalism imposed by the exchange-coupling model and the increasingly large field strengths or large values of the y parameter. However, at these larger field strengths, the derivation of the induced magnetic moments $M(T, J, H)$ from the ΔP_M values requires the formation of the derivatives $d(\Delta P_M)/dH$ or $d(\Delta P_M)/dy$, as shown by Eqs. (70). The formal complexity of both the x and y dependence of the strong-field moments imposes this elaborate approach toward their derivation from the correspondingly complex theoretical representation of the strong-field ΔP_M values.

We turn finally to the discussion of the two derivative thermal properties, heat capacity and expansion co-efficient of the magnetized solid, the formal structure of which has been derived and studied above, in the previous sections. We give in Fig. 3 a series of heat capacities as a function of the temperature, at a series of field strengths $C_{s,M}/R$ at melting, including the heat

capacity of the liquid at melting $C_{L,M}/R$. The bottom solid heat-capacity curve is that of the free solid at melting, and the others refer to 20-, 40-, 60-, 80-, and 100-kG field strengths, respectively. The heat capacity of the liquid at melting was given by us recently[8] to a fair degree of approximation over a wide temperature range. Similarly the free-solid exchange heat capacity at melting was obtained by us recently using though a large-volume estimate of J_M, the exchange energy parameter, of 1.25m°K. As shown earlier,[8] to the approximation of a practically temperature-independent volume change at melting, ΔV_M, the intersection of the solid, $C_{s,M}(T)$, and liquid, $C_{L,M}(T)$, heat capacities at melting locates the temperature T_b of the inflection or bend-over point of the melting-pressure line. The recent Florida J value[6] of 0.7m°K may be taken, approximately, to be representative of an averaged exchange energy over a relevant if reduced temperature interval over which the volume and pressure variations at melting are already small or moderate. With the new and smaller J value, the temperature of the inflection point $T_b(J)$ is depressed from about 10.5m°K associated with the large-volume estimate J of 1.25m°K, to somewhat above 7m°K with the smaller 0.7m°K J value, as noted by the Florida workers.[6] The decrease in melting pressure of solid He[3] on magnetization was shown above to arise from the entropy decrease resulting from the additional order imposed by the external field. This effect is qualitatively similar to an apparent increase of the strength of the internal exchange field through the increase of the parameter J. This is seen at once on the entropy formula (6), which is a decreasing function of J,

$$[\partial S(J/kT)/\partial J]_T < 0. \qquad (84)$$

The apparent increase of J is in turn equivalent to an increase of the spin-ordering temperature. The location of the temperature of the inflection point of the melting pressure $P_M(J/kT)$ is the root T_b of

$$C_{L,M}(T_b) = C_{s,M}(J/kT_b), \qquad (85)$$

to the approximation of a constant ΔV_M. Since $C_{s,M}$ is an increasing function of J, it is seen that

$$dT_b/dJ > 0, \qquad (86)$$

which accounts for the above-mentioned variations of the free solid T_b with J. With the magnetization of the solid being equivalent, in a qualitative sense, to an apparent increase of its exchange parameter J, (86) requires that

$$dT_b/dH > 0. \qquad (87)$$

It seems useful to give an analytical approximation or estimate of the temperature of the bend-over or inflection point of the melting-pressure line of the magnetized solid, which we will call $T_b(J,H)$. This estimate results, with the limitation arising from the assumed constancy of ΔV_M, from a generalization of
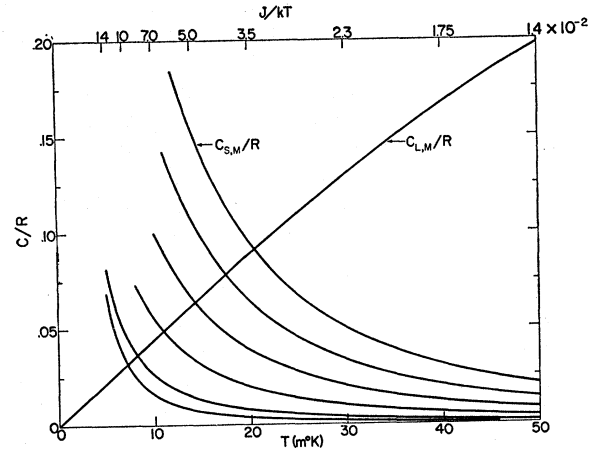

