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Theory of Paramagnetic Solid Helium Three*

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The object of the present paper is the elaboration of a model formalism for solid He^3. This is based on postulating isotropic first-neighbor antiferromagnetic $-J_1$ and second-neighbor ferromagnetic J_2 exchange interactions between pairs of atoms. Using some of the averaged pressure data along isochores of the unmagnetized and magnetized solid, due to Kirk and Adams, and assuming the exchange-strength functions to have constant logarithmic volume derivatives γ_1 and γ_2, we derive $-J_1(V)$, $J_2(V)$, γ_1, and γ_2; a satisfactory theoretical representation of all the isochore data, restricted to the asymptotic high-temperature range, is thereby achieved. The complete verification of the model formalism, however, can only be performed once data become available on various thermal properties of the solid, in absence and presence of external uniform and constant magnetic fields of moderate strength, at low enough temperatures $T < 10$ mK. Earlier high-temperature nuclear-paramagnetic-susceptibility data of low precision can be accounted for by the model and its parameters at small-volume isochores, but numerical discrepancies develop at the larger volumes. Accurate susceptibility measurements are required for a detailed comparison of theoretical and experimental values of the asymptotic Curie temperatures of the solid along a series of isochores.

I. INTRODUCTION

The present paper is devoted to the elaboration of a theoretical model of solid He^3 in the high-temperature paramagnetic region. Recent experimental work of Kirk and Adams[1,2] on the pressure variations along isochores of the unmagnetized and magnetized body-centered-cubic (bcc) solid has proved that the simplest nearest-neighbor isotropic antiferromagnetic exchange interaction scheme failed to account for the pressure variations along isochores of the magnetized solid calculated earlier.[3] These accurate magnetothermodynamic measurements, however, first established qualitatively the antiferromagnetic character of this solid, as indeed they should, according to the formalism advanced by us.[3] The magnetic isochore measurements suggested, as indicated by Kirk and Adams,[1] that the dominant antiferromagnetic interactions, arising possibly from nearest-neighbor-pair exchange couplings, were opposed by second- or higher-neighbor-pair ferromagnetic exchange interactions.

The formalism of model systems subject to first- and second-neighbor-pair exchange interactions had been established earlier, at high temperatures, by several investigators.[4-6] It appeared interesting to extend the formalism to the case of solid He^3. The experimental data had then to be used, with certain limitations, to derive with them the approximate empirical parametric functions which define the strength of the antiferromagnetic and ferromagnetic pair exchange interactions. The determination of the parametric functions is restricted to the asymptotic high-temperature range of the data. The empirical strength functions could then be used with the formalism to predict the various thermal properties of the model solid at sub-asymptotic temperatures, which range has not as yet been explored experimentally. Comparison of low-temperature data with the predictions of the model could contribute to an improved understanding of the nature of the forces responsible for the anomalous properties of solid He^3 discussed earlier.[7] The magnetic isochore measurements of Kirk and Adams[1] may be said to have already disclosed the increasingly complex aspects of solid He^3.

The present work is restricted to bcc-solid He^3 in its paramagnetic region.

II. SOME HIGH-TEMPERATURE PROPERTIES OF CUBIC SOLIDS WITH FIRST- AND SECOND-NEIGHBOR-PAIR EXCHANGE INTERACTIONS

The nearest-neighbor-pair or first-neighbor-pair isotropic exchange interactions in a system of localized atoms with spin vectors S, with the spin Hamiltonian

$$\mathcal{H}_{\mathrm{I}} = -2J_1 \sum_{i,j} \vec{S}_i \cdot \vec{S}_j , \tag{1}$$

determine the component partition function of the system,

$$Z_S^{\mathrm{I}} = \mathrm{tr}(e^{-\mathcal{H}_{\mathrm{I}}/kT}) . \tag{2}$$

In (1), the (i,j) subscripts refer to first-neighbor pairs alone, with the parameter $J_1 < 0$ in an antiferromagnet. At high temperatures, Z^{I}_S is seen to be expandable into an infinite series of ascending powers of J_1/kT. The combinatorial problem resulting from the spin summations in (1) and (2) has been the subject of many investigations.[8] For our present purposes, it will be justified to consider the various series expansions limited to relatively few terms of powers[9] of J_1/kT. The generalization of (1) to include second-neighbor-pair exchange interactions may be written down at once as

$$\mathcal{H}_{\mathrm{II}} = -2J_2 \sum_{l,m} \vec{\mathrm{S}}_l \cdot \vec{\mathrm{S}}_m , \tag{3}$$

where J_2 is the effective second-neighbor-pair exchange-energy parameter and the l, m subscripts refer to second-neighbor atoms only. In any given crystal structure, the second-neighbor sites define their own specific lattice, secondary to the dominant lattice arising with the first-neighbor sites. In the present system with the component spin Hamiltonian

$$\mathcal{H} = \mathcal{H}_{\mathrm{I}} + \mathcal{H}_{\mathrm{II}} , \tag{4}$$

the associated partition function is

$$Z_S = \mathrm{tr}(e^{-(\mathcal{H}_{\mathrm{I}}+\mathcal{H}_{\mathrm{II}})/kT}) . \tag{5}$$

According to (4), the formalism assigns every atom or spin to two lattices with specific interaction parameters of effective character J_1 and J_2. In the low-pressure or large-volume modification of solid He^3, the two lattices are, respectively, the dominant antiferromagnetic first-neighbor bcc lattice, and the secondary ferromagnetic simple-cubic lattice of the second neighbors, with the effective parameters $J_1 < 0$ and $J_2 > 0$.

In the single-lattice approximation, the partition function (2) leads, at high temperatures, to the infinite-series representation of the heat capacity[9,10]

$$C_{\mathrm{I}}/Nk = \sum_{p\geq 2} (-)^p c_{p0} (J_1/kT)^p . \tag{6}$$

Henceforth J_1 will refer to the numerical value of this parameter. The coefficients c_{p0} depend on S, the coordination number z of the lattice, and on characteristic numerical quantities, the so-called structure constants.[8-10] Correspondingly with (5) one is led, as first shown by Wojtowicz,[4] to the heat-capacity double series

$$C_{\mathrm{II}}/Nk = \sum_{\substack{p,r=0\\ p+r\geq 2}} (-)^p c_{pr} (J_1/kT)^p (J_2/kT)^r , \tag{7}$$

where the p summation, with $r = 0$, refers to the antiferromagnetic first-neighbor bcc lattice, in the present system, and the r summation, with $p = 0$, to the ferromagnetic second-neighbor simple-cubic-lattice heat-capacity components. The mixed coefficients c_{pr}, $p \neq 0$, $r \neq 0$, depend on S, on both lattice coordination numbers z_1 and z_2, as well as on specific structure constants referring to both lattices. It is convenient to rewrite the two-lattice heat capacity (7) in the following form:

$$C_{\mathrm{II}} = C^{(1)}_{\mathrm{I}} + C^{(2)}_{\mathrm{I}} + C^{(1,2)}_{\mathrm{II}} , \tag{8}$$

with

$$C^{(1)}_{\mathrm{I}}/Nk = \sum_{p\geq 2} (-)^p c_{p0} (J_1/kT)^p , \tag{9a}$$

$$C^{(2)}_{\mathrm{I}}/Nk = \sum_{r\geq 2} c_{0r} (J_2/kT)^r \tag{9b}$$

the component single-lattice heat capacities, and

$$C^{(1,2)}_{\mathrm{II}}/Nk = \sum_{\substack{p,r\geq 1\\ p+r\geq 3}} (-)^p c_{pr} \left(\frac{J_1}{kT}\right)^p \left(\frac{J_2}{kT}\right)^r \tag{9c}$$

the mixed heat capacity. Since in the present work we will be concerned with the entropy of the model system at hand, it is convenient to introduce it here. One has with (8) and (9)

$$\begin{aligned} S(J_1, J_2, T)/Nk &= \int_0^T (C_{\mathrm{II}}/Nk)(dT/T) = \left(\int_0^\infty - \int_\infty^T\right)(C_{\mathrm{II}}/Nk)(dT/T) \\ &= \ln(2S+1) - \sum_{p\geq 2} (-)^p (c_{p0}/p)(J_1/kT)^p - \sum_{r\geq 2} (c_{0r}/r)(J_2/kT)^r \\ &\quad - \sum_{\substack{p\geq 1, r\geq 1\\ p+r\geq 3}} (-)^p [c_{pr}/(p+r)] \times (J_1/kT)^p (J_2/kT)^r . \end{aligned} \tag{10}$$

