24% over the initial preshock value ν_0 with a like 24% decrease from ν_0 at the tail. This effect is illustrated by the middle curve, 100 Torr, with a $\nu_0/\omega = 1.1$. The bottom curve for 6 cm Hg again illustrates the case of mostly positive rotation, since initially $\nu_0/\omega = 0.65$.

Although the results in Fig. 2 and 3 show crossover from positive to negative Faraday rotation, it still remains to find the change in slope in $n_o - n_e$ at $\nu/\omega = 1.7$ experimentally. This change in slope may exist in the data of the 200-Torr curve of Fig. 3. However, the qualitative nature of the experiments and the changing nature of the collision frequency and electron density as they were shocked precluded locating that point.

[1]D. L. Jones, in *Exploding Wires*, edited by W. Chace and H. Moore (Plenum, New York, 1968), Vol. 4, p. 96, Fig. 1.

[2]D. L. Jones, Phys. Fluids 11, 247 (1968).

[3]F. C. Fehsenfeld, J. Chem. Phys. 39, 1653 (1963).

[4]B. W. McDaniel, *Collision Phenomena in Ionized Gases* (Wiley, New York, 1964), p. 19.

[5]D. L. Jones and R. M. Gallet, in *Exploding Wires*, edited by W. Chace and H. Moore (Plenum, New York, 1962), Vol. 2, p. 127 ff.

[6]L. D. Landau, in *Collected Works of Landau*, edited by D. ter Haar (Gordon and Breach, New York, 1965), p. 437.

Nuclear-Spin-Ordering Effects in Magnetized Solid ^{3}He†

W. P. Kirk and E. D. Adams
Physics Department, University of Florida, Gainesville, Florida 32601
(Received 24 May 1971)

The thermodynamic pressure $P_V(T, H)$ has been measured in solid ^{3}He in the presence of high magnetic fields. Modification of nuclear-spin ordering by the field is observed, indicating antiferromagnetic behavior. The results indicate that the behavior is not quantitatively as expected from the Heisenberg model. A depression of the melting curve in a field has also been observed.

We have measured the thermodynamic pressure $P_V(T,H)$ in solid ^{3}He for molar volumes $V \simeq 24$ cm^3/mole in applied magnetic fields of 60, 40, and ~0 kG. In addition to the specific interest in magnetized solid ^{3}He,[1-4] this study provides information of use in extending our understanding of fundamental problems in magnetism.

The major results of this study are as follows: (1) An applied field has a pronounced effect on the thermodynamic properties of solid ^{3}He. This is the first time these effects have been seen. In a recent study no effect of the field was observed.[5] (2) Time constants are short enough to permit solid ^{3}He to be magnetized in high fields. (3) The exchange energy J is negative, corresponding to antiferromagnetism. (4) The behavior at high magnetic fields is not adequately described by the series expansions[6] for the Heisenberg Hamiltonian.

In solid ^{3}He the magnetic properties of the nuclear spin system are determined by the exchange interaction which arises from the large zero-point motion and the resulting overlap of wave functions of neighboring atoms. An interesting consequence of this is that there should be a transition to a magnetically ordered state at $T_c \simeq 3|J|/k$, where J is the exchange energy. Measurements of $P_V(T)$ in zero magnetic field have been used to determine $|J|$.[7] For the solid near melting with $V \simeq 24$ cm^3/mole, it was found that $|J|/k \simeq 0.7$ mK or $T_c \simeq 2$ mK.[8] Recently, susceptibility measurements have shown that J is negative, corresponding to antiferromagnetic ordering.[9,10]

Solid ^{3}He has been considered to be probably the best example of a Heisenberg antiferromagnet because of the absence of interactions other than exchange and the symmetry provided by the bcc lattice. In order to make a detailed comparison between theory[1,6] and experiment, the measurements in zero magnetic field need to extend well into the critical region near $T_c = 2$ mK. However, as Goldstein[1] has pointed out, a detailed comparison should be possible in the paramagnetic region at temperature well above T_c if one applies a large magnetic field.

Taking the Heisenberg Hamiltonian to represent the exchange interaction, the Hamiltonian for the system is[11]

$$\mathcal{H} = -2J \sum_{i<j}^{N} \vec{I}_i \cdot \vec{I}_j - H \sum_{i}^{N} \mu_{zi}, \qquad (1)$$

where $\vec{I}$ is the nuclear spin operator, H the mag-

netic field (applied in the z direction), and $\vec{\mu}$ the magnetic moment. Various physical quantities are given in terms of the partition function $Z = \mathrm{Tr}\exp(-\mathcal{H}/kT)$. High-temperature ($T > T_c$) series expansions of Z for the spin-$\frac{1}{2}$ case have been made by Baker *et al.*[6] The expansion gives for the thermodynamic pressure, $P_V(T,H) = kT(\partial \ln Z/\partial V)_T$,

$$P_V(T,H) = \frac{R}{V}\frac{J}{k}\frac{\partial \ln|J|}{\partial \ln V}[3x - \tfrac{3}{2}x^2 + \cdots + y^2(2 + 12x + 52x^2 + \cdots) + y^4(-1.33 - 23x + \cdots) + \cdots], \quad (2)$$

where $x = J/kT$ and $y = \mu H/kT$, and V is the molar volume.