FIG. 3. The heat capacity $C_{L,M}/R$ of liquid He[3] at melting, and the heat capacities $C_{s,M}/R$ of solid He[3] at melting, as a function of the temperature (m°K) (lower scale) and the parameter J/kT (upper scale). The lowest solid heat capacity is that of the unmagnetized solid, the others refer, successively upward, to field strengths of 20, 40, 60, 80, and 100 kG, respectively.

(85) to the magnetized solid. Or,

$$C_{L,M}(T_b) = C_{s,M}(x_b, y_b)$$
$$= C_{s,M}(J_M/kT_b, \mu H/kT_b). \qquad (88)$$

An analytic estimate of $T_b(J_M,H)$ results at once from (88), with the help of the limiting very-low-temperature expression of $C_{L,M}(T)$, or

$$\lim_{T \ll T_{0,M}} C_{L,M}(T)/R = \gamma_{L,M}T, \quad \gamma_{L,M} \sim 4.55°K^{-1}, \qquad (89)$$

where $T_{0,M}$ is the characteristic temperature of the spin system of the liquid at melting, discussed previously by us.[8] The asymptotic high-temperature heat capacity of the magnetized solid at melting, resulting from Eqs. (49) and (50), to lowest order in J_M/kT and $\mu H/kT$, i.e., to order $(J_M/kT)^2$ and $(\mu H/kT)^2$, is

$$C_{s,M}/R \sim [3J^2 + (\mu H)^2]/(kT)^2, \quad J_M/kT \ll 1, \mu H/kT \ll 1. \qquad (90)$$

Hence, by (88),

$$T_b(J_M,H) = T_b(J_M,0)[1 + \tfrac{1}{3}(\mu H/J_M)^2]^{1/3}, \qquad (91)$$

with the free-solid bend-over temperature

$$T_b(J_M,0) = [3(J/k)^2/\gamma_{L,M}]^{1/3}. \qquad (92)$$

The latter estimate yields, with J_M/k of 1.25m°K, and the indicated $\gamma_{L,M}$ value, a T_b of 10.1m°K, while the graphical solution of the exact Eq. (85) gave earlier[8] 10.5–10.6m°K. With J_M/k of 0.7m°K, (92) gives a T_b of 6.86m°K, while the graphical solution is about 7.15–7.20m°K. It should be noted that while $T_b(J_M,0)$ refers to a single asymptotic approximation, the form (91) of $T_b(J_M,H)$ is a superposition of two such approximations. It is to be noted that while $T_b(J_M,0)$ given by (92) yields an approximation of the exact T_b by defect,

$T_b(J_M,H)$, or (91), at low field strengths, is a similar estimate, but becomes an estimate by excess at larger field strengths.

The heat-capacity curves of Fig. 3 show the locations of the inflection points of the melting-pressure line of magnetized solid He³. To the approximation of $C_{L,M}$ and of the approximate evaluation of the Eq. (71) of $C_{s,M}$, these inflection points vary from about 7m°K, for the unmagnetized solid, to 21m°K, at 100 kG. The very large intrinsic values of the heat capacity of the magnetized solid, at accessible field strengths and temperatures, should facilitate its measurements with fair accuracy. Ultimately, accurate heat-capacity measurements might be exploited toward determining some coefficients $a_n{}^{(0)}$ and $a_n{}^{(1)}$ appearing in Eqs. (49), at low or moderate field strengths. At larger field strengths, the correspondingly large heat capacities might become usable in the determination of some coefficients $a_n{}^{(s)}$, $s>1$, according to the heat-capacity formulas (71) and (72). Such an analysis of the measured heat capacities of the magnetized solid could thus contribute to ascertaining whether or not the assumed exchange-coupling model represents the spin system of the solid in a satisfactory way.