A number of coefficients c_{p0}, c_{0r}, and c_{pr}, $p + r = 3$, 4, and 5, have been derived for all S and various cubic structures. The volume dependence of the entropy (10) is indirect through the parametric functions $J_1(V)$ and $J_2(V)$. Hence, the temperature coefficient of the pressure along the isochore of volume V is

$$\left(\frac{\partial p(J_1,J_2,V,T)}{\partial T}\right)_V=\left(\frac{\partial S}{\partial V}\right)_T=(R/V)\left\{\left(\frac{\partial \ln J_1}{\partial \ln V}\right)_T\sum_{p\geq 2}(-)^p c_{p0}(J_1/kT)^p\right.$$
$$+\left(\frac{\partial \ln J_2}{\partial \ln V}\right)_T\sum_{r\geq 2}c_{0r}(J_2/kT)^r+\sum_{\substack{p,r\geq 1\\ p+r\geq 3}}(-)^p(c_{pr}/p+r)$$
$$\left.\times\left[p\left(\frac{\partial \ln J_1}{\partial \ln V}\right)_T+r\left(\frac{\partial \ln J_2}{\partial \ln V}\right)_T\right]\left(\frac{J_1}{kT}\right)^p\left(\frac{J_2}{kT}\right)^r\right\}. \tag{11}$$

The pressure variations along the isochores V, $p(J_1,J_2,V,T)$, with $p(J_1,J_2,V,T\rightarrow\infty)\rightarrow 0$, are, on integration,

$$p(J_1,J_2,V,T)=(RT/V)\left\{\left(\frac{\partial \ln J_1}{\partial \ln V}\right)_T\sum_{p\geq 2}(-)^p(c_{p0}/p-1)(J_1/kT)^p\right.$$
$$+\left(\frac{\partial \ln J_2}{\partial \ln V}\right)_T\sum_{r\geq 2}(c_{0r}/r-1)(J_2/kT)^r+\sum_{\substack{p,r\geq 1\\ p+r\geq 3}}(-)^p(c_{pr}/p+r)(1/p+r-1)$$
$$\left.\times\left[p\left(\frac{\partial \ln J_1}{\partial \ln V}\right)_T+r\left(\frac{\partial \ln J_2}{\partial \ln V}\right)_T\right](J_1/kT)^p(J_2/kT)^r\right\}. \tag{12}$$

Since the pressure variations along a series of isochores of bcc-solid He^3 have been measured earlier,[11] the way was open to attack the problem of deriving from the data approximate empirical first- and second-neighbor-pair exchange-energy parameters $J_1(V)$ and $J_2(V)$, as well as their logarithmic volume derivatives

$$\gamma_1(V)=\left(\frac{\partial \ln J_1}{\partial \ln V}\right)_T,\quad \gamma_2=\left(\frac{\partial \ln J_2}{\partial \ln V}\right)_T. \tag{13}$$

Once these quantities are available it becomes possible to explore, in some approximation, the isochores of the model over an extended temperature range reaching into the subasymptotic interval. Similarly, the knowledge of $J_1(V)$ and $J_2(V)$, though necessarily approximate, should enable the calculation of approximate solid entropies down to quite low temperatures. This in turn leads to an estimation of the spin-ordering temperature by bracketing it with earlier conjectured transition-temperature or critical entropy values. However approximate, the entropy of the generalized second-neighbor-pair exchange-coupling model should yield new melting pressures which may reasonably be expected to be improvements of those obtained earlier with the more restricted nearest-neighbor-pair coupling model.[7] These latter problems, however, will not be discussed in the present paper, which is devoted to the generalized treatment of the high-temperature bcc solid phase of He^3.

III. PARAMAGNETIC SOLID WITH FIRST- AND SECOND-NEIGHBOR-PAIR EFFECTIVE EXCHANGE INTERACTION IN UNIFORM MAGNETIC FIELDS OF MODERATE STRENGTH

Before proceeding to the calculation and discussion of the various thermal properties of the free unmagnetized model solid given in Sec. II, it is appropriate to generalize them first to the case of the magnetized solid to which we now turn.

In a constant, uniform external magnetic field of low or moderate strength $|\vec{H}|$, the nuclear paramagnetic moment induced in the system is, per atom or per spin,

$$M(J_1,J_2,T,H)=\chi(J_1,J_2,T)H, \tag{14}$$

χ being the paramagnetic susceptibility per spin. In the present model, with $\chi_0(T)$ denoting the limiting high-temperature ideal Curie-Langevin susceptibility per spin, one has[4]

$$\frac{\chi(J_1,J_2,T)}{\chi_0(T)}=1+\sum_{\substack{m,n=0\\ m+n\geq 1}}(-)^m a_{mn}x_1^m x_2^n,$$
$$x_1=J_1/kT,\quad x_2=J_2/kT. \tag{15}$$

This results from the partition function of the magnetized system whose Hamiltonian is

$$\mathcal{H}_H=\mathcal{H}_{\mathrm{I}}+\mathcal{H}_{\mathrm{II}}-g\beta\vec{H}\cdot\sum_i\vec{S}_i, \tag{16}$$

with the first two component Hamiltonians defined by (1) and (3); g is the relevant splitting factor and β the appropriate elementary magnetic dipole moment associated with the spins S. As was the case with the properties considered in Sec. II, the susceptibility ratio (15) may be rewritten in terms of the two separated lattice susceptibilities, the first-neighbor antiferromagnetic bcc lattice and the second-neighbor ferromagnetic simple-cubic lattice, together with the mixed susceptibility,

$$\frac{\chi(J_1,J_2,T)}{\chi_0(T)}=1+\sum_{m=1}(-)^m a_{m0}x_1^m+\sum_{n=1}a_{0n}x_2{}^n+\sum_{m,n\geq 1}(-)^m a_{mn}x_1^m x_2^n\,. \tag{17}$$

A number of single-lattice susceptibility expansion coefficients a_{m0} or a_{0n} have been obtained sometime ago for various spin values S and cubic structures, as well as for several types of exchange interactions, such as the totally anisotropic Ising type, the isotropic Heisenberg type, and their combinations.[6,8-10,12] A number of the mixed coefficients a_{mn} referring to these structures and interactions have also been derived.[4,6,12]

From thermodynamics the molar entropy change of the N-spin system on isothermal magnetization up to a field strength H, of moderate value, is

$$\frac{\Delta S(x_1,x_2,y)}{Nk}=\int_0^{\mu H}\frac{1}{\mu}\left(\frac{\partial M(x_1,x_2,y)}{\partial(kT)}\right)_H d(\mu H)\,, \tag{18}$$

with M given by (14) and (17); here

$$y=\mu H/kT\,,\quad \mu=g\beta S\,. \tag{18a}$$

With

$$\chi_0(T)=\overline{\mu}^2/kT\,,\quad \overline{\mu}^2=(g\beta)^2 S(S+1)/3\,, \tag{18b}$$

one obtains, using (17),

$$\frac{\Delta S(x_1,x_2,y)}{Nk}=-\tfrac{1}{2}y^2\left[1+\sum_{m=1}(-)^m(m+1)a_{m0}x_1^m+\sum_{n=1}(n+1)a_{0n}x_2^n+\sum_{m,n=1}(-)^m(m+n+1)a_{mn}x_1^m x_2^n\right]. \tag{19}$$

To order H^2 or y^2, the total entropy of the magnetized solid is

$$S(x_1,x_2,y)=S(x_1,x_2,y=0)+\Delta S(x_1,x_2,y), \tag{20}$$

with $S(x_1,x_2,y=0)$, the entropy in the absence of a magnetic field, given by (10). Correspondingly, the heat capacity of the magnetized system becomes

$$C(x_1,x_2,y)=C(x_1,x_2,y=0)+\Delta C(x_1,x_2,y)\,, \tag{21}$$

with $C(x_1,x_2,y=0)$ given by (8) and (9), and

$$\frac{\Delta C(x_1,x_2,y)}{Nk}=\tfrac{1}{2}y^2\left[2+\sum_{m=1}(-)^m(m+1)(m+2)a_{m0}x_1^m+\sum_{n=1}(n+1)(n+2)a_{0n}x_2^n+\sum_{m,n=1}(-)^m(m+n+1)(m+n+2)a_{mn}x_1^m x_2^n\right]. \tag{22}$$