Previous measurements[7] in zero field, which extended to $T \simeq 7T_c$, required only the linear term in x and therefore depended only on $|J|$. In the presence of a field, the term in y^2 reveals the sign of J. In addition, the degree to which this expansion fits the data provides a test of the correctness of the Heisenberg model, or of the expansions resulting from it.

Our results, in the form $P_V(T,H)$ vs T^{-1}, are shown in Figs. 1 and 2. Only pressure differences are shown (or are significant), with the zero of ΔP chosen such that each curve extrapolates through zero at $T^{-1} = 0$. Open symbols represent data taken on warming; closed ones, while cooling. The scatter is indicative of the long-term stability of the gauge. Examination of Eq. (2) shows that the downward curvature in a field indicates antiferromagnetic behavior or $J < 0$. This is in agreement with susceptibility measurements.[9,10] Although it was found that $J < 0$ in three separate susceptibility studies, we consider this confirmation to be significant since it is the first non-NMR determination of the sign of J. The result $J < 0$ is independent of the Heisenberg model as will be demonstrated thermodynamically below.

The solid lines shown on each figure are the calculations of $P_V(T,H)$ from Eq. (2) using the J's obtained in zero field and with $\partial \ln|J|/\partial \ln V = 17.5$.[7] Only terms written out explicitly in Eq. (2) were used; higher-order terms contribute $< 1\%$. The dashed lines near the ends of these are for fields differing by ± 3 kG from that for the solid curves. The magnitude of the effect of the field is much less than would be expected from Eq. (2). This is particularly apparent at 60 kG, for which Eq. (2) predicts a maximum in P at $T \approx 40$ mK for $J/k = -0.604$ mK. At the lowest temperature, $T \approx 20$ mK, the pressure was still increasing. This can

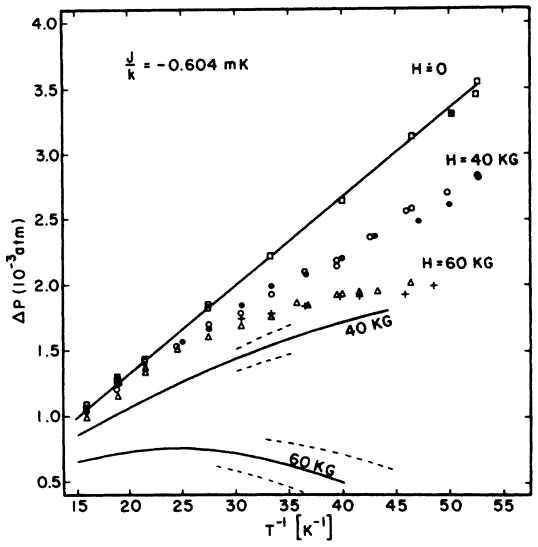

FIG. 1. Pressure differences versus T^{-1} for $V = 23.88$ cm^3/mole in fields of 60, 40, and 0.5 ($H \approx 0$) kG. Various symbols for a given H are for different traversals of the temperature region. The solid curves are calculated behavior based on the Heisenberg model.

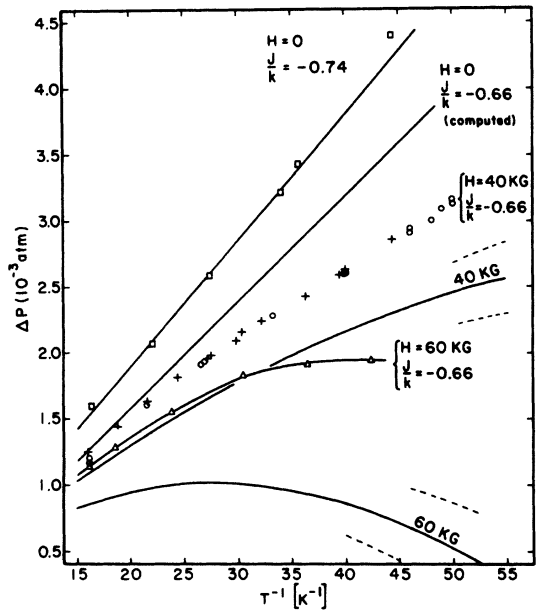

FIG. 2. Pressure differences versus T^{-1} for $V = 24.15$ cm^3/mole in zero field, and for $V = 24.0$ cm^3/mole in the fields of 40 and 60 kG. Various symbols for a given H are for different traversals of the temperature region. The curve through the 60-kG data is simply to connect the points. Other curves are calculated behavior based on the Heisenberg model.

be stated with much greater certainty than is apparent from the scatter in the data. Always on warming by as little as 1 mK at the lowest temperature, a definite decrease in pressure was observed. Thus, the exchange interaction still dominates the behavior in high fields.