We turn finally to the discussion of the isobaric expansion coefficient $\alpha_p(x,y)$, or of the temperature coefficient of the pressure $[\partial p(x,y)/\partial T]_V$ which is equivalent to it. As shown in Secs. 2 and 3, one of the effects of magnetizing solid He³ is to yield a positive component entropy, given by $S^{(i)}(x,y)$ arising from the interaction of the x and y fields. This interaction entropy is given by (21) to order y^2, and at larger field strengths or large y values by (67). This entropy $S^{(i)}$ gives rise, in turn, to a normal or positive component of the isobaric volume expansion coefficient $\alpha_p{}^{(i)}(x,y)$ or the associated temperature coefficient of the pressure $[\partial p^{(i)}(x,y)/\partial T]_V$. Stated in other terms, the entropy $S^{(i)}$ has normal pressure and volume derivatives. At the low temperatures, the free solid is thermally anomalous, having a negative expansion coefficient $\alpha_p(x)$ and a negative temperature coefficient of the pressure, $[\partial p(x)/\partial T]_V$, an early prediction[7,8] fully verified by the Florida group,[6] as mentioned above. On magnetization, the normal magnetic expansion coefficient $\alpha^{(i)}(x,y)$ will reduce the starting anomalous or negative expansion coefficient of the free solid. The derivatives $\alpha_p(x,y)$ or the associated $[\partial p(x,y)/\partial T]_V$, that is, $[\alpha_p(x,y)/\chi_T]$, where χ_T is the isothermal compressibility of the free solid, omitting magnetoelastic effects, given by Eqs. (51) and (73), do not involve the asymptotic ideal spin system. This contrasts with the entropy, melting pressure, magnetic moment, and heat capacity of the magnetized solid which all have components arising with the asymptotic ideal spin system. This is so because the ideal spin system involves only in its description the parameter y or $\mu H/kT$. On the other hand, the properties $\alpha_p(x,y)$ or $[\partial p(x,y)/\partial T]_V$ depend on the derivatives $(\partial \ln J/\partial p)_T$ or $(\partial \ln J/\partial V)_T$, written above as

total derivatives since J is not expected to vary explicitly with T. The other thermal and magnetic properties do not involve the derivatives of J.

Before analyzing more closely the derivatives α_p or $(\partial p/\partial T)_V$ in the magnetized solid, it seems of interest to consider briefly these same properties of the unmagnetized solid, taking into account the averaged Florida[6] J value, at volumes approaching those at very low temperatures near or at melting, instead of the estimated larger J value referring to a larger melting volume used by us recently.[8] With J equal to 0.7m°K, we find the zero of $\alpha_p(x)$ or of $[\partial p(x)/\partial T]_V$ to be at $T\sim0.25$°K, using a probable upper limit estimate of the compressibility of the solid at low temperatures and at melting. Below about 70m°K, where the contributions of the phonon system to these derivative properties may safely be neglected, our recent estimate of

$$(R/V_{s,M})|d\ln J/dp| \lesssim 0.3/°K \qquad (93)$$

corresponds, in terms of the Florida determination[6] of $d\ln J/dV$, to a compressibility of about 5.5×10^{-3}/atm. This appears to be fully compatible with recent determinations of the compressibility of solid He³ along the melting line at higher temperatures due to Straty and Adams.[13]

As a consequence of the competition between $\alpha_p(x)$ and $\alpha_p{}^{(i)}(x,y)$, or $[\partial p(x)/\partial T]_V$ and $[\partial p^{(i)}(x,y)/\partial T]_V$, these derivative properties may be grouped into three classes. At low field strengths, the magnetic components of these derivatives stay numerically smaller than their free-solid component. The anomalous behavior of the latter will not be changed qualitatively by the former, that is on magnetization. At intermediate field strengths and low temperatures, the magnetic component may become larger than the free-solid component so that below some temperature $T_{\alpha,H}$ these derivatives become normal, to become anomalous again at $T>T_{\alpha,H}$, because of the more rapid fall with temperature of the normal magnetic component compared with the slower algebraic increase with temperature of the anomalous free-solid component. Finally, at high enough field strengths, the normal magnetic component may become so large as to ensure, with the contribution of the phonon system, a positive α_p or $(\partial p/\partial T)_V$, at all temperatures.