With the entropy (20) the temperature coefficient of the pressure along isochores of the magnetized solid becomes

$$\left(\frac{\partial p(x_1,x_2,y)}{\partial T}\right)_{V,H}=\left(\frac{\partial S(x_1,x_2,y)}{\partial V}\right)_T=\left(\frac{\partial p(x_1,x_2,y=0)}{\partial T}\right)_V+\left(\frac{\partial\Delta p(x_1,x_2,y)}{\partial T}\right)_{V,H}, \tag{23}$$

where the first zero-field derivative was given above, Eq. (11), and the second is, by (19), with the definitions of γ_1 and γ_2 given by (13),

$$\left(\frac{\partial\Delta p}{\partial T}\right)_{V,H}=\left(\frac{\partial\Delta S}{\partial V}\right)_{T,H}=-\tfrac{1}{2}(R/V)y^2\left[\gamma_1\sum_{m=1}(-)^m m(m+1)a_{m0}x_1^n+\gamma_2\sum_{n=1}n(n+1)a_{0n}x_2^n+\sum_{m,n=1}(-)^m(m+n+1)\times(m\gamma_1+n\gamma_2)a_{mn}x_1^m x_2^n\right]. \tag{24}$$

The temperature coefficient of the pressure arising from the effect of the applied external field is seen to be normal, or positive, in contrast with the anomalous negative temperature coefficient of the pressure in the absence of a magnetic field. This is so because, over the temperature range of convergence of the series in (24), the contributions of the antiferromagnetic first sum on the right side of (24) are expected to remain dominant and the sign of this derivative is expected to be that of the first term of the antiferromagnetic series, which is negative. The normal-temperature coefficient of the field-dependent pressure $\Delta p(x_1,x_2,y)$ along isochores tends to reduce first the magnitude of the anomalous negative temperature coefficient of the pressure of the unmagnetized system to end up by overcoming the latter completely so as to modify the thermally anomalous unmagnetized system to become of normal thermal properties in presence of the field. This result was already obtained by us earlier in connection with the magnetized nearest-neighbor-pair exchange-coupled antiferromagnetic system.[3]

The pressure variations along isochores of the magnetized system are, on integration of (24) and with $\Delta p(x_1, x_2, y)_{T\to\infty} \to 0$, that is, the second term on the right side of (23),

$$\Delta p(x_1, x_2, y) = \tfrac{1}{2}(RT/V)y^2 \times \left[\gamma_1 \sum_{m=1} (-)^m m a_{m0} x_1^m + \gamma_2 \sum_{n=1} n a_{0n} x_2^n + \sum_{m,n=1} (-)^m (m\gamma_1 + n\gamma_2) a_{mn} x_1^m x_2^n\right]. \tag{25}$$

The resulting pressure variations become then

$$p(x_1, x_2, y) = p(J_1, J_2, V, T) + \Delta p(x_1, x_2, y), \tag{26}$$

with $p(J_1, J_2, V, T)$ given by (12). Again as was the case with (24), in the region of validity of the series in (25), $\Delta p(x_1, x_2, y)$ is seen to increase with increasing temperatures in contrast with the anomalous field-free term $p(J_1, J_2, V, T)$. At low enough temperatures, the magnetic pressure term is expected to overcome the zero-field pressure term.

This achieves the general formulation of those thermodynamic and magnetothermodynamic properties of the present model of paramagnetic solid He^3 which we propose to discuss in some detail in the present paper. Before applying the above formalism, it is necessary to obtain the empirical parametric functions $J_1(V)$ and $J_2(V)$ of the first- and second-neighbor-pair exchange energies. These will be shown to result from the currently available isochore data of Kirk and Adams[1] as well as from earlier isochore data of Adams and his collaborators.[11] We should like to emphasize, however, that the parametric functions $J_1(V)$ and $J_2(V)$ derived here are subject to various limitations. First of all, the experimental errors of the data affect necessarily these functions. The method of derivation, within the limitations of the above model, succeeds only under a restrictive assumption on the volume dependence of these functions. While this assumption appears indispensable for exploiting the still relatively meager isochore data, it could be omitted if simultaneous measurements existed either along isochores or isobars on the free unmagnetized solid heat capacities and the paramagnetic susceptibilities in the asymptotic high-temperature range. This latter approach, requiring a major experimental program, may, however, present specific difficulties preventing the accurate determination of the functional values $J_1(V)$ and $J_2(V)$ without involving their derivatives.

IV. DERIVATION OF THE EFFECTIVE EMPIRICAL EXCHANGE-ENERGY PARAMETERS OF SOLID He^3

At the present time, essentially all the isochore data on free and magnetized solid He^3 refer to the asymptotic high-temperature range. At these temperatures, $p(x_1, x_2, y)$ reduces to

$$\lim_{T\text{ large}} p(x_1, x_2, y) = (RT/V)\{c_{20}\gamma_1 x_1^2 [1 + (c_{02}/c_{20})(\gamma_2/\gamma_1)(x_2/x_1)^2] - \tfrac{1}{2}a_{10}\gamma_1 y^2 x_1 [1 - (a_{01}/a_{10})(\gamma_2/\gamma_1)(x_2/x_1)]\}, \tag{27}$$

keeping only the lowest-power terms in $(1/T)$ in the zero-field series as well as in the factor series of y^2 in the magnetized system, in (26), using (12) and (25). The asymptotic terms of the pressure variations along isochores refer to the lattices of the antiferromagnetic first-neighbor spins and the ferromagnetic second-neighbor spins. At these high temperatures, these two contributions simply add. The relevant lattice expansion coefficients are the following[9]:

$$c_{20} = \tfrac{2}{3}z_1[S(S+1)]^2, \quad c_{02} = \tfrac{2}{3}z_2[S(S+1)]^2,$$
$$a_{10} = \tfrac{2}{3}z_1 S(S+1), \tag{28a}$$
$$a_{01} = \tfrac{2}{3}z_2 S(S+1).$$

Since,

$$z_1 = 8, \quad z_2 = 6, \quad S = \tfrac{1}{2},$$

one has

$$c_{20} = 3, \quad c_{02} = \tfrac{9}{4},$$
$$a_{10} = 4, \quad a_{01} = 3. \tag{28b}$$

It is seen that, with J_1 and J_2 assumed to be independent of the temperature, in the absence of a magnetic field, $y = 0$,

$$\gamma_1 (J_1/k)^2 + (z_2/z_1)\gamma_2 (J_2/k)^2 = \tau^2(V) \tag{29}$$

is the square of a characteristic temperature of the system along an isochore V. If one introduces the restrictive assumption wherein γ_1 and γ_2, the logarithmic volume derivatives of $J_1(V)$ and $J_2(V)$, are constants independent of the volume, or that

$$J_1(V) \propto V^{\gamma_1}, \quad J_2(V) \propto V^{\gamma_2}, \tag{30}$$

then isochore measurements at four distinct volumes supply, in the asymptotic range, four transcendental equations in the four unknowns J_1, J_2, γ_1, γ_2, the exchange energies referring to one of the four volumes. This system of simultaneous equations enables, in principle, the determination of these four quantities subject to the limitations tied to the restrictive assumptions on the volume dependence of $J_1(V)$ and $J_2(V)$, as well as to those referring to the empirical values of $\tau^2(V)$, or, by (27) and (29),

$$\tau^2(V)=p(J_1,J_2,V,T)/[c_{20}(R/VT)], \tag{31}$$

where p, T and V are measured quantities. Before considering the determination of the four indicated unknowns, it appears interesting to discuss a very much simpler idealized problem. In the latter, one attempts to approximate the system through its dominant first-neighbor lattice subject to an apparent fictitious exchange coupling mechanism of antiferromagnetic character. This yields, instead of (29),

$$[J_a(V)/k]^2\left(\frac{\partial \ln J_a(V)}{\partial \ln V}\right)_T=\tau^2(V). \tag{32}$$

Then let V_1 and $V_1+\Delta V_1$ or V_2 refer to two isochores along which $\tau^2(V_1)$ and $\tau^2(V_2)$ have been evaluated from the measurements of $p(J_a(V_1), V_1, T)$ and $p(J_a(V_2), V_2, T)$. One then obtains at once, with (32), the system of two transcendental equations

$$\begin{aligned}&\gamma_a[J_a(V_1)/k]^2=\tau^2(V_1),\\&\gamma_a[J_a(V_1)/k]^2[1+(\Delta V_1/V_1)]^{2\gamma_a}=\tau^2(V_2),\end{aligned} \tag{33}$$

where use was made of the assumption equivalent to (30), according to which

$$J_a(V_1+\Delta V_1)=J_a(V_1)[1+(\Delta V_1/V_1)]^{\gamma_a}. \tag{34}$$

By (32), γ_a is seen to be positive. We denote by J_a the numerical value of the parameter, as we have done consistently with the effective antiferromagnetic nearest-neighbor parameter $J_1(V)$. The system (33) yields at once

$$\gamma_a=\frac{\ln[\tau(V_2)/\tau(V_1)}{\ln(V_2/V_1)}, \tag{35}$$

$$[J_a(V_1)/k]^2=\tau^2(V_1)\frac{\ln(V_2/V_1)}{\ln[\tau(V_2)/\tau(V_1)]}. \tag{36}$$