The reason for the discrepancy between the Heisenberg model and experiment is not clear, although a number of possibilities can be readily suggested. These include the validity of the model, inclusion of nearest neighbors only, and convergence or correctness of the series expansion.

We can examine the data thermodynamically using the Maxwell relation $(\partial P/\partial H)_{V,T} = (\partial M/\partial V)_{H,T}$, where M is the magnetization. Using this with (1) the fact that $\partial \ln|J|/\partial \ln V > 0$ and (2) the present result that $(\partial P/\partial H)_{V,T} < 0$, one can conclude that $J < 0$, independent of any model. To obtain $P_V(T,H)$ from the Maxwell relation requires $M(V)$. Since the *low-field limit* $M = CH/T(1 - 4J/kT)$ is inadequate at high fields, further analysis of the data in terms of the Maxwell relation will be left to a subsequent paper.

We have also measured the melting pressure $P_m(T,H)$ in fields of 1 and 60 kG. This was done by measuring the pressure change relative to the minimum in the melting curve, $P_m(T,H) - P_{\min}$. A depression of the melting curve in 60 kG relative to that in 1 kG was seen. At 20 mK the magnitude of this effect was 0.030 ± 0.006 atm, consistent with the calculation of Goldstein.[1] (The relatively large uncertainty is due to two sources: We are subtracting two numbers ≈ 4 atm which are almost equal. Because of the large melting curve slope $|dP_m/dT| \approx 42$ atm/K, a shift in temperature of only 0.1 mK gives a pressure change of 0.004 atm.)

The pressure was measured using a capacitance strain gauge[12] with a welded bundle of fine copper wires attached to the chamber walls to provide a large surface area.[13] This bundle, consisting of 26 000 No. 50 wires, gave a surface area of 200 cm^2 for contact to $\simeq 0.3$ cm^3 of sample. The bundle interfered in no way with the operation of the gauge. There was no indication of hysteresis and the pressure resolution was 2×10^{-6} atm.

Temperatures were produced with a ^{3}He-^{4}He dilution refrigerator. A Speer carbon resistor served as the secondary thermometer. To avoid problems with magnetoresistance in the high fields, the resistance thermometer was located outside the main field in a region where the fringe field was 2.5 kG for 60 kG on the sample. The resistor was shielded entirely from the fringe field by placing it within a Nb_3Sn cylinder capable of excluding fields of 25 kG. Thermal contact between the resistor and the sample was made by a bundle of copper wires soldered to the chamber by using tin. Magnetic fields, $H \geq 500$ G, were applied at the sample to make the tin normal for all measurements.

Two other Speer resistors were mounted on the sample chamber itself. These showed a strong negative magnetoresistance $\Delta R/R_0 \propto HT^{-1}$ for fields up to 20 kG. At higher temperatures and low fields the error in T would be relatively small if the magnetoresistance is ignored. However, at $H = 40$ kG and $T = 25$ mK the error in T would be $\sim 50\%$. Also some use was made of a glass-ceramic capacitance thermometer.[14] This was located in the field and was relatively little affected by it. Full details will be reported elsewhere.

The carbon resistor was calibrated using the melting pressure $P_m(T)$ of ^{3}He as a primary thermometer.[15] The pressure was related to temperature using the data of Scribner and co-workers[15] for $T \gtrsim 40$ mK and those of Johnson *et al.*[16] for $T \lesssim 40$ mK. In order to make $P_m(T)$ and dP_m/dT for the two sets of data join smoothly, the results of Johnson *et al.* were reduced by 0.08 atm and those of Scribner and co-workers were reduced by ~ 0.02 atm near 40 mK. The approximate validity of this temperature scale is indicated by the linearity of the curves for $H \approx 0$ in Figs. 1 and 2.

Magnetic fields were produced by a superconducting solenoid operating in the persistent mode and were determined from the current using data accurate to 1%, furnished by the manufacturer. This should determine fields to 3% or better when hysteresis loops are avoided as was done.