A series of expansion coefficients, including that of the free solid (bottom curve), are given in Fig. 4. They refer, respectively, to the following field strengths: 20, 40, 60, 80, and 100 kG. The insert gives the expansion coefficients at 45-, 50-, and 55-kG field strengths, showing their positive and negative regions around their zeros. The curves of the expansion-coefficient graph are seen to refer to two of the three classes of expansion coefficients considered above. To the approximation of the present calculations, below about 30 kG, the mag-

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[13] G. C. Straty, thesis, University of Florida, 1967 (unpublished); G. C. Straty and E. D. Adams (to be published).

netized solid He^3 expansion coefficients are anomalous up to their zero arising from the normal phonon contributions, which at 30 kG is about the same as in the absence of the external field. Between about 30 and 100 kG, the expansion coefficient has two zones. The first, at low temperatures, arises from the competition between the two component expansion coefficients of opposite sign discussed above in connection with (73) and (74). The second, at higher temperatures, is due to the compensating effect of the normal phonon expansion coefficient. It would appear that only at field strengths larger than 100 kG would the magnetized solid become completely normal with respect to the pressure and volume variations of its entropy. Equation (51) shows that, at intermediate and accessible field strengths, the technique developed by the Florida group[6] should enable one to analyze the change in the derivative $[\partial p^{(i)}(x,y)/\partial T]_V$ arising from the magnetization of the low-pressure solid He^3 with good accuracy. Since by (51), at moderate field strengths,

$$[\partial p^{(i)}(x,y)/\partial T]_V = \alpha^{(i)}(x,y)/\chi_T$$

$$= -(R/V_M)(d \ln J/dV)(\tfrac{1}{2}y^2)$$

$$\times \sum_{n=1} (-)^n n(n+1)a_n^{(1)}x^n, \qquad (94)$$

as noted already above, χ_T being the isothermal compressibility of the solid, omitting magnetoelastic effects, the determination of at least the first two coefficients $a_1^{(1)}$ and $a_2^{(1)}$ in the x series appears to be feasible. At the present time this approach could be also advantageous, at accessible temperatures, for a verification of the degree of validity of the exchange-coupling model used here to describe the thermal properties of magnetized solid He^3.

In conclusion, it may be expected that experimental investigations of magnetized solid He^3, at or near melting and in statistical thermodynamic equilibrium, will be initiated in due course, at accessible temperatures and magnetic field strengths. Nuclear magnetic susceptibility determinations through measurements of melting-pressure changes on magnetization as well as
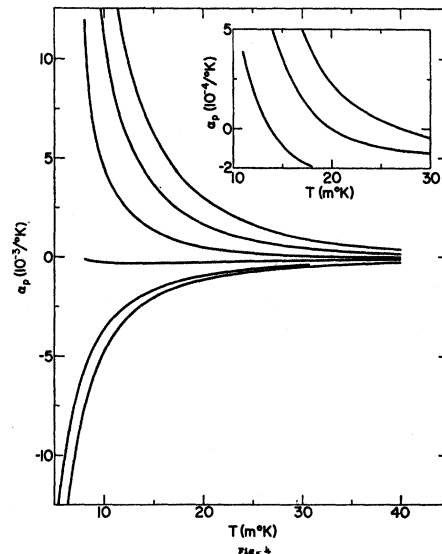

FIG. 4. The approximate isobaric expansion coefficient at melting ($10^{-3}/°K$) of unmagnetized solid He^3 (bottom curve), and of the magnetized solid, as a function of the temperature ($m°K$). The field strengths refer, from the second lowest curve upward, to 20, 40, 60, 80, and 100 kG, respectively. The insert gives short arcs of expansion coefficients around their zeros, from left to right, at 45-, 50-, and 55-kG field strengths, respectively.

the qualitative modification of the anomalous derivative thermal properties of the free solid on magnetization appear to be of particular interest. As shown and discussed above, the suggested experimental studies of the magnetized solid should contribute to the elucidation of the nature and form of interatomic couplings in solid He^3 governed by its nuclear spin system.

ACKNOWLEDGMENTS