Of the five isochores measured by Adams and his collaborators,[11] it is convenient to choose four with the largest volumes V_i. The four isochores V_1, V_2, V_3, and V_4 define six pairs of Eqs. (33). It is clear that if the assumptions leading to (33) are valid over the volume range V_4-V_1, and if the $\tau^2(V_i)$ values given by the pressure variations, the solid volumes V_i, and temperatures T had been measured with sufficient accuracy, the six pairs of Eqs. (33) should yield essentially the same $J_a(V_i)$'s and a unique γ_a, regardless of the isochore pairs (V_i, V_j) used to calculate them. Table I gives the values of $\gamma_a(V_i, V_j)$ obtained with the six pairs of experimental isochores.[11] It is seen that while the larger volume isochores, V_2, V_3 and V_4, give rise to the almost constant γ_a of 20.5–20.6, the pairs involving V_1 yield γ_a values ranging from 15.7 to 18.4.

Table II gives the $J_a(V)$ values at the four volumes V_i, as obtained with the indicated isochore pairs (V_i, V_j), as defined by the experimental volumes of Table I. It is seen again that the J_a values obtained with the isochore pairs involving the smallest volume V_1 tend to range over a larger interval. The J_a values at the larger volume isochores are again almost constant.

With regard to the smallest volume isochore V_1, it should be noted that its pressure variations are small compared with those along the larger volume isochores, tending to decrease the precision of their measurements. Furthermore, the restrictive assumption on the constant value of γ_a, while valid approximately at the larger volumes, may become less valid at the small solid volumes. The difficulties raised by the small-volume isochore data foreshadow certain problems in connection with the extended task of deriving $J_1(V)$, $J_2(V)$, γ_1, and γ_2 using simultaneously the four isochores

TABLE I. The logarithmic volume derivatives γ_a of the apparent nearest-neighbor exchange energy $J_a(V)$. The isochore pairs (V_i, V_j) used for the γ_a's refer to the row and column volumes (in units of cm^3/mole) (Data from Ref. 11.)

	V_2	V_3	V_4
	23.32	23.72	24.02
V_1(22.84)	15.7	17.7	18.4
V_2(23.32)	···	20.6	20.6
V_3(23.72)	20.6	···	20.5

TABLE II. Numerical values of the apparent exchange energies $[J_a(V_1)/k]_{V_iV_j}$ mK, obtained with the isochore pairs (V_i, V_j) defined in Table I.

$J_a(V_1)/k$ (mK)	$J_a(V_2)/k$ (mK)	$J_a(V_3)/k$ (mK)	$J_a(V_4)/k$ (mK)
V_2 : 0.294	V_1 : 0.404	V_1 : 0.532	V_1 : 0.676
V_3 : 0.273	V_3 : 0.347	V_2 : 0.493	V_2 : 0.638
V_4 : 0.268	V_4 : 0.347	V_4 : 0.494	V_3 : 0.640

V_i, $i=1,2,3,4$, to which we return now.

Given the four experimental isochores V_i, one obtains with them the quantities $\tau^2(V_i)$ through (31), as

$$\tau^2(V_i)=p(J_1,J_2,V_i,T)/c_{20}(R/V_iT)\,,\quad i=1,2,3,4\,,\quad c_{20}=3\,. \tag{37}$$

These define the simultaneous system of four transcendental equations

$$\gamma_1(V_i/V_1)^{2\gamma_1}[J_1(V_1)/k]^2+\tfrac{3}{4}\gamma_2(V_i/V_1)^{2\gamma_2}\times[J_2(V_i)/k]^2=\tau^2(V_i)\,,\qquad i=1,2,3,4 \tag{38}$$

where, by (29), (z_2/z_1) reduces to $\frac{3}{4}$. These four equations determine, in principle, the four unknowns $J_1(V_1)$, $J_2(V_2)$, γ_1, and γ_2. In the above formulation, the last three equations of the system involve the volume ratios V_i/V_1, $i=2,3,4$, at the high powers $2\gamma_1$ and $2\gamma_2$, as expected from the γ_a values of the equivalent apparent nearest-neighbor-pair model obtained above and first derived by Adams and his collaborators[11] through a somewhat different numerical approach. These expected large exponents, γ_1 and γ_2, suggest that unless the four volumes V_i are known with sufficient accuracy, a satisfactory solution of the system (38) in terms of unique values of the four unknowns may be difficult to achieve.

At the four molar volumes given in Table I, the $\tau^2(V_i)$ values derived from the data were found to be approximately 1.32, 2.48, 5.01, and 8.39 mK^2 for the four isochores, in increasing order of their volumes. The system of four transcendental equations (38) was investigated by my colleague, Myron L. Stein, from the Computer Science Division, using the MANIAC computer. Various procedures with reasonable starting values for the unknowns failed to yield, however, a satisfactory solution. As implied above, this state of affairs was not particularly surprising since the empirical volumes V_i of the various isochores are known, at best, to within a precision of 0.5%, and their ratios to within 1%, a rather optimistic estimate.[13] The apparently relatively small errors on the volumes and the volume ratios affect, however, considerably the factors $(V_i/V_1)^{2\gamma_1}$ and $(V_i/V_1)^{2\gamma_2}$, and with them the solution of the system of equations. We will return below briefly to the problem of the system (38), after consideration of a second approach to the determination of the empirical parametric functions and their logarithmic volume derivatives.

The Kirk-Adams[1] data on the isochores of the magnetized solid can indeed be used effectively with the stated assumption of the volume independence of γ_1 and γ_2 to obtain the latter logarithmic volume derivatives, $J_1(V)$ and $J_2(V)$. However, the magnetic isochore data[1] cannot as yet be normalized in a satisfactory fashion so as to ensure their correct limit behavior $p(J_1,J_2,T\to\infty,H)\to 0$, as was the case with the isochores of the unmagnetized solid. This lack of satisfactory normalization will necessarily affect the functions $J_1(V)$ and $J_2(V)$, as well as γ_1 and γ_2.

The magnetic isochore measurements at a chosen molar volume V of the solid are also performed at this volume in the absence of a magnetic field, enabling one to determine the details of the modification brought about by the applied field. The zero-field data give, as above, at the volume V_i, equations of the type (38) and (37). In presence of a field, and with the help of (28b), (27) yields

$$\begin{aligned}[p(J_1,J_2,V_i,y=0)-p(x_1,x_2,y)]/2(R/V_i)y^2 &=\Delta p(x_1,x_2,y)/2(R/V_i)y^2\\ &=\gamma_1[J_1(V_i)/k]-\tfrac{3}{4}\gamma_2[J_2(V_i)/k]\\ &=T(J_1,J_2,V)\,,\end{aligned} \tag{39}$$

a characteristic temperature. At two volumes, V_i and V_j, one has two independent equations in J_1, J_2, γ_1, and γ_2; or at V_j,

$$T(V_j)=\gamma_1(V_j/V_i)^{\gamma_1}[J_1(V_i)/k]-\tfrac{3}{4}\gamma_2(V_j/V_i)^{\gamma_2}[J_2(V_i)/k]\,. \tag{40}$$

In absence of the field, at these two volumes, one has, with (38),

$$\tau^2(V_i)=\gamma_1[J_1(V_i)/k]^2+\tfrac{3}{4}\gamma_2[J_2(V_i)/k]^2 \tag{41}$$

and

$$\tau^2(V_j)=\gamma_1(V_j/V_i)^{2\gamma_1}[J_1(V_i)/k]^2+\tfrac{3}{4}\gamma_2(V_j/V_i)^{2\gamma_2}[J_2(V_i)/k]^2\,. \tag{42}$$

Equations (39)–(42) enable one, in principle, to obtain the four unknowns of the problem.