Data were taken in two different ways. Usually the procedure was first to cool the sample to the lowest temperature and hold it there for a few hours. Then data were taken with the temperature regulated electronically, spending about 5 to 10 min at each point. To check for long time constants or nonequilibrium effects, data were taken in some runs, at both 40 and 60 kG, with the temperature decreasing as rapidly as could be achieved (~ 1 mK/min at $T = 40$ mK). In no case was there evidence of a time lag between the ^{3}He magnetization [as indicated by $P_V(T,H)$] and the temperature. It is difficult to translate this into a time constant, but we estimate an upper limit $\tau < 1$ min for the spin-lattice relaxation time. Thus it should be possible to magnetize ^{3}He in high fields

at temperatures in the range of a few millikelvins.

Samples formed by the blocked capillary technique were annealed as they slowly cooled through the region just below the melting temperature. The ^{4}He impurity content was 400 ppm. However, *in situ* purification occurred through the phase separation at ~100 mK. After phase separation, the samples were kept below 65 mK where the ^{4}He remaining in the ^{3}He-rich phase is only ~10 ppm. Thus the measurements should be for "pure" ^{3}He.

We believe that the major explanation of the null result found by Osgood and Garber[5] (OG) can be related to the large negative magnetoresistance of the carbon resistance thermometers, assuming that their Speer resistor behaves similarly to ours. OG compare a resistor in the magnet with another "outside of the magnet" throughout the temperature range (and with the melting curve only above 100 mK) and "estimate" a magnetoresistance correction to the temperature of less than 5% in 57 kG. However, as noted, we have checked the magnetoresistance in fields up to 60 kG and find that $\Delta R/R_0 \propto HT^{-1}$ for $H \leq 20$ kG and $T \geq 20$ mK. This would cause a rapidly increasing error in T^{-1} as the temperature is lowered, resulting in a decidedly downward curvature in the data of OG. We suggest that possibly the failure of OG to observe large magnetoresistance at the lower temperatures was because the resistor "outside of the magnet" was not adequately shielded from the fringing field of the magnet; hence it suffered a magnetoresistance effect itself. Also, the "outside" resistor may have been in poor thermal equilibrium with the resistor inside the magnet. Another way of showing that the magnetoresistance is the source of the trouble is to use our unshielded resistors as thermometers without accounting for the magnetoresistance. In this case, our 60 kG results would be almost the same as for $H = 0$, and the low-T 40-kG data would fall slightly to the left of that for $H = 0$.

It was argued by OG that errors in thermometry could not account for the discrepancy between their results and calculations since at 57.2 kG pressure changes were observed which were larger than the maximum calculated [from our Eq. (2)]. This is understood in light of our findings that the pressure changes are in fact considerably larger than those calculated by the Heisenberg model. Indeed, if we assume that their lowest temperature was in each case ~13 mK ($T^{-1} \sim 75$), then their ΔP for 57.2 kG is consistent with our 60 kG results. In making this comparison, the instability and lower resolution of their capacitance strain gauge has to be considered.

We acknowledge useful discussions with Louis Goldstein and the enthusiastic assistance of Mike Ryschkewitsch with parts of the apparatus. We also thank Al Menard for assistance in the initial phase of this work.

---

†Work supported in part by the National Science Foundation.

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

[2]L. Goldstein, Phys. Rev. Lett. 25, 1095 (1970).

[3]W. P. Wolf, M. F. Thorpe, and R. Alben, Phys. Rev. Lett. 26, 756 (1971).

[4]R. E. Walstedt, L. R. Walker, and C. M. Varma, Phys. Rev. Lett. 26, 691 (1971).

[5]E. B. Osgood and M. Garber, Phys. Rev. Lett. 26, 353 (1971).

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

[7]M. F. Panczyk and E. D. Adams, Phys. Rev. 187, 321 (1969).

[8]See H. Meyer, J. Appl. Phys. 39, 390 (1968), for a review of relaxation-time studies of exchange.

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

[10]J. R. Sites, D. D. Osheroff, R. C. Richardson, and D. M. Lee, Phys. Rev. Lett. 23, 835 (1969); P. B. Pipes and W. M. Fairbank, Phys. Rev. Lett. 23, 520 (1969).

[11]Other terms in the Hamiltonian are unimportant in the temperature range of this study.

[12]G. C. Straty and E. D. Adams, Rev. Sci. Instrum. 40, 1393 (1969).

[13]W. P. Kirk, S. H. Castles, and E. D. Adams, to be published.

[14]W. N. Lawless, Rev. Sci. Instrum. 42, 561 (1971). We are grateful to Dr. Lawless and Corning Glass Works for making one of these thermometers available to us.

[15]R. A. Scribner, M. F. Panczyk, and E. D. Adams, J. Low Temp. Phys. 1, 313 (1969); R. A. Scribner and E. D. Adams, Rev. Sci. Instrum. 40, 287 (1970).

[16]R. T. Johnson, O. V. Lounasmaa, R. Rosenbaum, O. G. Symko, and J. C. Wheatley, J. Low Temp. Phys. 2, 403 (1970).