The Kirk-Adams[1] data used here to obtain the above system of four transcendental equations referred to $V_i=23.34$ cm^3/mole, at $H=0$ and 70 kG, as well as to $V_j=24.0$ cm^3/mole, at $H=0$ and 40 kG. As was the case with $\tau^2(V_i)$, the characteristic temperatures $T_H(V_i)$ and $T_H(V_j)$ exhibited deviations from constant temperature-independent values, at the various temperatures of the data. This was expected because of the finite precision of the data and the lack of satisfactory normalization of the magnetic isochores. In addition, the data had another type of defect. At a chosen volume, at two field strengths H_a and H_b or two y values y_a and y_b, at the same temperature, one has, by (39),

$$\Delta p(x_1, x_2, y_a)/\Delta p(x_1, x_2, y_b) = y_a^2/y_b^2 = H_a^2/H_b^2 . \tag{43}$$

The empirical Δp ratios satisfied the preceding requirement only marginally, a fact soon recognized by Kirk and Adams.[1] This may be another manifestation of the lack of satisfactory normalization of the magnetized-solid isochore data.

The $\tau^2(V)$ values at $V_i = 23.34$ cm^3/mole and $V_j = 24.0$ cm^3/mole were found to be approximately 2.69 and 7.62 mK^2, respectively. At 70 kG, T(23.34 cm^3/mole) amounted to 3.37 mK, approximately, while T(24.0 cm^3/mole) at 40 kG varied between about 4.02 and 4.60 mK, with a median value of 4.36 mK, approximately.

The solution of the system of four simultaneous equations (39)–(42) has been obtained by Mr. Stein. This time the solution procedure converged fairly rapidly for all three values of the characteristic empirical temperatures $T(V = 24$ cm^3/mole) and $H = 40$ kG given above. Starting values of the unknowns were similar to those used in attempting to solve the system (38) referring to the four isochores in the absence of a magnetic field. We give in Table III the solutions $J_1(V)$, $J_2(V)$, γ_1, and γ_2 referring to 23.34 cm^3/mole. Over the indicated $T(V = 24$ cm^3/mole) range, J_1 and γ_1 are seen to vary moderately, while J_2 and γ_2 show larger changes. These solutions are all of fair accuracy, and we intend to use those referring to the median value of 4.36 mK of $T(V = 24.0$ cm^3/mole).

The question which arose here was to what extent did these solutions, using two isochores of the unmagnetized and magnetized solid, verify the system of four Eqs. (37) referring all to the unmagnetized solid. One finds that with the solutions given in Table III, the differences between the two sides of the Eqs. (37) vary between 2 and 8.3%, which may be termed acceptable. This could further be improved and these percentages reduced by a factor of 0.1 on modifying slightly the molar volumes of the unmagnetized solid appearing in the system (37). These modifications were computed by Stein with the following results: 22.89 instead of 22.84, 23.29 instead of 23.32, 23.73 instead of 23.72, and 24.06 instead of 24.02, all in units of cm^3/mole. The modified system of Eqs. (37) had solutions essentially identical with those given in Table III, referring to $T(V = 24$ cm^3/mole), at 40 kG, of 4.36 mK, approximately. The indicated small modifications of solid volumes are entirely within the present accuracy of molar volume measurements of the free unmagnetized solid.[13]

We mentioned briefly that an experimental approach is available wherein asymptotic heat-capacity and paramagnetic susceptibility measurements, along the same isochores, enable the empirical determination of $J_1(V)$ and $J_2(V)$ without involving γ_1 and γ_2. One has, indeed, in the asymptotic range,

$$\lim_{T\,\text{large}} C/Nk = c_{20}(J_1/kT)^2 + c_{02}(J_2/kT)^2 , \tag{44}$$

and with (15),

$$\lim_{T\,\text{large}} \chi(J_1, J_2, T)/\chi_0(T) = 1 - a_{10}(J_1/kT) + a_{01}(J_2/kT) , \tag{45}$$

with the coefficients given by (28). In the asymptotic range, the susceptibility ratio may be rewritten as

$$\lim_{T\,\text{large}} \chi/\chi_0 = (1 - \theta_p/T)^{-1} , \tag{46}$$

with

$$\theta_p = -a_{10}(J_1/k)[1 - (a_{01}/a_{10})(J_2/J_1)] \tag{47}$$

the asymptotic paramagnetic Curie temperature, which is negative in the present case, J_1 referring to the numerical value of the nearest-neighbor-pair exchange energy and anticipating here the factor in the parentheses to be positive.

The limiting paramagnetic Curie point θ_p is seen to be decreased numerically from its value in the limit of $J_2 \to 0$, or it is increased algebraically. The actual spin-ordering temperature or Néel point, T_N, is accessible approximately through either the mean-field theory[14] or through a formalism[6] based on assuming a power-law approach, $(T + T_N)^{-\gamma}$ of the susceptibility, for instance, toward the critical transition tempera-

TABLE III. Numerical values of the parametric functions $J_1(V)$ and $J_2(V)$, $(\partial \ln J_1/\partial \ln V)_T$ or γ_1, and $(\partial \ln J_2/\partial \ln V)_T$ or γ_2, at the isochore of volume $V = 23.32$ cm^3/mole of bcc-solid He^3. The three characteristic temperatures of the second isochore, T(24.0 cm^3/mole), are possible choices allowed by the ischore data of Kirk and Adams. As discussed in the text, the above quantities refer to solutions of a simultaneous system of transcendental equations, (39)–(42), formed with the experimental isochores at the indicated volumes, with each isochore defining a pair of equations, in the presence and absence of an external magnetic field.

T (24.0 cm^3/mole) (mK)	J_1/k (mK)	J_2/k (mK)	γ_1	γ_2
4.02	0.368	0.143	16.9	26.4
4.36	0.362	0.154	17.2	24.8
4.60	0.357	0.161	17.4	23.7

ture T_N. While the discussion of the approximate determinations of T_N is outside the subject matter of the present paper, the following remarks appear to be of interest here. The mean-field approach may considerably overestimate T_N. There is a qualitative similarity between the two indicated approaches toward the calculation of T_N. Quantitatively, the power-law formalism should yield better approximations to the actual Néel points than the mean-field theory over most of the range of the J_2/J_1 ratios relevant in the present model of bcc-solid He^3.

The system of the two simultaneous equations (44) and (45), over their range of validity and for the same isochore V, determines $J_1(V)$ and $J_2(V)$. The volume dependence of these exchange energies becomes accessible through the rather laborious approach of the determinations of J_1 and J_2 through measurements of heat-capacity and susceptibility ratios over a series of isochores $V_1, V_2, \ldots, V_n$. The asymptotic susceptibility ratios (45), or (46), will have to be measured with great precision in order to detect reliably with them their deviations from unity. The precision and accuracy required for a satisfactory determination of θ_p, Eq. (47), may be quite high for its successful use in combination with the heat capacity for a derivation of acceptably approximate $J_1(V)$ and $J_2(V)$ values.

In attempting to assess the approximations involved in the here-obtained $J_1(V)$ and $J_2(V)$ parametric functions resulting from the isochore data, one must keep in mind the limitations of the model, the assumed constant values of γ_1 and γ_2, the logarithmic volume derivatives of $J_1(V)$ and $J_2(V)$, and the limitations arising from the finite precision of the data combined with the fairly serious lack of satisfactory normalization of the magnetic isochore data. The general failure of the data to satisfy acceptably the requirements expressed by Eq. (43) cannot but introduce discrepancies between the experimental pressures and their theoretically derived values. To some limited extent, the $J_1(V)$ and $J_2(V)$ functions derived here must be termed as being of tentative character. They are to be tested through new measurements of various thermal and magnetothermal properties of bcc-solid He^3 in the subasymptotic temperature range. We turn now to the calculation of some of these properties.

V. VARIOUS THERMAL PROPERTIES OF PARAMAGNETIC SOLID He^3 ACCORDING TO THE FIRST- AND SECOND-NEIGHBOR-PAIR ISOTROPIC EXCHANGE INTERACTION MODEL

In order to calculate the various thermal properties of the solid model, one must use polynomial approximations to their formal infinite-series representations given in Sec. III. For our present purposes, it will suffice to limit the polynomials of the heat capacity and entropy to terms up to $1/T^4$. The coefficients of these polynomials defined through Eqs. (7)–(9) are thus c_{p0}, with p equal to 2, 3, and 4, referring to the nearest-neighbor antiferromagnetic bcc lattice. The c_{0r} coefficients refer to the ferromagnetic second-neighbor simple-cubic lattice, $r=2,3,4$. The mixed coefficients c_{pr} needed here are such that $p, r=1,2,3$. These coefficients are as follows:

$$\begin{aligned}
&c_{20}=3\,, \quad c_{30}=-3\,, \quad c_{40}=\tfrac{21}{4}\\
&c_{02}=\tfrac{9}{4}\,, \quad c_{03}=-\tfrac{9}{4}\,, \quad c_{04}=-\tfrac{81}{16}\,,\\
&c_{21}=27\,, \quad c_{31}=-18\,, \quad c_{12}=c_{13}=0\,,\\
&c_{22}=63\,.
\end{aligned} \tag{48}$$

General expressions for the single-lattice coefficients c_{p0} or c_{0r} in cubic structures were given up to p or r equal to 7 by Rushbrooke and Wood[9] for any S. For $S=\frac{1}{2}$, Baker *et al.*[10] gave numerical values of c_{p0} or c_{0r} up to p or r equal to 10. General expressions for the mixed coefficients of cubic structures for any spin S were first given by Wojtowicz[4] up to $p+r=5$. It is convenient, however, to rearrange terms in the polynomial approximations of the various series so as to group together all terms of the same power in $1/T$. The coefficients of these polynomials now become functions of $J_1(V)$ and $J_2(V)$. A number of these latter coefficients of the free-energy and paramagnetic susceptibility series of the isotropic second-neighbor-pair ferromagnetic exchange interaction model for spin-$\frac{1}{2}$ systems were given earlier for cubic lattices.[5,6] For cubic and some planar lattices, a number of these coefficients were also given for both isotropic and anisotropic exchange interactions.[12] Up to the indicated power in $1/T$, the generalized coefficients, depending on J_2/J_1 as given earlier,[5] are identical with those calculated here, if account is taken of the negative sign of this ratio in the coefficients given earlier.[5,15]

The present polynomial approximation to the heat capacity series is

$$\begin{aligned}
&C(J_1, J_2, V, T)/R=3[J_1(V)/kT]^2\Gamma(J_1, J_2, T)\,,\\
&\Gamma(J_1, J_2, T)=C_0+C_1(J_1/kT)+\tfrac{7}{4}C_2(J_1/kT)^2\,.
\end{aligned} \tag{49}$$

We recall that $J_1(V)$ refers to the numerical value of the first-neighbor-pair isotropic antiferromagnetic exchange parameter. One has

$$\begin{aligned}
&C_0=1+\tfrac{3}{4}\alpha^2\,, \quad C_1=1+9\alpha-\tfrac{3}{4}\alpha^3\,,\\
&C_2=1+\tfrac{24}{7}\alpha+12\alpha^2-\tfrac{27}{28}\alpha^4\,, \quad \alpha=J_2/J_1\,.
\end{aligned} \tag{50}$$

The heat-capacity high-temperature series in terms of the generalized coefficients is given in the form[5]

$$C/R=\sum_{m=2} (-)^m m(m-1)e_m(J_1/kT)^m/2^m m!\,. \tag{51}$$

One has thus

$$e_2=12C_0,\quad e_3=24C_1,\quad e_4=168C_2\,. \tag{52}$$

The entropy in the absence of a magnetic field is, up to $(1/T)^4$,

$$\begin{aligned}S(J_1,J_2,V,T)/R&=\ln 2-\tfrac{3}{2}(J_1/kT)^2 s(J_1,J_2,T),\\ s(J_1,J_2,T)&=C_0+\tfrac{2}{3}C_1(J_1/kT)+\tfrac{7}{8}C_2(J_1/kT)^2\,.\end{aligned} \tag{53}$$

In the limit $\alpha\to 0$, (49) and (53) are seen to reduce to the polynomial approximations of these properties for the nearest-neighbor-pair isotropic exchange-coupling model.

The method of probing into the detailed physical behavior of bcc-solid He^3 through isochore measurements may be expected to be extended into the subasymptotic temperature range. It thus appears of interest to give several items of the model-isochore-property series. One finds with the series given in Sec. II, Eq. (11),

$$\begin{aligned}\left(\frac{\partial p}{\partial T}\right)_V&=\left(\frac{\partial S}{\partial V}\right)_T\\ &=(-)3(R/V)\gamma_1(J_1/kT)D(J_1,J_2,T),\end{aligned} \tag{54}$$

with

$$D=D_0+D_1(J_1/kT)+\tfrac{7}{4}D_2(J_1/kT)^2\,; \tag{55}$$

$$\begin{aligned}D_0&=1+\tfrac{3}{4}(\gamma_2/\gamma_1)\alpha^2,\\ D_1&=1+3[(2\gamma_1+\gamma_2)/\gamma_1]\alpha-\tfrac{3}{4}(\gamma_2/\gamma_1)\alpha^2,\\ D_2&=1+\tfrac{6}{7}[(3\gamma_1+\gamma_2)/\gamma_1]\alpha\\ &\quad+6[(\gamma_1+\gamma_2)/\gamma_1]\alpha^2-\tfrac{27}{28}(\gamma_2/\gamma_1)\alpha^4\,.\end{aligned} \tag{56}$$

It is seen that the coefficients D_i reduce to the C_i's, Eqs. (50), in the limit of $\gamma_2\to\gamma_1\to 1$.

On temperature integration of (54), and with $p(V,T\to\infty)\to 0$, one obtains the pressures along isochores in the above polynomial approximation. One finds,

$$\begin{aligned}p(J_1,J_2,V,T)&=3(R/V)(J_1/k)\gamma_1 x_1 D_p(x_1,\alpha),\\ D_p&=D_0+\tfrac{1}{2}D_1x_1+\tfrac{7}{12}D_2x_1^2,\quad x_1=J_1/kT\,.\end{aligned} \tag{57}$$

The asymptotic high-temperature forms of the heat capacity, entropy, and pressure along isochores are seen to reduce to the following expressions:

$$\begin{aligned}&\lim_{T\text{ large}} C/R=3C_0x_1^2,\\ &\lim_{T\text{ large}} S/R=\ln 2-\tfrac{3}{2}C_0x_1^2,\\ &\lim_{T\text{ large}} p=3(R/V)(J_1/k)\gamma_1D_0x_1\,.\end{aligned} \tag{58}$$

The characteristic $1/T^2$ behavior of the asymptotic heat capacity and entropy, as well as the $1/T$ behavior of the asymptotic pressure, are seen to appear in (58). Inasmuch as the coefficients C_i and D_i are positive over the volume range of interest here, $V\lesssim 24\ \mathrm{cm}^3/\mathrm{mole}$, the heat capacity and pressure along isochores at subasymptotic low temperatures increase over and above their asymptotic values.

We turn now to the calculation of the thermal properties of the magnetized solid. The paramagnetic susceptibility ratio will be given up to terms in $1/T^4$. The relevant a_{m0}, a_{0n}, and a_{mn} coefficients of the series (17) are as follows:

$$\begin{aligned}&a_{10}=4,\quad a_{20}=12,\quad a_{30}=\tfrac{104}{3},\quad a_{40}=\tfrac{575}{6},\\ &a_{01}=3,\quad a_{02}=6,\quad a_{03}=11,\quad a_{04}=\tfrac{165}{8},\\ &a_{11}=24,\quad a_{12}=84,\quad a_{21}=105,\quad a_{13}=232,\\ &a_{31}=391,\quad a_{22}=535.5\,.\end{aligned} \tag{59}$$

On rearranging the polynomial approximation to the susceptibility-ratio series (17) one obtains

$$\chi/\chi_0=1-4k_1x_1+12k_2x_1^2-\tfrac{104}{3}k_3x_1^3+\tfrac{575}{6}k_4x_1^4, \tag{60}$$

with the k_i coefficients given as follows:

$$\begin{aligned}&k_1=1-\tfrac{3}{4}\alpha,\quad k_2=1-2\alpha+\tfrac{1}{2}\alpha^2,\\ &k_3=1-\tfrac{315}{104}\alpha+\tfrac{63}{26}\alpha^2-\tfrac{33}{104}\alpha^3,\\ &k_4=1-\tfrac{2346}{575}\alpha+\tfrac{628.2}{115}\alpha^2\\ &\qquad-\tfrac{1392}{575}\alpha^3+\tfrac{99}{460}\alpha^4\,.\end{aligned} \tag{61}$$

The susceptibility ratio (60) is found to be identical with the one given directly earlier[5] up to terms in T^{-4}, and written as

$$\chi/\chi_0=1+\sum_{m=1}(-)^m j_m x_1^m/2^m m!\,. \tag{62}$$

On identification, the connection between the j_m's and the k_i's are found to be as follows, using $-\alpha$ for α in the j_m's:

$$j_1=8k_1,\quad j_2=96k_2,\quad j_3=1664k_3,\quad j_4=36\,800k_4\,. \tag{63}$$

The polynomial approximation to the entropy change $\Delta S(x_1,x_2,y)$ given by the series representation (19) may be rearranged to yield

$$\Delta S/R=-\tfrac{1}{2}y^2\sigma(x_1,\alpha), \tag{64}$$

with

$$\sigma = 1 - 8k_1x_1 + 36k_2x_1^2 - \tfrac{416}{3}k_3x_1^3 + \tfrac{2875}{6}k_4x_1^4 . \qquad (65)$$

The corresponding heat-capacity change is then

$$\Delta C(x_1, x_2, y)/R = T\frac{d(\Delta S/R)}{dT}$$

$$= y^2 c(x_1, \alpha) , \qquad (66)$$

with

$$c(x_1, \alpha) = 1 - 12k_1x_1 + 72k_2x_1^2 - \tfrac{1040}{3}k_3x_1^3 + \tfrac{2875}{2}k_4x_1^4 . \qquad (67)$$

The polynomial approximation to the magnetic term of the temperature coefficient of the pressure, Eq. (24), will be seen to take on the following form after rearrangement:

$$\left(\frac{\partial(\Delta p)}{\partial T}\right)_V = 4(R/V)\gamma_1 x_1 y^2 P(x_1, \alpha) , \qquad (68)$$

with

$$P(x_1, \alpha) = P_0 - 9P_1x_1 + 52P_2x_1^2 - \tfrac{2875}{12}P_3x_1^3 , \qquad (69)$$

and

$$\begin{aligned} P_0 &= 1 - \tfrac{3}{4}(\gamma_2/\gamma_1)\alpha , \\ P_1 &= 1 - [(\gamma_1 + \gamma_2)/\gamma_1]\alpha + \tfrac{1}{2}(\gamma_2/\gamma_1)\alpha^2 , \\ P_2 &= 1 - \tfrac{105}{104}[(2\gamma_1 + \gamma_2)/\gamma_1]\alpha \\ &\quad + \tfrac{21}{26}[(\gamma_1 + 2\gamma_2)/\gamma_1]\alpha^2 - \tfrac{33}{104}(\gamma_2/\gamma_1)\alpha^3 , \\ P_3 &= 1 - \tfrac{1173}{1150}[(3\gamma_1 + \gamma_2)/\gamma_1]\alpha + \tfrac{314.1}{115}[(\gamma_1 + \gamma_2)/\gamma_1]\alpha^2 \\ &\quad - \tfrac{348}{575}[(\gamma_1 + 3\gamma_2)/\gamma_1]\alpha^3 + \tfrac{99}{460}(\gamma_2/\gamma_1)\alpha^4 . \end{aligned} \qquad (70)$$

It is seen that the coefficients P_i reduce to the coefficients k_i, Eqs. (61), in the limit $\gamma_1 \to \gamma_2 \to 1$.

The magnetic pressure decrease along isochores given by the series (25), when rewritten in the polynomial approximation takes on the following form:

$$\Delta p(V, T, H) = -2(R/V)(J_1/k)\gamma_1 y^2 Q(x_1, \alpha) , \qquad (71)$$

with

$$Q(x_1, \alpha) = P_0 - 6P_1x_1 + 26P_2x_1^2 - \tfrac{575}{6}P_3x_1^3 . \qquad (72)$$

This achieves the formulation of the various thermal properties of the free and magnetized solid to the approximation of the model and within the limitations of the polynomial representation of these properties, to terms quadratic in the magnetic field strength. We turn now to the numerical evaluation of these properties, performed by Stein, with the approximate empirical parametric functions here obtained, $J_1(V)$ and $J_2(V)$, and their logarithmic volume derivatives γ_1 and γ_2, restricted by assumption to be constant.

The pressure variations $p(J_1, J_2, V, T)$ and $p(J_1, J_2, V, T, H)$ were calculated both at asymptotic and subasymptotic temperatures using (17), (67), and their superposition defined by (26). Comparison of the data with the calculated asymptotic pressure variations is displayed on Figs. 1–3. The three graphs refer to the three currently available sets of isochore measurements in presence of a magnetic field. As stated in Sec. IV, we used the averaged data points at 70 kG, at 23.34 cm^3/mole, Fig. 1, through the characteristic temperature T(23.34 cm^3/mole), as well as the zero-field characteristic temperature τ^2(23.34 cm^3/mole). It is seen that at this molar volume, there is satisfactory agreement between the theoretical pressure curves and the data. The discrepancies may be attributed to

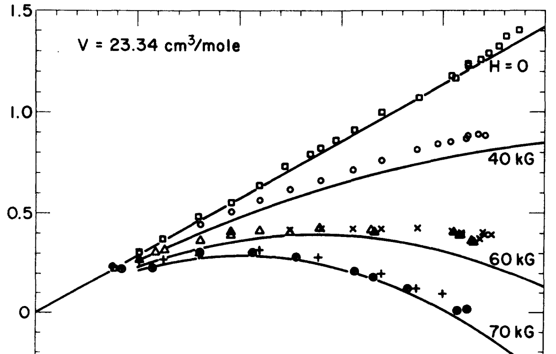

FIG. 1. Theoretical asymptotic high-temperature pressure variations, 10^{-3} atm units, as a function of the reciprocal temperature, in K^{-1} units, along the isochores of volume $V = 23.34$ cm^3/mole in free and magnetized bcc-solid He^3, at the indicated field strength H, in kG units. The data points are those of Kirk and Adams, Refs. 1.

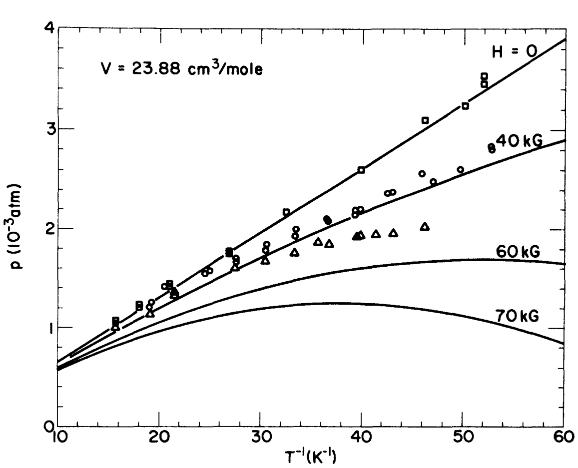

FIG. 2. Same as Fig. 1 at $V = 23.88$ cm^3/mole. The data refer to $H = 0$, 40, and 60 kG.

the failure of the magnetic isochore data to satisfy the important field-normalization condition (43).

At the intermediate volume of 23.88 cm^3/mole, Fig. 2, whose data points are not involved in the determination of the J functions and the γ's, the discrepancies at 60 kG are seen to be relatively large. Again, this indicates, to a certain extent, failure of the data to satisfy the field-ratio condition (43). The zero-field experimental points appear well represented by the calculated $1/T$ function (58).

Finally, at 24 cm^3/mole, where use was made in deriving the J's and γ's of the 40-kG averaged data through the characteristic temperature T(24 cm^3/mole), the 40-kG data points are well accounted for by the calculations. However, at 60 kG, at the lower temperatures, discrepancies seem to develop. This again must be due in part to the failure of the data to satisfy the field-ratio condition (43). However, the present model with the derived J functions and the derived γ's can only be judged in part through future data at subasymptotic temperatures, $T \leqslant 10$ mK, for instance. The calculation of the pressures at the lower temperatures may be performed without difficulties, to the given order in $1/T$, through the formulation given above.

As shown in the earlier discussion of the nearest-neighbor-pair exchange interaction model,[3] the competition between spin ordering arising from the antiferromagnetic interspin couplings and the ordering process imposed by the applied external magnetic field reappears also in the present generalized model. This competition manifests itself through the external field imposing normal thermal behavior on the pressures along isochores, which are anomalous in the absence of a magnetic field, since they decrease on temperature increase. The temperature coefficient of the pressure is negative, i.e., $(\partial p/\partial T)_V < 0$, or, equivalently, isothermal volume increase is accompanied by entropy decrease, $(\partial S/\partial V)_T < 0$, or isothermal compression leads to entropy increase, $(\partial S/\partial p)_T > 0$. In the asymptotic temperature range, the system becomes normal at temperatures $T < T_p$, T_p being the temperature where the anomalous temperature derivatives $[\partial p(V, T, H)/\partial T]_{V,H}$ vanish. By (23), (54), and (67),

$$\begin{aligned}\lim_{T\,\text{large}} &[\partial p(V, T, H)/\partial T]_{V,H} \\ &= (\partial S/\partial V)_{T,H} \\ &= (-)3(R/V)\gamma_1(J_1/kT)^2 D_0 \\ &\times\left[1-\tfrac{4}{3}\left(\frac{y^2}{(J_1/kT)}\right)(P_0/D_0)\right],\end{aligned} \tag{73}$$

where D_0 and P_0 are given by (56) and (69). Explicitly, at asymptotic temperatures,

$$\begin{aligned}T_p = {} & \tfrac{4}{3}(\mu H/k)^2 \\ & \times(J_1/k)^{-1}[1-\tfrac{3}{4}(\gamma_2/\gamma_1)\alpha]/[1+\tfrac{3}{4}(\gamma_2/\gamma_1)\alpha^2]\ ,\end{aligned} \tag{74}$$

which is quadratic in the applied field strength at moderate field strengths. At a constant field strength then,

$$T_p(H_1)/T_p(H_2) = (H_1/H_2)^2\ . \tag{75}$$

The measured pressures along the isochore of smallest volume, $V = 23.34$ cm^3, at 70 kG, clearly exhibit $T_p(H)$ through the maximum of the pressure in the $(p, 1/T)$ representation.

Always in the asymptotic range, one has, by (26), (54), and (70),

$$\begin{aligned}\lim_{T\,\text{large}} [p(J_1, J_2, V, T, H) &= 3(R/V)(J_1/k)\gamma_1(J_1/kT)D_0 \\ &\times\{1-\tfrac{2}{3}[(\mu H)^2/J_1(kT)](P_0/D_0)\},\end{aligned} \tag{76}$$

which is seen to change sign at temperatures $T < T_{p,0}$, with

$$T_{p,0} = \tfrac{2}{3}(\mu H/k)^2[1/(J_1/k)](P_0/D_0) \tag{77}$$

the zero of the pressure $p(J_1, J_2, V, T, H)$. In this asymptotic range,

$$T_{p,0}(H)/T_p(H) = \tfrac{1}{2}\ . \tag{78}$$

At the lower field strengths, both T_p and $T_{p,0}$ recede rapidly, quadratically in the field strength, toward lower temperatures.

The existence of the characteristic temperature $T_{p,0}$, the zero of the pressures along isochores

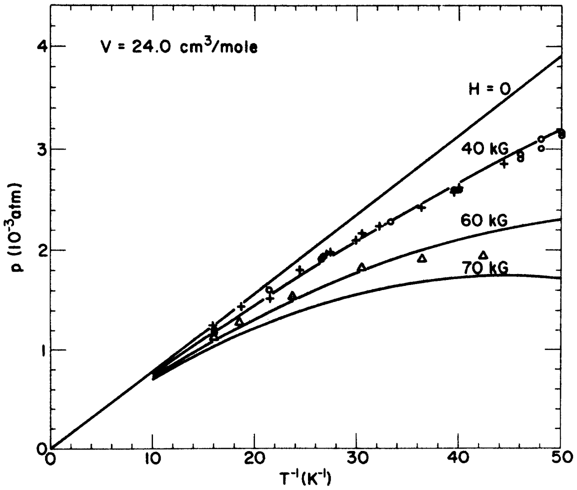

FIG. 3. Same as Figs. 1 and 2, at $V = 24.0$ cm^3/mole. The actual data refer to $H = 40$ and 60 kG.

of the magnetized solid, should help for purposes of normalization of the measurements at the higher field strengths. At this temperature, the pressure variations arising from the exchange interactions, in absence of magnetic field, are exactly compensated by the pressure variations of opposite sign arising from the effect of the external magnetic field, as clearly exhibited formally by (76), referring to the asymptotic temperature range. This compensation effect is, of course, a general feature of the pressure variations along isochores of the magnetized solid.

Our concluding topic of discussion refers to the paramagnetic susceptibility of bcc-solid He^3. The measurements along isochores by Kirk *et al.*[16] yielded the asymptotic paramagnetic Curie temperatures at four solid volumes, all negative as expected. The θ_p values, in mK units, derived from the asymptotic high-temperature susceptibilities, were found to be as follows: -2.9 ± 0.7, -1.3 ± 0.3, -0.48 ± 0.16, and -0.44 ± 0.30, at the respective volumes of 24.0, 23.1, 22.0, and 21.0 cm^3/mole. The θ_p values derived by Sites et al.,[17] at a pressure of about 34 atm, may be associated approximately with a solid volume of about 24.0–24.2 cm^3/mole. This θ_p is given as -2.7 ± 0.3 mK. With the here-obtained $J_1(V)$, $J_2(V)$, and $\alpha(V)$, and γ_1 and γ_2, which parameters give an acceptable account of the pressure variations, in the asymptotic temperature range, along several isochores in the absence and presence of a magnetic field, the calculated θ_p values differ significantly from those derived from susceptibility measurements. Using Eqs. (47), with the approximate $J_1(V)$'s and $J_2(V)$'s, one finds at the volume indicated by Kirk *et al.*,[17] in order of decreasing volumes: -1.42, -0.85, -0.42, and -0.20 mK. It is seen that while at the smaller volumes the experimental θ_p values of rather low precision are compatible with the calculated θ_p values, the discrepancies at the two largest volumes are quite large. It may be hoped that new and more precise susceptibility measurements along isochores will be performed in due course, enabling a detailed discussion of the data in the light of the model and its parameters advanced in the present work.[18]

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[1] W. P. Kirk and E. D. Adams, Phys. Rev. Lett. 27, 392 (1971); Proceedings of the Thirteenth International Conference on Low Temperature Physics, Boulder, Colorado, 1972 (unpublished).

[2] S. B. Trickey, W. P. Kirk, and E. D. Adams, Rev. Mod. Phys. 44, 668 (1972).

[3] L. Goldstein, Phys. Rev. 171, 194 (1968).

[4] P. J. Wojtowicz, J. Appl. Phys. 35, 99 (1964); P. J. Wojtowicz and R. I. Joseph, Phys. Rev. 135, A1314 (1964).

[5] N. W. Dalton and D. W. Wood, Phys. Rev. 138, A779 (1965).

[6] K. Pirnie, P. J. Wood, and J. Eve, Mol. Phys. 11, 551 (1966).

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

[8] Cf. the review paper of C. Domb, Adv. Phys. 9, 149 (1960).

[9] See, for instance, G. S. Rushbrooke and P. J. Wood, Mol. Phys. 1, 257 (1958).

[10] G. A. Baker, Jr., H. E. Gilbert, J. Eve, and G. S. Rushbrooke, Phys. Rev. 164, 800 (1967).

[11] M. F. Panczyk, R. A. Scribner, G. C. Straty, and E. D. Adams, Phys. Rev. Lett. 14, 1102 (1967); M. F. Panczyk and E. D. Adams, Phys. Rev. 187, 321 (1969).

[12] N. W. Dalton, Proc. Phys. Soc. 88, 659 (1966).

[13] E. R. Grilly, J. Low Temp. Phys. 4, 615 (1971).

[14] See, for instance, J. S. Smart, *Effective Field Theories of Magnetism* (Saunders, Philadelphia, 1966), pp. 69–74.

[15] There appears to be a misprint in Ref. 4 for the full expression given there for c_{22}. Namely, the structure constant $p_1(y;zy)$ should there be replaced by $p_1(y;yz)$, which vanishes in cubic structures. Without this correction, the generalized coefficient e_4 of $(1/T^4)$ given in Ref. 5 could not be obtained.

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

[17] J. R. Sites, D. D. Osheroff, R. C. Richardson, and D. M. Lee, Phys. Rev. Lett. 23, 836 (1969).

[18] It is appropriate to mention here that L. I. Zane [Phys. Rev. Lett. 28, 420 (1972); J. Low Temp. Phys. 9, 219 (1972)] has invoked multiple-exchange-type of interactions to account semiquantitatively for the isochores of magnetized solid He^3. In our approach discussed in the present paper, effective first-neighbor-pair and second-neighbor-pair exchange couplings are used. The various multiple exchange interactions are expected to be distributed phenomenologically between the here assumed effective pair interactions.